Solar wind noble gases in targets from the genesis mission

Author[s]:
Grimberg, Ansgar

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SOLAR WIND NOBLE GASES IN TARGETS FROM THE GENESIS MISSION

A dissertation submitted to the

ETH ZÜRICH

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presented by

ANSGAR GRIMBERG
Diplom-Geologe, Universität Bonn (Germany)

born 22.02.1974
citizen of Germany

accepted on the recommendation of:

Prof. Dr. Rainer Wieler, ETH Zürich, examiner
Prof. Dr. Peter Bochsler, Universität Bern, co-examiner
Prof. Dr. Bernard Marty, CRPG Nancy, co-examiner
Prof. Dr. Bernard Bourdon, ETH Zürich, co-examiner

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Frontcover

*upper half:* SOHO-EIT image in resonance lines of eight and nine times ionized iron (Fe IX/X) at 171 Angstroms in the extreme ultraviolet showing the solar corona at a temperature of about 1 million K. This image was recorded on 11 September 1997. It is dominated by two large active region systems, composed of numerous magnetic loops. *lower half:* This artist’s conception shows the Genesis spacecraft in collection mode. The cover of the science canister contains one set of collection materials. It is the same kind of hexagonal silicon wafer array that comprises the stack of four arrays that were rotated out of the interior of the canister when the spacecraft began to orbit L1. The two solar panels, shown in blue in this depiction, which extend to the side of the spacecraft bus, provide electrical energy for the functions performed by the rest of the spacecraft.

Backcover

(from top down) *first:* G-switch sensor that was installed in inverted orientation and thus was unable to sense sample return capsule deceleration during atmospheric entry and initiate parachute deployments. *second:* Genesis capsule after crash-landing in the Utah desert. *third:* Bulk metallic glass target in the middle of the scrap of the Genesis capsule. *fourth:* SEM image of the Bulk metallic glass after etching, showing the negative relief of a dendritic crystalline phase that got etched preferentially compared to the glass matrix.
CONTENTS

ABSTRACT

ZUSAMMENFASSUNG

I. INTRODUCTORY CHAPTER

1. FOREWORD AND OUTLINE OF THESIS 14
2. INTRODUCTION 14
   2.1. The Sun – Memory of the Solar System’s Origin 14
   2.2. Nucleosynthesis and its Effect on the Solar Composition 15
   2.3. The Solar Wind – Deliverer of Solar Matter 16
   2.4. Solar Wind Formation and Regimes 17
   2.5. From the Sun to the Solar Wind – Processes Controlling Compositional Variations
      2.5.1. Incomplete Hydrogen Burning 21
      2.5.2. Gravitational Settling 21
      2.5.3. First Ionisation Potential and Time 22
      2.5.4. Coulomb Drag 23
   2.6. Solar Wind Sample Return Missions 25
      2.6.1. Solar Wind Composition Experiment 26
      2.6.2. The Genesis Mission 27

II. SAMPLE TREATMENT AND NOBLE GAS ANALYSIS

1. BULK METALLIC GLASS – BMG 36
2. SAMPLE FRAGMENTATION 37
3. CLEANING 37
4. NOBLE GAS MEASUREMENT 40
   4.1. Closed System Stepwise Etching – CSSE 40
   4.2. Pyrolytical Gas Extraction 42
   4.3. Gas Cleaning and Separation 42
   4.4. Mass Spectrometry 42
   4.5. Interferences 42
   4.6. Blanks 43
III. Irradiation Experiments

1. INTRODUCTION
2. TRAPPING EFFICIENCY
3. EXPERIMENTAL TECHNIQUE
   3.1. CASYMS
   3.2. Samples
   3.3. Analysis of Implanted Ions
   3.4. SRIM Calculations
4. RESULTS
   4.1. Trapping Efficiency
   4.2. Depth Distribution of Different Isotopes
5. CONCLUSION

IV. Solar Wind Neon and the Lunar Noble Gas Record

Solar Wind Neon from Genesis: Implications for the Lunar Noble Gas Record

V. Composition of Light Solar Wind Noble Gases

Composition of Light Solar Wind Noble Gases in the Bulk Metallic Glass flown on the Genesis Mission

1. INTRODUCTION
2. BULK SOLAR WIND HELIUM AND NEON DATA
3. DEPTH DEPENDENCE OF TRAPPED SOLAR Ne COMPOSITION IN THE BMG
4. ISOTOPIC FRACTIONATION OF THE SOLAR WIND
5. CONCLUSION

VI. Helium, Neon, and Argon Isotopic and Elemental Composition

Solar Wind Helium, Neon, and Argon Isotopic and Elemental Composition:
Data from the Metallic Glass flown on NASA’s Genesis Mission

1. INTRODUCTION
2. SAMPLE
3. EXPERIMENTAL PROCEDURE
   3.1. Pyrolytic Analysis
   3.2. Closed System Etching Technique
   3.3. Blanks
3.4. Uncertainties 90
3.5. Artificial Irradiations 91
3.6. SRIM Simulation of Depth Distributions of SW Atoms 92

4. RESULTS 92
4.1. Isotopic Composition 92
4.1.1. Neon 92
4.1.2. Helium 95
4.1.3. Argon 99
4.2. Elemental Composition 101

5. DISCUSSION 103
5.1. Mass Separation upon Implantation 103
5.2. Bulk Solar Wind Composition 105
5.3. Fractionation of Low Speed Solar Wind 106

6. CONCLUSIONS 108

APPENDIX: Elemental and Isotopic Fractionation by Inefficient Coulomb Drag in the Solar Wind

VII. CONCLUSIONS AND OUTLOOK
1. CONCLUSIONS 122
2. OUTLOOK 124

ACKNOWLEDGEMENTS 125

CURRICULUM VITAE
ABSTRACT

During solar system formation ~99.8 % of the mass of solar nebula material was incorporated into the Sun. Material in the Sun’s outer convection zone and photosphere is believed to have survived the entire solar systems lifespan of ~4.6 Gyr without larger compositional changes. Material from the photosphere is constantly carried to interplanetary space by the solar wind that originates in the solar atmosphere. Hence, the solar wind provides an important source of information about the isotopic and elemental composition of the Sun and thus is fundamental for the understanding of cosmochemistry. NASA’s Genesis mission sampled the solar wind for about 2 ½ years in space. The general science objectives are to study the solar wind composition and possible fractionation mechanisms influencing it in order to derive precise information of the photospheric composition. In this framework noble gases are an important diagnostic to examine different processes influencing the solar wind composition because of their high abundance, their almost inert chemical character and the fact that their analysis is relatively unproblematic compared to most other elements.

The aim of this study was to measure light noble gases from the solar wind that were implanted into a bulk metallic glass (BMG) target for 853 days on the Genesis mission. The target itself was especially designed for our main purpose to determine the depth distributions of solar He, Ne, and Ar trapped into the target via gas extraction by closed system stepwise etching (CSSE). From the depth distribution of implanted ions, in turn, it is possible to derive information about the variation of composition on particle speed. These measurements were complemented by total extraction analyses of the trapped noble gases from BMG samples to obtain data on the bulk composition of the solar wind for the relatively long exposure period.

Implantation of ions into solid matter, the basic principle of Genesis’s solar wind collection, is a process that is not sufficiently well understood for low energies that are typical for the solar wind. Therefore, a preliminary study focussed on the trapping properties of He and Ne into different target materials, e.g. isotopic fractionation and the distribution of isotopes with depth, but also on target material properties like the depth resolution in the BMG using the etching method. The associated irradiations have been carried out with the calibration system for mass spectrometers at the University of Bern. The results were then compared to simulations calculated with the open-access SRIM computer code. The artificial irradiation experiments revealed substantial problems of SRIM for the prediction of He backscattering from different target materials and also led to the conclusion that SRIM overestimates the isotopic separation of He within the BMG upon implantation.
The bulk isotopic composition for He, Ne, and Ar measured in the BMG by total extraction is in good agreement with the mean values observed in foils from the Solar Wind Composition (SWC) experiment carried out during the lunar landings of Apollo 11 to Apollo 16. Also the elemental composition deduced from the BMG fits with the data from the single SWC experiments. However, the fluence of He derived from the BMG is several percent lower compared to values obtained in-situ in space or reported from other Genesis bulk targets. This discrepancy in the fluence hampers the interpretation on the elemental abundance of He relative to Ne and Ar.

The main expectations on the BMG to obtain depth profiles of the distribution for all three elements with CSSE were totally fulfilled. The isotopic composition of He, Ne, and Ar varies with depth within the BMG in a way generally consistent with SRIM simulations of a solar wind of uniform composition and adopting the measured solar wind velocity distribution. The similarity of the release patterns with the depth dependent distribution of trapped solar He, Ne, and Ar reported for lunar and asteroidal regolith samples shows that also the solar noble-gas record of extraterrestrial samples can be explained by isotope separation of implanted solar wind with depth. This result confirms that contributions from high-energy particles to the solar wind fluence are very minor. This fact is consistent with in-situ flux observations and solves a long-standing problem of an unreasonable high apparent flux of solar energetic particles thought to be represented by an enigmatic “SEP” noble gas component in lunar regolith samples and solar-gas rich meteorites.

An unexpected observation is represented by a small fraction of the total solar gas in the BMG released from shallow depths. It is markedly enriched in the light isotopes relative to deeper sited gas and to results from implantation simulations. Atmospheric contamination or fractionation due to diffusion cannot explain this enrichment. Neither do contributions from a neutral interstellar or solar component, because the resulting concentrations would be too small to explain this gas fraction. Fractionation in the corona by inefficient Coulomb drag, however, is a feasible mechanism that suffices to explain the isotopic composition of the superficially implanted gas by attributing it to very low-speed, current sheet related SW. In this model Ne is accelerated more efficiently relative to Ar, which would also account for the relatively high Ne/Ar elemental ratios in the same gas fraction from shallow sites. The higher depth range of implantation simulated for current-sheet related SW, however, somewhat conflicts with the small penetration depth observed in the BMG.
ZUSAMMENFASSUNG


Der Prozess der Implantation von Ionen in festes Material (das Grundprinzip der Beprobung des Sonnenwinds durch die Genesis-Mission) ist für tiefe Energien, wie sie für den Sonnenwind typisch sind, nicht genügend verstanden. Aus diesem Grunde wurde eine Vorstudie durchgeführt, mit dem Ziel die Einfangeigenschaften wie Isotopenfraktionierung oder Tiefenverteilung für He und Ne in verschiedenen Kollektormaterialien zu erforschen und zudem Materialeigenschaften

Die Gesamtzusammensetzung der He, Ne und Ar Isotope, die mittels Totalextraktion gemessen wurde, stimmt gut mit den Mittelwerten aus den Folien der „Solar Wind Composition“ (SWC) Experimente überein, welche während der Mondlandungen von Apollo 11 bis Apollo 16 durchgeführt wurden. Gleiches gilt für die Elementzusammensetzung im BMG, welche ebenfalls den Daten der einzelnen SWC Experimente entspricht. Jedoch erschwert eine um mehrere Prozent niedrigere Heliumkonzentration, als sie durch in-situ Messungen im All bestimmt oder von anderen sog. Bulk-SW Kollektoren berichtet wurde, die Interpretation der He-Konzentration gegenüber Ne und Ar.


Etwas unerwartete Ergebnisse lieferte solares Gas, welches aus sehr geringer Tiefe des BMGs freigesetzt wurde. Verglichen zu tiefer sitzendem Gas und den Resultaten der Simulationen ist der oberflächliche Gasanteil merklich angereichert an leichten Isotopen, was nicht durch
atmosphärische Kontamination erklärbar ist. Ebenso wenig kann dies die Folge eines Beitrags einer neutralen interstellaren oder solaren Komponente sein, da ihre Konzentrationen viel zu gering sind, um die freigesetzte Gasmenge zu erklären. Eine Fraktionierung von sehr langsamen Sonnenwind in der Corona (Current-Sheet Sonnenwind) durch ineffiziente Übertragung von Coulomb Kräften (Coulomb drag Modell) ist hingegen ein möglicher Prozess welcher die Isotopenzusammensetzung der oberflächlich implantierten Edelgase erklären könnte. Zudem wird bei diesem Modell Ne deutlich effizienter als Ar beschleunigt, was wiederum die relativ hohen Ne/Ar Elementverhältnisse in eben dieser Gasfraktion berücksichtigt. Hingegen steht die mit SRIM ermittelte größere Reichweite implantieter Ionen des Current-Sheet Sonnenwinds in gewissem Widerstand zu der geringen Eindringtiefe der Gasfraktion im BMG.
I. Introductory Chapter
1. FOREWORD AND OUTLINE OF THESIS

This PhD thesis focuses on solar noble gases collected with a metallic glass target onboard NASA’s Genesis mission from 2001 to 2004. It is a continuation of studies by V.S. Heber (2002) as part of her PhD thesis, who choose the target actually flown by testing several materials for their suitability as noble gas collectors.

The thesis in hand has been conducted in the noble gas laboratories at ETH under the supervision of Rainer Wieler and Heinrich Baur, and in close collaboration with Peter Bochsler and Fritz Bühler from the University of Bern, Don Burnett from the California Institute of Technology in his position as the principal investigator of the Genesis mission, and the entire Genesis science team. The introductory chapter will give the motivation of this study and the Genesis mission itself, briefly summarise the main cosmochemical and solar physical concepts necessary to understand why we study the sun and use the solar wind, and set the Genesis mission in a historical frame of sample return missions in space. The following two chapters address the methods I used in this study and describe calibration experiments that we carried out before the return of the Genesis satellite. Chapter IV and V are published articles and chapter VI is a manuscript submitted to *Geochimica et Cosmochimica Acta*. The last chapter summarises all results and points on questions that remained open or came up as a result of this work. The references cited are always given at the end of each chapter.

2. INTRODUCTION

2.1 The Sun – Memory of the Solar System’s Origin

The solar system originates from a rotating and flattened gas and dust disk, the solar nebula, that formed from a collapsing molecular cloud in our Galaxy ~4.6 Gyr ago (Boss, 2003). Much of the motivation for cosmochemical studies of meteorites, comets, and other primitive bodies stems from the desire to constrain the physical and chemical conditions in the solar nebula from which the sun and the planets formed. A cornerstone in this view was a seminal paper published by H.E. Suess and H.C. Urey (1956) on the abundances of the elements in the solar system based on meteorite data. The current theories of solar system evolution and planetary formation attribute the differences in the elemental and isotopic composition of different bodies in the solar system to a set of processes that over time resulted in spatial inhomogeneities in the solar nebula, from which the various planets formed. To decipher and understand these processes,
the initial conditions of the solar nebula have to be known to high precision. The most primitive material in the solar system, which is available on earth, and therefore the material closest to the solar nebula is, according to common understanding, represented by primitive meteorites as the carbonaceous chondrite Orgueil. Their elemental abundances agree very well with that of the Sun except for the volatile elements H, C, N, and O, the noble gases, and Li (Anders and Grevesse, 1989). The depletion of volatile elements is a common observation for all solid matter in the solar system. Fortunately, when the solar system formed, about 99.8 % of solar nebular material was incorporated into the Sun. And though nuclear reactions have modified the composition in its core, it is generally accepted that the Sun preserved the original, largely unfractionated composition of the solar nebula (Bochsler, 2000). Therefore, knowledge of the solar abundances of elements and their isotopic composition is the key to decipher the solar nebula composition and in turn to have a reference for the different processes that led to the inhomogeneities observed in planetary materials.

2.2 Nucleosynthesis and its Effect on the Solar Composition

The Sun consists to 73.5 % by mass of hydrogen which is constantly burned in the core to helium, with 24.9 % by mass the second most abundant element, by the pp chain (Meyer and Zinner, 2006). The basic reaction is proton-fusion via deuterium (ppI chain),

\[ ^1\text{H} + ^1\text{H} \leftrightarrow ^2\text{H} + \gamma \]
\[ ^2\text{H} + ^1\text{H} \rightarrow ^3\text{He} + \gamma \]
\[ ^3\text{He} + ^3\text{He} \rightarrow ^4\text{He} + 2^1\text{H}, \]

in which the p + p reaction produces a deuteron. Deuterium subsequently captures a proton to make \(^3\)He, which, in the principal pp chain (ppI), captures another \(^3\)He to make \(^4\)He and two protons. The other two pp chains (II and III) also produce \(^4\)He simultaneously with ppI by the capture and subsequent radioactive decay of \(^7\)Be-\(^7\)Li and \(^7\)Li-\(^8\)Be-\(^8\)B. Further nucleosynthetic reactions cannot occur in the Sun at present, because of the maximum temperature of only \(1.3 \times 10^7\) K that is reached in the core. Therefore, over its present lifespan the Sun completely burned deuterium and changed in composition only for H, HD and the Helium isotopes. All other elements are essentially present in their initial composition.

To complete the pp chain requires temperatures >10^7 K, a fact which restricts the production of He mainly to the core, with minor exceptions as will be discussed in Section 2.5. The core of the sun, however, is surrounded by the radiation zone (Fig. 1.1) where the material is hot
and dense enough that energy is transferred via thermal radiation and no thermal convection of material occurs. Thus the radiation zone essentially prevents mixing of core material, especially nucleosynthetic He, with the convection zone overlying the radiation zone. At present, the Sun is about halfway through its main-sequence evolution. According to standard evolution models for the main-sequence phase of stars, the composition of the Sun’s convective zone remained largely unchanged over its present lifespan (Bochsler, 2000), at least its outermost part which is therefore treated separately and termed outer convective zone (OCZ). Hence, the initial abundances of elements of the solar system and thus the solar nebula are represented by the composition of the OCZ and its visible surface (Wiens et al., 2004), the photosphere, with minor exceptions as will also be discussed in Section 2.5.

2.3. The Solar Wind – Deliverer of Solar Matter

The composition of the solar photosphere and the OCZ can be studied by two different approaches. Observations using helioseismology or spectroscopy (e.g. Hall, 1973; 1975) succeeded in determining elemental and a few isotopic ratios, but the photospheric values inferred this way often have large uncertainties due to complex model assumptions (Von Steiger

Fig. 1.1. Illustration of the interior of the Sun with the main processes affecting the composition of either the outer convective zone or that of the solar wind.
et al., 2001). The other approach to study the photospheric composition is the analysis of the solar wind (SW). This stream of charged particles originates in the chromosphere and the corona, which are directly coupled to the photosphere, and mainly consists of electrons, protons, and $\alpha$-particles, but also all heavier elements in minor amounts. The SW can be studied either in-situ with spacecrafts instruments or by analyses of SW particles implanted into lunar or asteroidal soils or collection substrates. In-situ instruments, e.g. onboard the advanced composition explorer (ACE) (Gloeckler et al., 1998; Mason et al., 1998; Smith et al., 1998; McComas et al., 2003) or the Solar and Heliospheric Observatory (SOHO/CELIAS), successfully determined the isotopic and elemental SW composition for a wide range of elements. These measurements, however, still suffer from lack of precision to infer very precise photospheric values, a precision which is also necessary to correct for fractionation processes in the SW that have to be accounted for. These processes will be discussed in Section 2.5. Trapped SW particles analysed in lunar or asteroidal soils have the advantage to represent a long-term history of the SW composition (Pepin et al., 1970; Becker and Pepin, 1994; Wieler, 1998; Hashizume et al., 2000; Heber et al., 2003; Hashizume and Chaussidon, 2005). But, this record is often modified by processes affecting the sub-micron thin, superficial layer that hosts the trapped SW particles, e.g. particle losses or fractionation due to surface sputtering, diffusion or erosion. Additionally, the determination of the solar particle composition is complicated by an admixture of cosmogenic atoms produced via spallation.

In contrary to the different methods mentioned above, collection substrates exposed to the SW on space missions and returned to laboratories, however, allow in general accounting for all these problems to permit the high precision required (Burnett et al., 2003; Geiss et al., 2004; Wiens et al., 2007).

2.4 Solar Wind Formation and Regimes

The source of particles for the SW is the solar corona. Atoms in the underlying chromosphere get ionised by the corona’s UV radiation and subsequently expanded into the corona itself (Fig. 1.2). The corona is heated up by, e.g., Alfén or magneto-hydrodynamic waves and/or magnetic reconnection on the solar surface to maximum temperatures of $1-2 \times 10^6$ K. These temperatures are sufficient that protons and electrons continuously escape from the gravitational field of the Sun just by their thermal speeds (Bochsler, 2000).

The incorporation of heavier particles than protons into this particle flow, however, requires additional processes like Coulomb collisions (Geiss et al., 1970b) or wave-particle interaction
As soon as the particles are accelerated, they flow along magnetic field lines to interplanetary space typically with velocities between 300 to 900 km/s. Variations in the arrangement and geometry of magnetic field lines on the solar surface result in the formation of regions with specific SW acceleration and thus different SW regimes: high-speed streams from coronal holes, low-speed interstream wind, and the transient wind associated with coronal mass ejections (Neugebauer, 1991; Von Steiger et al., 1997; Fisk et al., 1998). The low-speed SW is thought to be associated to equatorial coronal streamers, large magnetic loops which are connected to active regions and/or prominences in the corona. According to von Steiger (1998), the low-speed SW originates from the streamer’s roots in the corona, and the plasma flows from the roots along field lines at the streamers periphery out into interplanetary space. The low-speed SW measured for the period of Genesis had a broad speed interval with an average of 400 km/s at ~1 AU and made up 45.7 % of the proton fluence.

The source region of high-speed streams are coronal holes (Von Steiger, 1998) which are low-density regions of the corona from which single-polarity magnetic fields open out to interplanetary space (Neugebauer, 1991). During solar minimum of the solar cycle, coronal holes are usually located at the poles of the Sun, while at solar maximum they can open up at any latitudes. The high-speed SW made up 32.3 % of the entire proton fluence during the Genesis exposure period at an average speed of ~590 km/s, also with a broad speed interval at 1 AU. These two SW types do not mix as they are separated by a sharp boundary, even at larger distance in interplanetary space (Von Steiger, 1998). However, it is believed that the two quasi-stationary SW types sometimes are disturbed by coronal mass ejections (CMEs),

Fig. 1.2. Schematic illustration showing the location of the ionisation and acceleration processes for particles (labelled as X) that form the solar wind.
which are transient events of large-scale magnetic structures. The \textit{CME-driven SW} or \textit{transient solar wind disturbance} (Gosling and Forsyth, 2001) is the third regime that was identified by the algorithm used for the Genesis in-situ instruments and differentially sampled. CMEs arise from closed field regions in the corona and thus originate above magnetic neutral lines in the solar photosphere (Gosling and Forsyth, 2001). The coronal field lines open up and the plasma is then dynamically ejected to interplanetary space (Gosling, 1997), a process that is up to date not well understood (Cane and Lario, 2006). For the exposure time of Genesis CME related SW made up a surprisingly large fraction of 22.1 \% and showed the broadest speed interval with an average velocity of about 380 km/s at 1 AU.

A differentiated analysis of these SW regimes allows studying the acceleration mechanisms at the source of the solar wind and their variability with time. At the same time, it is important to scale their influence as these processes finally lead to elemental and isotopic fractionation of SW particles and control the composition as it is observed in interplanetary space, e.g. 1 AU.

Besides the SW also particles with higher energy are emanated from the Sun to interplanetary space. These \textit{solar energetic particles (SEPs)} usually make up less than 1 \% of the entire solar corpuscular radiation (Fig 1.3). Their energy ranges between 5 keV/amu and several tens of keV/amu (Reisenfeld, Wiens and Mewaldt). The fluence of solar energetic particles, which have energies $\sim >5 \times 10^{-3}$ MeV/nucleon, compared to SW particles with energies between 0.35 and 5 keV/nucleon, usually make up less than 1 \% of the entire solar corpuscular radiation. (Data kindly provided by R.A. Mewaldt, G.M. Mason, C.M.S. Cohen, R.A. Leske and, M.E. Wiedenbeck).
hundreds of MeV/amu depending on the acceleration process. Two different types of distinct SEP acceleration process have been identified. The seed material of gradual SEP events is thought to be suprathermal ions in the solar wind or corona that are accelerated by large shocks driven by fast coronal mass ejections (Leske et al., 2003). Impulsive SEPs are samples of hot (10^7 K) solar flare matter accelerated by small impulsive flares (Stone, 2007). Both types of SEPs exhibit variations in their composition. Impulsive SEPs exhibit large enhancements in heavy ions and especially ³He, in cases with resulting ³He/⁴He ratios of about ~1 (Stone, 2007), arising from the impulsive acceleration process. Gradual SEPs show variations in the elemental and isotopic abundances that result from differences in the sources of suprathermal ions and from fractionation during shock acceleration and interplanetary propagation. Additionally, both types also display a fractionation relative to photospheric abundances that depends on the first ionisation potential (FIP) which is similar to the FIP-effect described below in section 2.5.3. Recent studies using the Solar Isotope Spectrometer onboard the ACE satellite have found that the SEP isotopic composition varies widely from event to event, with the ²⁰Ne/²²Ne ratio varying by more than a factor of 4. It also seems that these variations change during the solar cycle. However, the long-term average ²⁰Ne/²²Ne ratio from in-situ measurements, which seems to be distinctly lower than that of the SW, supported for a long time a supposed noble gas component reported from lunar and asteroidal regolith material. This “SEP” noble gas component showed a relatively fixed composition different from the SW but, on the other hand, always required a very high abundance of up to 40% of the entire solar particle fluence, which made it questionable. In this thesis we could show that the “SEP” noble gas component is not reflecting a surprisingly high abundant SEP component but rather is an artefact of a separation of SW isotopes within the regolith grains upon implantation.

2.5 From the Sun to the Solar Wind – Processes Controlling Compositional Variations

The composition of the OCZ remained largely unaltered over the past 4.6 Gyr (Geiss et al., 1995). However, some processes have somewhat changed the isotopic and elemental composition of the OCZ from that of the bulk Sun and, in turn, change the composition of the SW from that of the OCZ. Incomplete hydrogen burning and gravitational settling both influenced the composition of the OCZ over the Sun’s lifespan, whereas inefficient Coulomb drag and the first ionisation potential and first ionisation time of an element fractionate the SW composition at the source region. These processes have to be corrected for to derive precise photospheric values from the SW record or by the interpretation of helioseismology and spectroscopy data. Though all four processes will be discussed in the following, only Coulomb drag and the first ionisation potential are a subject of this study.
2.5.1 Incomplete Hydrogen Burning

$^3$He is an intermediate product of the ppl-chain. In the central core of the Sun ($M/M_\odot < 0.2$, where $M/M_\odot$ is the distance from the centre relative to the solar radius) temperatures are sufficiently high to convert $^3$He into $^4$He. In the outermost layers ($M/M_\odot < 0.9$) the $^3$He content remains basically unchanged because even the p – p reaction does not proceed. Near $M/M_\odot \sim 0.6$, where temperatures are sufficiently high to produce $^3$He but remain too low to further process $^3$He to $^4$He, a $^3$He rich region gradually evolves (Bochsler et al., 1990). This region within the radiation zone should grow during solar history and slowly move outward if no large scale mixing throughout the Sun is involved (Bochsler, 1992).

In standard solar evolution models, the $^3$He abundance of the OCZ reflects the sum of primordial $^3$He and original (“cosmological”) deuterium in the solar nebula (Gloeckler and Geiss, 2000). Deuterium was burned in the pre-Main Sequence phase of the Sun and could not survive at the base of the OCZ. Non-standard evolution models involving modest admixture to the OCZ of material from the $^3$He regions below predict an increase in the $^3$He/$^4$He ratio at the solar surface (Bochsler et al., 1990). However, most recent $^3$He/$^4$He measurements of lunar regolith grains indicate that long time averages of the helium isotopic composition in the solar wind have been constant within a few percent during the past 3–4 Gyr (Heber et al., 2003; Wieler and Heber, 2003). Thus mixing of the Sun down to intermediate depth was very limited throughout much of solar history (Geiss et al., 2004).

2.5.2 Gravitational Settling

The solar He abundance can very precisely be measured by helioseismology (Gough, 1998). Data of He/H show that He is depleted in the OCZ by 10% relative to the initial value (Bochsler, 2000); a difference that is attributed to gravitational settling of He throughout solar history. This process also affects the isotopic composition as, e.g., $^3$He settles more slowly than $^4$He, which has resulted in a decrease of 2–3% in the $^3$He/$^4$He ratio in the OCZ (Gautier and Morel, 1997). According to Vauclair (1998) settling of elements at the lower boundary of the convection zone is not only due to gravitation but also thermal diffusion and radiative acceleration and leads to abundance variations of ~10% also of elements heavier than He. However, Bochsler (2000) argues that the effect of element settling is generally not taken into account when comparing photospheric abundances (relative to hydrogen) with other solar system abundances. First, accounting for this effect is not needed because the uncertainties of photospheric abundances are usually larger than the expected variations by gravitational settling. Second, gravitational
settling seems to affect elements in the same mass range similarly. Hence, the error introduced by using photospheric abundance ratios (except those including hydrogen) instead of using initial solar abundances accounting for the effect of gravitational settling is probably smaller than 10%. However, gravitational settling might have measurable influences on the isotopic composition data analysed in the laboratory, e.g. of Genesis samples, should provide isotopic ratios with an uncertainty in the %-range. Therefore, isotopic effects due to gravitational settling reflected by compositional variations in the SW should become identifiable.

2.5.3 First Ionisation Potential and Time

Atoms in the photosphere are generally believed to be in a neutral state (Bochsler, 2000). In the chromosphere atoms get ionized by the coronal UV radiation and subsequently separated from the neutrals as they move into the corona. A number of mechanisms have been suggested to explain the ion-neutral separation but there is still no generally accepted model (Feldman and Widing, 2002). However, it is accepted that ionisation plays a crucial role in establishing solar wind abundances. From the systematic of coronal abundances as derived from solar energetic particles (SEPs) or from solar wind ions, compared with the photospheric abundances, it appears that the first ionization potential (FIP) is the most important atomic parameter. The FIP of an element is the energy required to strip it of the first electron, which produces a positive electric charge. Elements with a low FIP are ionised more easily and quicker than high-FIP elements, leading to an enrichment of low-FIP elements in the corona where the SW roots. The FIP-dependence of coronal abundances derived from solar energetic particles (SEPs) shows a

![Fig. 1.4.](image-url) Solar energetic particle derived coronal abundances relative to photospheric abundances and normalized to the oxygen abundance (the so-called first ionization potential (FIP) plot, from Bochsler, 2000). Left panel: The solar energetic particle (SEP) derived abundances are from (Breneman and Stone, 1985) using an improved Q/M fractionation law (Garrard and Stone, 1994). The photospheric abundances are from Grevesse and Sauval (1998). Right panel: Interstream solar wind abundances normalized with oxygen relative to photospheric abundances as given by Grevesse and Sauval (1998).
one-step pattern (Fig. 1.4): elements with low FIP, i.e., FIP below the Lyman α limit at 10.2 eV, are generally overabundant by a factor of 3–5 in the equatorial corona; and elements with FIP >10.2 eV show no enrichment in comparison with the photosphere (Bochsler, 2000). The FIP-pattern deduced from SW ions (Fig. 1.4) shows a similar step, though less pronounced and with a less defined low-FIP plateau. Additionally, coronal hole–associated solar wind appears to be less fractionated by the FIP-effect than interstream solar wind. The enrichment of low-FIP elements in coronal-hole related SW relative to the high-FIP elements amounts to typically a factor of 2 only (Geiss et al., 1995). The FIP-dependence as described is valid for most elements. However, the heavy noble gases Kr and Xe are exceptional, as they are both high-FIP elements but are enriched in the SW relative to the photosphere (Wieler and Baur, 1995). This abnormal behaviour from the FIP-effect can be explained, if the first ionisation time (FIT) is assumed to be the relevant parameter controlling elemental fractionation.

2.5.4 Coulomb Drag

According to theoretical models the importance of the FIP- and FIT-effect for isotopic fractionation seems to be rather small (Marsch et al., 1995; Peter, 1996). However, Kallenbach et al. (1998) reports possible isotopic fractionation of Si, Ne, and Mg isotopes in the SW that varies with SW speed. Their data indicate a small systematic depletion of the heavier isotopes in the low-speed SW on the order of (1.4±1.3) % per amu compared to their abundances in the

![Fig. 1.5. Expected isotope fractionation factors associated with He/H depletion factors. For instance, a depletion of He/H to 30% of its normal value in the equatorial streamer belt implies an enhancement of the $^{28}\text{Si}^{12+}/^{30}\text{Si}^{12+}$ ratio up to 1.7% over its normal coronal value. (from Bochsler, 2000)
high-speed streams from coronal holes. According to Kallenbach et al. (1998) the influence of gravitational settling in the OCZ, the FIP-effect or interplanetary shocks on the observed fractionation can be ruled out. Instead, the theoretical model of an inefficient Coulomb drag (Bodmer and Bochsler, 1998) predicts the observed trends with right magnitude and leads to minor elemental fractionation also.

Coulomb drag works by the momentum transfer of protons to heavier ions. In the transition region and the corona (~1-10 solar radii) where Coulomb drag is the relevant process, protons are incorporated into the SW by their thermal speed. Coulomb friction with protons couples the heavier ions into the proton flux and finally accelerates them to the proton speed. The acceleration is dependent on the mass of an atom and its charge state, and therefore is affecting not only the elemental but also the isotopic composition.

Regarding isotopes of the same element and the same charge state, an inefficient Coulomb drag preferentially accelerates the light species which leads to an enrichment of light isotopes in the SW relative to its source (Fig. 1.5). To carry a certain ion species in the SW a minimum flux of protons is required. The efficiency of Coulomb drag is reduced in regions where the protons are less abundant. This is observed especially for current-sheet related SW where a rapid superradial expansion of the magnetic field lines regulates the number density of protons right at the roots (Fig. 1.6). If the occasional helium depletion in the equatorial current sheet to He/H abundance ratios as low as 0.01 is indeed caused by the special magnetic field topology and the concurrent inefficiency of Coulomb drag in the inner corona, then isotopic fractionation effects of the order of 30% have to be expected for the $^3\text{He}/^4\text{He}$ ratio (Bochsler, 2000). For heavier elements the theoretical fractionation is smaller as shown in Fig. 1.3 for H, O, N, Ne, Mg, and Si isotopes.

![Fig. 1.6. Illustration of the acceleration of heavy ions due to coulomb collisions with protons. In the current-sheet magnetic field lines experience are rapid superradial expansion, compared to e.g. the inter-stream region, which regulates the number density of protons right at the roots as they follow along the magnetic field lines. This reduction in the proton number density in turn leads to an inefficiency in the acceleration of ions heavier than protons and lowers the fluence of these particles in the current-sheet related low-speed solar wind.](image-url)
2.6 Solar Wind Sample Return Missions

To study the composition of the SW and the OCZ by in-situ analysis of the SW or by the trapping of SW ions in solids on sample-return missions, the in-situ instruments or trapping devices have to be placed outside of any strong magnetic field, e.g. generated by large planetary bodies, which deflects the charged particles (Fig. 1.7). One possible position was used by experiments on the Apollo missions that collected SW particles on the lunar surface (Geiss et al., 2004). The Moon has only a very weak magnetic field and essentially no atmosphere in comparison to the Earth and thus charged SW particles reach the lunar surface essentially unhindered. Another possibility is to place a satellite in a solar orbit at a position where it can be stationary relative to the Sun and the Earth because it is only affected by the gravity of the two large objects. Such a position is called Lagrangian point. Five Lagrangian points exist around any orbital configuration of two larger objects. The most suitable position for SW analysis is the Lagrangian point L₁, which is about $1.5 \times 10^6$ km away from Earth towards the Sun where gravitation of both compensates each other. NASA and ESA operate a number of spacecraft in orbit around the Sun-Earth L₁ point, namely SOHO, ACE, the International Sun/Earth Explorer 3 (ISEE-3), and the Genesis satellite.

Fig. 1.7. This illustration shows a CME blasting off the Sun’s surface in the direction of Earth. The left portion is composed of an EIT 304 image showing the sun (extreme UV light with the He" emission line at 304 Å, formed primarily at 60,000-80,000 K) superimposed on a LASCO C2 coronagraph white light image of the corona (both instruments onboard SOHO). The paths emanating from the Earth’s poles represent magnetic field lines that deflect the charged SW particles. The magnetic cloud of plasma can extend to 30 million miles wide by the time it reaches earth. Objects in the illustration are not drawn to scale. (Image taken from the SOHO web page)
2.6.1 Solar Wind Composition Experiment

The solar wind composition (SWC) experiment was designed by scientists from the University of Bern to measure elemental and isotopic abundances of the light noble gases in the solar wind, and to investigate time variations in the solar-wind composition (Geiss et al., 2004). On July 20, 1969, Apollo 11 landed in Mare Tranquillitatis and at 03:35 UT Astronaut Edwin E. Aldrin deployed the SWC device at a distance of 4 meters from the lunar module (Fig. 1.8). This was the first of six lunar landings. On the next four, astronauts also deployed a SWC aluminium foil. The foils with an exposed area of 30 cm × 130 cm were fixed on a telescopic pole and exposed to the solar corpuscular radiation for 77min on Apollo 11 to 45hr 5min on Apollo 16. Finally they were returned to Earth, where the collected noble gas particles were analysed in ultra-high vacuum mass spectrometer systems mainly at the University of Bern. Analyses were carried out over more than one decade (Bühler et al., 1969; Geiss et al., 1970a; Geiss and Reeves, 1972; Cerutti, 1974; Filleux, 1975; Filleux et al., 1980) and the obtained

Fig. 1.8. Apollo 11 Astronaut Edwin E. Aldrin standing besides the SWC experiment in Mare Tranquillitatis on July 21, 1969. Photograph by Commander Neil A. Armstrong (NASA Photo AS11-40-5873).
results are still more precise than can be obtained by spacecraft instruments today (Wiens et al., 2004). However, due to the short exposure duration these measurements reflect only short-term variations in solar wind composition and analyses are limited to noble gases. Historically, these experiments are of great importance in that they established that deuterium in terrestrial oceans \((D/H = 1.6 \times 10^{-4})\) is enriched by almost an order of magnitude relative to the protosolar composition \((D/H = 1.94 \times 10^{-5})\), and also that atmospheric neon is distinctly different from the solar composition.

2.6.2 The Genesis Mission

The Genesis mission is the fifth in NASA's Discovery program which is a series of relatively low-cost scientific space missions (Burnett et al., 2003). It was the first sample return mission from beyond the orbit of the Moon. Genesis was launched on August 8, 2001, and crash-landed on September 8, 2004 because a design flaw prevented the deployment of its main parachute. After launch, Genesis spent ~5 months to reach the \(L_1\) point, collected SW for ~28 months during 5 halo orbits around \(L_1\), and returned to earth in 5 months to land in the Utah desert (Fig. 1.9).

Genesis successfully followed the footsteps of the SWC experiment in that it collected SW particles by implantation into solid targets. But, the Genesis mission had several improvements compared to the SWC experiments: 1) a several hundred times longer exposure time of targets

\[\text{Fig. 1.9. The Genesis spacecraft trajectory from Earth to L1 and return (from Burnett et al., 2003). The Earth-L1 distance is about } 1.5 \times 10^6 \text{ km. The lunar orbit is shown for scale. Arrows indicate the outbound and return trajectories. There are 5 'halo' orbits about L1. The large loop behind the Earth on the return trajectory positions the spacecraft for daylight re-entry.}\]
allows analysis of almost all elements in the periodic system in contrast to the SWC experiments that allowed the analysis of He, Ne, and Ar only, 2) a SW concentrator raises SW fluences in one special target by a factor of 20 to allow analysis of nitrogen and oxygen (Nordholt et al., 2003; Wiens et al., 2003), 3) no severe contamination of SW-loaded dust particles had to be expected as on the lunar surface, 4) constant monitoring of SW conditions with instruments onboard Genesis and the satellites ACE and SOHO, 5) the possibility to study the long-term SW variability due to the long exposure time, and 6) differentiated SW sampling at different conditions by three SW regime arrays. Based on these improvements the main objectives of Genesis are (a) to obtain solar isotopic abundances to the level of precision required for the interpretation of planetary science data, (b) to significantly improve knowledge of solar elemental abundances, (c) to measure the composition of the different solar wind regimes, and (d) to provide a reservoir of solar matter to serve the needs of planetary science in the 21st century (Burnett et al., 2003).
Fifteen different high purity target materials were exposed onboard the Genesis satellite, mostly as hexagonal wafers attached to 5 different collector arrays (Fig. 1.10). The most important selection criteria for the materials were: bulk purity, surface cleanliness, and the retentivity for solar-wind components under mission conditions. Further, these flight-qualified materials can physically withstand the environment the spacecraft sees during launch, cruise, and landing phases. Finally, the collector-material designated for a given element (or isotope) had to be compatible with the technique(s) most likely to be used for the eventual laboratory analysis (Jurewicz et al., 2003). The targets were either exposed to the SW for the entire time the science canister of the satellite was open or only when one specific SW regime was identified. Identification of regimes was performed in-situ by an algorithm (Neugebauer et al., 2003) on the basis of data from the Genesis ion monitor and the Genesis electron monitor (Barraclough et al., 2003), measuring the proton speed, number density, and temperature, the $\alpha$-particle number density, and the streaming direction of electrons (see Fig. 1.11 for the arrangement of instruments and targets onboard the Genesis satellite). This algorithm also protected the satellite from solar high energy events and Genesis went into a safe mode for more than 10 days during the solar super storms of 2003.

The targets of Genesis were assiduously handled when mounted on the satellite prior to launch. However, Genesis suffered from two massive backstrokes. During exposure in space a molecular film mainly consisting of Si, O, C, and F formed on many targets (Allton et al., 2006). One source of this film (informally known as Brown Stain) appears to be evaporated and recondensed glue used to fix screws with which the target of this study was mounted. Though the trapping of ions was not substantially affected by the molecular film, it caused another substantial problem. Because it got polymerized by solar UV radiation it became difficult to remove and compromised the effectiveness of several methods used to extract the trapped SW particles. And, due to the crash-landing of the satellite almost all samples were broken into shards ranging from cm-size to submicron particles. Additionally, dust and wet soil had access to the science canister when it cracked while impacting into the Utah desert at a speed of ~360 km/h. Both mishaps complicate the handling and analyses and caused substantial delay of the scientific schedule. However, according to first results and studies of target materials it seems that all major science objectives of Genesis can be achieved without larger restrictions. The target flown on Genesis analysed in this study is a bulk metallic glass which was especially designed for the purpose of light noble gas measurement by a stepwise etching technique to study the depth dependence of implanted ions. The BMG collected solar particles for the entire Genesis exposure time which allows conclusions about the bulk SW composition. Noble gases are of special interest for studying the SW as they are almost chemically inert and relatively abundant.
in the SW, and therefore well suited to study processes causing compositional variations in the SW and the photosphere.

References


Burnett D. S., Barraclough B. L., Bennett R., Neugebauer M., Oldham L. P., Sasaki C. N.,


II. **SAMPLE TREATMENT AND NOBLE GAS ANALYSIS**
The sample material we analysed in this study is a metallic glass that was especially designed for the purpose of noble gas trapping and storage (Fig. 2.1). This amorphous metal was custom made at the California Institute of Technology (CalTech) by C.C. Hays (2001; 2005) and the final glass composition of Zr\textsubscript{58.5}Cu\textsubscript{15.6}Ni\textsubscript{12.8}Al\textsubscript{10.3}Nb\textsubscript{2.8} (in atom percent) was chosen after several etching tests by V.S. Heber (2002). The basic idea to choose such a complex material was to allow homogeneous etching to release the implanted gas stepwise with a high depth resolution. The second important reason was to trap the implanted ions in the material without subsequent diffusive losses or migration of trapped ions within the glass. This expectation, which was based on the fact that a crystal lattice or grain boundaries are absent and thus no preferred paths for diffusion of trapped gases exist, was unfortunately not fulfilled for He as turned out after analyses of several flown BMG samples. However, scanning electron microscopy (SEM) images show \(\mu\)m-sized dendrites in the glassy matrix that are enriched in Zr, Cu, and Ni, and Al-deficient relative to the surrounding matrix. These Laves-phase like dendrites result from imperfect quenching of the melt during the casting process because of a low critical cooling rate to vitrify the composition of \(R_c = 1.75\) K/s. Due to the low areal fraction of \(<9\%\) (Hays, private communication) and their distribution the effect of these dendrites on trapping or diffusive losses can be neglected, however.

Fig. 2.1. The image shows the entire flown BMG target after removal. The target is fully intact without major superficial damage.
2. SAMPLE FRAGMENTATION

The specimen was prepared by vacuum injection of the liquid metal (Tm = 830 °C) into a metal mold at Howmet Research Corporation (HMM), Michigan. The dimensions of the entire plate were 13.97 cm by 30.48 cm with a thickness of 0.30 cm. The large plate was not fully amorphous which required the removal of the crystalline part. Then, the amorphous 1-2 mm thick layers were reheated to an annealing temperature of 400 °C to relieve quenched stress into residual stress. The amorphous plate was wire-machined to desired final collector geometry (circular) and hand-polished with 0.25 µm diamond powder. The amorphous material was characterized via standard methods (X-ray, DSC, TMA, etc.). The final dimension of the glass collector was 6.48 cm in diameter with a thickness of 1.02 mm.

After sample return and removal from the science canister of the Genesis satellite ~1/5 of the BMG was subdivided at ETH into 35 separate aliquots for analysis. The target was cut with a diamond blade from the backside to a remaining thickness <100 µm under constant airflow cooling to prevent diffusion of trapped gases. Finally, the single aliquots were broken off in order not to lose any of the area exposed to the SW (front side) by the cutting process.

3. CLEANING

After removal of targets from the Genesis science canister a brown stain was discovered by the Genesis curation team on several samples, which displayed transitions in colour and rainbow like fringes (Fig. 2.2). First studies showed that this contamination is a molecular film consisting of Si, C, O, and F (Fig. 2.3). Extensive surface mapping of the BMG using x-ray photoelectron spectroscopy together with S. Tosatti at the Department of Materials at ETH confirmed these results (Grimberg et al., 2005). Additionally, six XPS profiles were taken either between two screw holes or between one of the holes and the centre of the target. Along the profiles we measured ratios of Zr/Si and Zr/C to study the distribution of the contamination (the profiles are plotted in Chapter VI as the respective Fig. 1). Small ratios indicate a relatively thick contamination layer since the Zr signal only stems from the BMG while the Si and C signals represent the molecular film. Indeed, film thickness is highest near the holes, confirming the suspicion that the molecular film was caused by evaporating glue. However, the Zr signal is visible everywhere along the six profiles. Hence, the film thickness nowhere exceeds about 8 nm, the maximum depth of detection of the XPS system used.
Though the film thickness $<8$ nm was not harming the trapping of solar wind ions in space, it still caused problems because of its resistance against $\text{HNO}_3$, the etching solvent used for CSSE. In a first etching run of an uncleaned sample, essentially no He and Ne above blank level were detected and hence this run was abandoned after nine steps. As shown in Fig. 2.4, the neon isotopes measured in all steps had approximately atmospheric composition.

A cleaning procedure to gently remove the molecular film without affecting the BMG surface itself thus had to be developed. A very effective but gentle method used in the semiconductor industry to remove contaminants from wafers is reactive-ion etching, which will be termed plasma cleaning in the following. This method uses a chemically reactive plasma which is generated in a vacuum from a certain gas. The sample to be cleaned rests on a wafer platter which is electrically isolated from the grounded vacuum chamber. Plasma is initiated in the system by applying a strong radio frequency electromagnetic field to the wafer platter.

**Fig. 2.2.** Detailed image of the BMG showing part of a screw hole to the right (black), the area of the BMG shaded by the screw top (centre) and the exposed area (left). On the latter the molecular film is visible as the grey colour gradient.

**Fig. 2.3.** XPS spectra of an uncleaned flown BMG sample. The peak height is not directly comparable to the element abundance. Very distinct peaks are visible for O and C, which stem from the molecular film as well as the superficial atmospheric contamination. Si and F stem from the molecular film only. The small thickness of contamination is indicated by the presence of the Zr and Nb peak belonging to the BMG substrate. The elements are given with their binding state.
field is typically set to a frequency of 13.56 MHz, applied at a few hundred watts. The oscillating electric field ionizes the gas molecules by stripping them of electrons, creating a plasma. The wafer platter builds up large negative voltage due to absorbed electrons whereas the plasma is charged slightly positive. Because of the large voltage difference, positive ions tend to drift toward the wafer platter, where they collide with the samples to be etched. The ions react chemically with the materials on the surface of the samples but can also sputter some material. We first used the method of plasma ashing with pure oxygen as the reactive species, where the charging of the wafer platter is suppressed, thus the plasma ions are not accelerated and only react chemically with the contamination. The subsequent CSSE run was not successful and showed, similar to the uncleaned sample, mostly atmospheric composition of the released gases (Fig. 2.4). This suggested that ashing only oxidised the contaminants, leading to freshly formed silicon oxide that is as well resistant against HNO₃.

Fig. 2.4. ²⁰Ne/²²Ne ratios versus etching time of two flown BMG samples, one untreated and one cleaned with plasma ashing. Both samples show only very little to no solar wind contribution in their Ne composition, which proves that plasma ashing alone is not removing the molecular contamination.

Fig. 2.5. Secondary electron images of flown BMG samples that were treated differently. In image A) of an uncleaned sample that was etched for 1600 min. in HNO₃, dendrites are barely visible as a positive relief underneath the molecular contamination. Image B) shows a BMG sample after plasma ashing with O₂ and plasma etching with SF₆. Material inhomogeneities are better visible since the surface contamination is absent. Dendrites showing a positive relief are clearly visible now (white arrows) as well as surface structures like the diagonal scratch. Image C) shows a sample cleaned by plasma ashing and etching, which was exposed at CSSE to HNO₃ for over 13.000 min. The dendrites show a negative relief because they react more rapidly to the acid. The surface relief proves that plasma cleaning removed the contamination successfully. The surface structure of the glass matrix is very smooth indicating homogeneous etching.
Finally we used a combination of carbon oxidisation by gentle oxygen plasma ashing followed by a plasma etching procedure. For plasma etching the reactive ions are accelerated due to the negatively charged wafer platter. We used SF$_6$ gas as the reactive species to chemically transfer Si-compounds into volatile SiF$_4$, which was accompanied by sputtering at a low rate due to accelerated SiF$_6$ ions. This procedure was entirely successful as is proved by the subsequent CSSE runs (see sections IV, V, and VI) and visible in images shown in Fig. 2.5. The entire plasma cleaning was carried out together with O. J. Homan at the Centre for Micro- and Nanoscience at ETH.

4. NOBLE GAS MEASUREMENT

All noble gas measurements were carried out in the noble gas laboratory of the Departement Erdwissenschaften of ETH Zürich. The work was supported by the guidance of Dr. Heinrich Baur who designed the mass spectrometer and wrote the machine software, and the technical maintenance by our staff Urs Menet, Andreas Süsli, Donat Niederer and Bruno Rütsche.

4.1. Closed System Stepwise Etching - CSSE

The main extraction method used for this work is the release of noble gas via closed system stepwise etching (CSSE). As already mentioned, the BMG was custom made for this purpose and its etching behaviour tested by V. S. Heber (2002). One main advantage of this technique over the widely used stepwise extraction methods pyrolysis or combustion is that the gas is released essentially at room temperature by dissolving the target and not by diffusion at elevated temperatures. This avoids heat induced diffusive fractionation typically affecting the gas released at higher temperatures. The other advantage of CSSE is its high depth resolution of a few nm. This becomes very important for samples where the gas to be analysed is sitting in the outermost 100 nm of a material and variations in the noble gas composition are relatively small.

The CSSE line used for this study (Fig. 2.6) is essentially the same as described by Heber (2002). Experimental details are explained therein and by Graf et al. (1990) or H. Busemann (1998). All acid-exposed parts are made of Au or Pt only. The concentrated HNO$_3$ (1 ml, 65%) in the acid container is held at -20°C when stored only, but at room temperature during the actual etching procedure. The acid vapour is separated from the BMG aliquot stored in the sample container by valve G2A. One of the main differences to former studies with this line
II. SAMPLE TREATMENT AND NOBLE GAS ANALYSIS

is the larger volume of the sample container to allow horizontal storage of the single BMG aliquots. This is necessary to avoid drop formation of the acid on parts of the BMG surface that would touch the ground of the container.

Before each CSSE run new acid was loaded into the container and carefully degassed from dissolved noble gases by several cycles of heating to 50°C and subsequent freezing down to -198°C and later -20°C. The degassed vapour/gas mixture was pumped with an acid resistant turbo molecular pump (labelled acid pump in Fig. 2.6) through a special vacuum line equipped with a cold trap (-80°C). The residing acid blank gas was measured via a bypass that is connected to the cleaning system right after the sample container, to allow determination of acid blanks even when a sample was already loaded.

To etch the actual sample, HNO₃ vaporises at room temperature and expands into the sample container when valve G2A is opened. After the etching process that lasted usually between 30 min. and several days, the acid container was cooled down to -90°C for 65 min. and thus the HNO₃ vapour was removed from the sample container. This re-freezing stopped the etching process almost completely. The temperature was held at -90°C in order not to affect the mobility of Ar. The released noble gases subsequently passed valve G1A to be expanded into the cleaning part.
II. SAMPLE TREATMENT AND NOBLE GAS ANALYSIS

4.2. Pyrolytical Gas Extraction

For determination of the total gas amount and composition the samples were molten in a furnace. The molybdenum crucible is heated via electron bombardment to temperatures of 1500°C at most in the case of Ni foils and Ax1 analysis from the CASYMS irradiation experiments. For all samples analysed, similar amounts of unirradiated target material were measured for their blank noble gases content prior to analysis. The determination of the bulk Ar composition in the Genesis BMG was not possible via pyrolysis because blank gas contributions from the furnace were far too high. These values have been obtained from the integration of all etching steps in the respective run.

4.3. Gas Cleaning and Separation

The released gases were exposed to a series of getters in the stainless steel part of the line to remove interfering gas species like H\textsubscript{2}O, CH\textsubscript{4}, or CO\textsubscript{2}: first CaO, then TiZr, and finally AlZr (SAES®). The procedure was similar for CSSE and pyrolysis, with the exception of a getter made of CaO powder that was used for CSSE only. The released Ar was separated from He and Ne with activated charcoal held at -196 °C.

4.4. Mass Spectrometry

All measurements in this work have been carried out on the Albatros mass spectrometer at ETH. It is a non-commercial instrument with a 90° flight tube (21cm radius) and a mass resolution M/ΔM of 550. This is sufficient to resolve peaks of \(^3\)He and HD. The gas is ionised with electrons of 45 eV energy, which reduces the double ionisation of \(^{40}\)Ar and CO\textsubscript{2} and thus suppresses the interferences on \(^{20,22}\)Ne. The ionised noble gases were subsequently measured on two collectors, a Faraday cup for high ion currents and a multiplier operating in counting mode with an upper limit of 550,000 Hz, corresponding to \(3 \times 10^{-8} \text{ cm}^3\text{STP} \ ^4\text{He}\).

4.5. Interferences

The background gases are continuously monitored throughout all measurements and they are relatively constant during a single run. The mass resolution of Albatros is sufficient to resolve HD from \(^3\)He but also \(^{20}\)Ne from H\textsubscript{2}O\textsuperscript{18}O as has been shown by N. Vogel (2003). The interferences of \(^{40}\)Ar\textsuperscript{++} on \(^{20}\)Ne and CO\textsubscript{2}\textsuperscript{++} are suppressed by the low filament current of 45 eV. The contribution due to remaining interferences is generally below 1% for blank gas analyses.
and always smaller than 0.1% of the sample gas. Fortunately, these corrections are lower than uncertainties for all sample measurements.

4.6. Blanks

Blank gases corrected for all analyses were measured for the entire system used for a certain measurement and for all sample materials. The highest contributions from pyrolysis stem from the hot crucible and increase with temperature. However, blank gas contributions were only substantial for Ar, which hampered the measurement of the bulk composition of the flown BMG via pyrolysis. Blank Ar contributions for pyrolysis as well as for CSSE were corrected assuming $^{40}\text{Ar}$ to be entirely atmospheric. This is reasonable since $^{40}\text{Ar}$ is absent in the SW. The corrected fraction reached up to 95% due to gas incorporated during the manufacturing process in an Ar-rich atmosphere and minor from the degassing crucible. For gas rich steps with CSSE the correction via $^{40}\text{Ar}$ was generally 20-40%.

For CSSE two different types of He and Ne blanks can be distinguished. Procedural blanks cover the entire extraction line without sample and acid container and are measured after every five etching steps. Acid blanks are the second type which has to be considered and that is usually higher than the procedural blank. Blank corrections reached 1.5% for $^{21}\text{Ne}$ and 2.0% for $^{22}\text{Ne}$ in gas poor steps, but never exceeded 0.01% for $^{20}\text{Ne}$ or the He isotopes.
References


III. Irradiation Experiments
1. INTRODUCTION

The basic principle of the Genesis mission is the trapping of solar wind (SW) ions into solid materials to return them back to earth (Burnett et al., 2003). This technique follows the footsteps of two successful missions in space, the Apollo solar wind composition (SWC) experiment that captured solar wind ions on the lunar surface in aluminium foils (Geiss et al., 2004) and the COLLISA experiment that trapped interstellar light noble gases in a Be-oxide layer on BeCu foils onboard the MIR space station (Zastenker et al., 2002a; Zastenker et al., 2002b; Salerno et al., 2003; Busemann et al., 2006). For these studies extensive calibration tests about the trapping behaviour of the target materials used have been carried out using facilities at the University of Bern (Bühler et al., 1966; Meister, 1969; Filleux et al., 1980; Bassi, 1997). However, little is known about trapping efficiencies and depth ranges at energies below 5 keV/amu for other target materials like the bulk metallic glass (BMG) analysed in this study or the other 14 types of material exposed on Genesis (Jurewicz et al., 2003).

One essential tool to examine the irradiation of ions into matter is the SRIM (Stopping and Range of Ions in Matter) simulation code by Ziegler (2004). SRIM was very widely used for the prediction of mass separation effects on the impinging ions upon implantation and the final interpretation of the results obtained from different Genesis targets. According to data plots on the SRIM homepage www.srim.org the best fit of experimental data with simulation results is achieved at energies between 10 and 1000 keV/amu, thus clearly above the maximum implantation energy of 5 keV/amu for SW ions.

To check the accuracy of SRIM predictions for typical SW energies and to have an experimental calibration for the mass separation effects upon implantation we performed extensive irradiation experiments (with substantial support by A. Etter and F. Bühler) with the calibration system for mass spectrometers (CASYMS) in Bern (Ghielmetti et al., 1983; Steinacher et al., 1995). These experiments focussed on the trapping behaviour of different target materials for He and Ne isotopes and the depth distribution of Ne isotopes within the BMG. All experimental data were compared to results from SRIM simulations.

2. TRAPPING EFFICIENCY

An ion hitting a solid has a certain probability of becoming trapped in the material and not to be backscattered. This trapping efficiency ($\eta$) depends on various parameters, such as mass and
energy of the projectile, angle of incidence and the chemical properties of a target material. All these parameters are inputs to the SRIM code. The irradiations at the CASYMS were carried out to experimentally determine $\eta$ for different materials to measure the effect of atomic mass of target and projectile on the loss by backscattering. Additionally we intended to check SRIM at low energies and for the complicated composition of the metallic glass used.

According to Bühler et al. (1966), the actually measured gas amount can be smaller than it would follow from $\eta$, because ions already trapped get released during the ongoing irradiation. The different mechanisms responsible for the loss of trapped particles by further ion bombardment are: a) sputtering of target surface layers that contained trapped gases; b) an increased diffusion constant due to radiation damage; c) enhanced diffusion due to local heating of the lattice during the slowing down process; and d) diffusion due to an increased target temperature during exposure in space by solar radiation. For Genesis targets, a), b) and c) can be ruled out because the exposure time was too short for efficient surface sputtering and the amount of high energetic particles was too low to produce severe radiation damage. Instead, mechanism d) is a process that might have affected the collecting efficiency of the BMG since exposure temperatures were expected to reach 180°C in space, a fact that had to be considered and that SRIM calculations cannot account for.

3. EXPERIMENTAL TECHNIQUE

3.1. CASYMS

The ion irradiation experiments were carried out at the calibration system for mass spectrometers (CASYMS) at the University of Bern (Ghielmetti et al., 1983; Steinacher et al., 1995). The CASMYS operates with a Nier electron bombardment source and a 90° sector angle UHV mass spectrometer to separate the different isotopes (Fig. 3.1). The gas to be implanted was introduced into the ionisation chamber from monoisotopic gas reservoirs for $^3$He and $^4$He, whereas $^{20}$Ne and $^{22}$Ne were separated from an air mixture. After separation the ions were accelerated in the drift tube to energies ranging from 0.83 to 3.0 keV/amu, depending on the experiment, and implanted with an angle of incidence of $\sim0^\circ$, similar to the exposure situation of Genesis in space. The different isotopes were subsequently implanted in the order of increasing mass. In hindsight, the opposite order would have been more reasonable to avoid any knock-on effects on the light isotopes in shallow depth by the following heavy ones.
III. Irradiation Experiments

The ion flux was measured every 40 min during irradiation with a channel electron multiplier (CEM) in combination with a Faraday cup, both attached to the beam scanner located in the main vacuum chamber. The Faraday cup (aperture 5 cm\(^2\)) was used as an absolute standard measuring the ion current in the beam centre whereas the CEM, with a sensitive area of 0.1 cm\(^2\) only, measured the intensity in the beam centre as well as on 7×7 spots, covering the whole effective beam area of 10 cm by 10 cm to map the beam distribution. An ion flux ranging from \(2\times10^{5}\) cm\(^{-2}\)s\(^{-1}\) for \(^{22}\)Ne to \(1.4\times10^{6}\) cm\(^{-2}\)s\(^{-1}\) for He isotopes was obtained and the fluence was chosen to be at least 100 times above the material blank of the BMG. The estimated uncertainty of the total integrated beam current measurement was \(\sim10\%\).

Beam profiles measured with the CEM along the horizontal and vertical axes (labelled y and z in Figs. 3.2 and 3.3) through the beam origin showed an ion distribution at low absolute energies (2.5-3.3 keV) with a variation of 10-20 % from the beam centre to the edges. At higher absolute energies (17.0-18.7 keV) as for Ne, variations were larger with a complex beam shape in both axes. Notably, the variations were higher along the vertical axis than along the horizontal axis. To homogenise the ion distribution on the target in the vertical direction, the target mount was moved through the beam in vertical direction with 21 1-cm steps lasting 30 s each. In a second irradiation run the target mount was moved continuously, at constant speed, in vertical direction with each crossing of the beam lasting 60 s. The duration of irradiation for one isotope varied from 15.5 hours to more than 42 hours depending on the inlet gas concentrations used.

Fig. 3.1. Schematic diagram of CASYMS showing the gas inlet, the ion source with the mass spectrometer and the beam expander on the right-hand side, the drift tube in the middle, and the main vacuum chamber with the beam diagnostics the sample mount is connected to on the left-hand side. Picture modified from Steinacher et al. (1995).
3.2. Samples

The BMG used for the irradiation experiments labeled Ax1 is of similar composition as the flown BMG and consists of relatively heavy transition metals $\text{Zr}_{58.5}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3}\text{Nb}_{2.8}$ (atomic weight percent). However, the Ax1 target had somewhat different material properties compared to the BMG actually flown, namely, it contained less dendritic crystals due to a better quenching of the glass matrix in the manufacturing process.
Because of uncertainties in the flux measurement and thus in the determination of the amount of implanted ions, foils of Al, Ni and Au were irradiated simultaneously. These targets were used as standards for a relative comparison with SRIM, especially Al where backscattering was known to be small due to the small atomic weight of Al, and to study the effect of the atomic mass on trapping. Beside Apollo like Al-foils we used commercial foils of 99.9 % purity and 10 to 20 µm thickness. All targets were cleaned with ethanol prior to irradiation and care was taken not to touch the beam-facing surface after cleaning. The foils were cut in stripes of 50×5 mm size and mounted horizontally on a stainless steel plate (Fig. 3.3). The BMG was cleaved into 10×5 mm pieces and mounted amongst the foils.

Table 3.1. Trapping efficiencies for He, Ne, and Ar isotopes in Al and BMG

<table>
<thead>
<tr>
<th>Target</th>
<th>(^3)He</th>
<th>(^4)He</th>
<th>(^{20})Ne</th>
<th>(^{21})Ne</th>
<th>(^{22})Ne</th>
<th>(^{36})Ar</th>
<th>(^{38})Ar</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al(^a)</td>
<td>0.87±0.02</td>
<td>0.89±0.02</td>
<td>1.00±0.02</td>
<td>1.00</td>
<td>1.00±0.02</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>SRIM Al(^b)</td>
<td>0.899</td>
<td>0.919</td>
<td>0.992</td>
<td>0.993</td>
<td>0.994</td>
<td>1.000</td>
<td>1.000</td>
</tr>
<tr>
<td>CASYMS BMG(^c)</td>
<td>0.81±0.02</td>
<td>0.85±0.03</td>
<td>0.89±0.04</td>
<td>-</td>
<td>0.90±0.05</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>SRIM BMG(^b)</td>
<td>0.760</td>
<td>0.775</td>
<td>0.885</td>
<td>\textbf{0.892}</td>
<td>0.898</td>
<td>\textbf{0.948}</td>
<td>\textbf{0.952}</td>
</tr>
<tr>
<td>SRIM BMG(^d)</td>
<td>0.785</td>
<td>0.801</td>
<td>0.895</td>
<td>0.901</td>
<td>0.907</td>
<td>0.954</td>
<td>0.958</td>
</tr>
</tbody>
</table>

\(^a\) Experimental He and Ne trapping efficiencies adopted from Meister (1969), given with the 2σ uncertainty of the mean for He and Ne isotopes from the CASYMS irradiation results for 6 Al samples.

\(^b\) SRIM values modeled for the implantation energy of 0.83 keV/nucleon.

\(^c\) Experimental He and Ne trapping efficiencies determined at CASYMS with 0.83 keV/nucleon for 3 BMG samples, normalized to Al. Values are given with the 2σ uncertainty of the mean.

\(^d\) SRIM values modeled over the entire energy range of the SW velocity distribution from 0.3 to 4.0 keV/nucleon as measured for the Genesis exposure period.

Trapping efficiencies adopted for the BMG are plotted in bold. Uncertainties for values modeled with SRIM were estimated from uncertainties of the CASYMS experiments.

3.3. Analysis of Implanted Ions

The mass spectrometric analysis of all samples I carried out at the noble gas laboratory of the Department for Earth Sciences of ETH Zürich, Switzerland. The pyrolytical gas extraction and subsequent analysis procedure is similar to that described in Chapter II.

3.4. SRIM Calculations

The trapping efficiencies for all He and Ne isotopes I calculated using the SRIM2003 code. For each isotope three sets of 1,000,000 ions were calculated for all target materials and an implantation energy of 0.83 keV/amu.
SRIM calculates physical parameters such as ion ranges or ion losses due to backscattering based on the binary collision approximation by Robinson and Torrens (1974), accounting for the electronic stopping and interatomic potential (Ziegler et al., 1985). SRIM simulates the irradiation of a monoisotopic beam that has to be defined by the element species of the projectile, its atomic mass and implantation energy, the number of ions, and the angle of incident of the ion beam relative to the target surface. The target itself can be a multicomponent material that is defined by its compound elements in their stochastic fractions, the state of aggregation, the thickness of the substrate and its density, and the number of material layers that can be of different composition. The data sheet of a typical calculation includes the amount of backscattered or transmitted ions and the exact position (in three dimensions) for each of the trapped atoms in the target material.

### 4. RESULTS

#### 4.1. Trapping efficiencies

Very precise experiments on the trapping efficiency of aluminium foils were carried out by Bühler et al. (1966) and Meister (1969). In our study Al foils were used as a calibration standard for normalisation for all sample materials placed on the same vertical position. This normalisation eliminated the uncertainties in the absolute fluence determination. All ions were implanted with 0.83 keV/amu, corresponding to a SW at 400 km/s, with a fluence up to $1 \times 10^{11}$ atoms/cm$^2$. The average Al-normalised $\eta$ for $^3$He, $^4$He, $^{20}$Ne, and $^{22}$Ne deduced experimentally and with SRIM are shown in Tab. 3.1 and the adopted correction factors for the BMG are given in Tab. 3.2.

SRIM simulations predict that backscatter losses, and hence mass separation gets larger with increasing atomic mass of the target material, decreasing irradiation energy and decreasing atomic mass of the implanted ion. This leads to Al-normalised $^{20}$Ne/$^{22}$Ne ratios of gases remaining in the target ranging from 0.991 in Ni to 0.985 in Au. These SRIM values are consistent with measured data, although the sample-to-sample variation for some materials is relatively large (Fig. 3.4). However, the results make it reasonable to adopt the SRIM predicted $\eta$ for Ne and to deduce the correction factor $(1/\eta)$ for all isotopes $^{20,21,22}$Ne.

For $^3$He/$^4$He-ratios in a given material the sample-to-sample variation is considerably larger than it is for $^{20}$Ne/$^{22}$Ne ratios. The reason for this is still unexplained as experiments with
III. Irradiation Experiments

Improved homogenisation (mentioned above) of the ion distribution were showing similar differences. Nevertheless, the average measured $^3$He/$^4$He ratios are mostly lower than the values predicted by SRIM which suggests the predicted He isotopic mass fractionation to be doubtful. Additionally, the trapping efficiency of He isotopes in the BMG relative to that in Al was much higher than predicted and well reproducible. Therefore, the experimental values were adopted for the correction of data obtained from the flown BMG target. The $\eta$ values for He in the BMG not normalised to Al were calculated with $\eta$ values for Al given by Bühler et al. (1966) and Meister (1969) since their absolute calibration of the implanted ion fluence was better than the expected precision of our experiments at the CASYMS (F. Bühler, personal communication).

4.2. Depth Distribution of Different Isotopes

The main purpose of the metallic glass target was the determination of depth distributions to resolve noble gases implanted with different energies, especially those implanted with SW energies (0.5-4.0 keV/amu) from solar energetic particles (SEPs) with energies $>5$ keV/amu. For the final selection of the target composition, V.S. Heber (2002) already carried out tests of the etching behaviour and depth distribution of $^4$He and $^{20}$Ne in a metallic glass (Vitreloy 106) comparable to Ax1, though with a slightly different composition. These test samples were irradiated with $^4$He and $^{20}$Ne at energies of 10 keV/amu, therefore with a similar velocity for both isotopes. Subsequent CSSE experiments showed a homogeneous etching behaviour with depth, which favoured the metallic glass to other materials. To test whether CSSE would be

Fig. 3.4. Trapping efficiencies for $^4$He and $^{20}$Ne at SW energies in different targets as deduced from implantation experiments at the CASYMS compared to SRIM simulations.
III. Irradiation Experiments

capable to resolve a low-velocity from a high-velocity SW component and therefore also SW particles from SEPs, a modified implantation experiment was arranged. A single Ax1 aliquot was irradiated with $^{20}\text{Ne}$ at 12.7 keV and $^{22}\text{Ne}$ at 60.0 keV in a ratio of $\sim 11$, in order to gain a large variation of $^{20}\text{Ne}/^{22}\text{Ne}$ ratios with implantation depth. The implanted Ne isotopes were extracted by CSSE in 36 steps; the resulting $^{20}\text{Ne}/^{22}\text{Ne}$ ratios are plotted in Fig. 3.5.

<table>
<thead>
<tr>
<th>Table 3.2. Adopted backscatter correction factors ($1/\eta$) for He, Ne and Ar isotopes in the BMG.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Correction factor* ($1/\eta$)</td>
</tr>
<tr>
<td>------------------------------------------------------------</td>
</tr>
<tr>
<td>$^4\text{He}$</td>
</tr>
<tr>
<td>$^3\text{He}/^4\text{He}$</td>
</tr>
<tr>
<td>$^{20}\text{Ne}$</td>
</tr>
<tr>
<td>$^{20}\text{Ne}/^{22}\text{Ne}$</td>
</tr>
<tr>
<td>$^{21}\text{Ne}/^{22}\text{Ne}$</td>
</tr>
<tr>
<td>$^{36}\text{Ar}$</td>
</tr>
<tr>
<td>$^{36}\text{Ar}/^{38}\text{Ar}$</td>
</tr>
</tbody>
</table>

* For He deduced experimentally and modeled with SRIM for Ne and Ar.
The adopted uncertainties for backscatter correction are given as $2\sigma$ error: $^4\text{He}$: 3.5%, $^{20}\text{Ne}$: 4.8%, $^3\text{He}/^4\text{He}$: 5.1%, $^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$, and $^{36}\text{Ar}/^{38}\text{Ar}$: 1.2%, $^4\text{He}/^{20}\text{Ne}$ and $^{20}\text{Ne}/^{36}\text{Ar}$: 2.3%.

The pattern in Fig. 3.5 shows a constant decrease of measured $^{20}\text{Ne}/^{22}\text{Ne}$ ratios starting from values about 58 in shallow depth to ratios of 2.8 in the final step. This trend is somewhat in contrast to SRIM simulations for an irradiation with conditions to that used at the CASYMS. SRIM predicts a much higher mass separation of Ne isotopes in the first gas fraction released, with initial $^{20}\text{Ne}/^{22}\text{Ne}$ ratios of about 100. The most feasible explanation for this is a contribution of blank Ne incorporated in the Ax1 during manufacturing, for which the blank gas correction via the acid blank was not accounting for. This is suggested by the composition of Ne stored in between two etching days, which is always isotopically heavier than adjacent etching steps. Because the actual sample gas concentrations were very small for each step, the blank gas contributions had a significant effect on the resulting Ne composition and shifted the released pattern towards smaller $^{20}\text{Ne}/^{22}\text{Ne}$ ratios. However, the gradual release pattern demonstrates that the Ax1 etches very homogeneously with a high depth resolution, which finally allowed the separation of solar particles implanted with different velocities.

5. CONCLUSION

The irradiation experiments performed at the CASYMS facility in Bern were successful to a certain extent. The experimentally deduced trapping efficiencies are in good agreement with SRIM simulations. As will be discussed in the following chapters, the adopted backscatter correction factors for Ne and Ar lead to reasonable results and interpretations of the data obtained from the BMG flown on Genesis. In contrast, the He experiments showed larger deviations from the simulations. For Ni, Ag, and Au we observed a smaller trapping efficiency.
than predicted. Surprisingly, this was contrary for the BMG for that we measured significantly higher trapping efficiencies than simulated with SRIM. Since the measured values for the BMG are well reproducible we adopted the experimental values for the interpretation of the Genesis data, though the results are not as reliable as expected from Ne. However, as will be discussed in Chapter VI, the derived trapping efficiency for He in the BMG might underestimate backscattering as the bulk SW He fluences are 20-30% lower than measured in-situ or with other Genesis bulk targets. This might be a problem of the single implantation energy used in the irradiation experiments, which does not account for lower SW energies <0.83 keV/amu in space. F. Bühler (private communication) reports from significant differences between predictions of trapping efficiency, e.g. with SRIM, and irradiation experiments at low particle energies <50 eV carried out for the COLLISA experiment. Similar differences leading to underestimated backscatter losses could account for the assumed small fluence measured in the BMG.

The depth distribution analysed proved that CSSE with HNO$_3$ allows very homogeneous depth profiling in a metallic glass. The deviation between simulated and measured depth distribution is most likely an artefact of blank gas contributions that shifted the release pattern towards lower $^{20}$Ne/$^{22}$Ne than predicted by SRIM. Nevertheless, it turned out that CSSE of the BMG allowed for perfect depth profiling for He, Ne, and Ar isotopes to distinguish between solar particles implanted at different velocities.
III. Irradiation Experiments

Acknowledgements

At this point I want to specifically thank Adrian Etter for perfect technical support at the CASYMS in Bern and Fritz Bühler for so many scientific and personal discussions and help. In the end of the day, Fritz was the big brain behind the experiments run at the CASYMS!

References


III. Irradiation Experiments


IV. Solar Wind Neon
And the Lunar Noble Gas Record
Solar Wind Neon from Genesis: Implications for the Lunar Noble Gas Record

Ansgar Grimberg,1* Heinrich Baur,1 Peter Bochsler,2 Fritz Bühler,2 Donald S. Burnett,3 Charles C. Hays,1 Veronika S. Heber,1 Amy J. G. Jurewicz,5 Rainer Wieler1

1Isotope Geology and Mineral Resources, ETH Zürich NW, Clausiusstrasse 25, CH-8092 Zürich, Switzerland.
2Physikalisches Institut, University of Bern, Sidlerstrasse 5, CH-3012 Bern, Switzerland.
3Department of Geological and Planetary Sciences, Caltech, Pasadena, CA 91125, USA.
4Jet Propulsion Laboratory Caltech, Pasadena, CA 91109, USA.
5Arizona State University (ASU) Center for Meteorite Studies, ASU M/C 1404, Tempe, AZ 85287, USA.

*To whom correspondence should be addressed. Email: grimberg@erdw.ethz.ch

Abstract

Lunar soils have been thought to contain two solar noble gas components with distinct isotopic composition. One has been identified as implanted solar wind, the other as higher-energy solar particles. The latter was puzzling because its relative amounts were much too large compared with present-day fluxes, suggesting periodic, very high solar activity in the past. Here we show that the depth-dependent isotopic composition of neon in a metallic glass exposed on NASA’s Genesis mission agrees with the expected depth profile for solar wind neon with uniform isotopic composition. Our results strongly indicate that no extra high-energy component is required and that the solar neon isotope composition of lunar samples can be explained as implantation-fractionated solar wind.

Dust grains from both the lunar surface and from meteorites consisting of compacted dust from asteroidal surfaces contain noble gases with isotopic compositions close to those measured in the aluminum foils that trapped solar wind (SW) on the Moon during the Apollo missions (1). The SW ions are implanted into these grains during exposure on the lunar or asteroidal surface to depths of up to ~200 nm for SW velocities in the range of 300 to 800 km/s (2). Within this implantation zone, deeply sited solar noble gases are substantially enriched in heavy isotopes. Specifically, near the grain surface, 20Ne/22Ne ratios are ~13.8 {close to the SW value of 13.7 measured in the Apollo foils [(1) and references therein]}, whereas at greater depths, 20Ne/22Ne values of solar Ne appear to cluster around 11.2 in many samples (3, 4). This data pattern becomes particularly evident when stepwise in vacuo etching of lunar grains is performed, a technique that releases noble gases from progressively deeper layers (Fig. 1). Some fractionation has always been expected along a depth profile, because the normal SW carries all species at
roughly equal speeds, causing heavier isotopes to have somewhat higher energies and greater penetration depths \((5, 6)\). However, this effect will not produce a pronounced clustering near a composition of \(^{20}\text{Ne}/^{22}\text{Ne} = 11.2\) but rather a gradual enrichment of the heavier isotopes with depth (Fig. 1). Because of this fact and the fact that the heavy component in lunar dust was assumed to reside considerably deeper than the several-hundred-nanometer penetration depth of the fastest SW ions \((7)\), the heavy component was attributed to solar energetic particles (SEPs) from discrete high-energy events known from in situ measurements in space. Therefore, Ne with a \(^{20}\text{Ne}/^{22}\text{Ne}\) composition of 11.2 was labeled SEP-Ne \((3, 4, 8–10)\). Further support for this distinction was provided by in situ analyses done on the Interplanetary Monitoring Platform (IMP-8) and International Sun-Earth Explorer (ISEE-3) spacecraft of solar energetic particles in the mega–electron volt per atomic mass unit range \((11, 12)\), which indicated \(^{20}\text{Ne}/^{22}\text{Ne}\) ratios distinctly lower than the SW value. In the following discussion, SEP-Ne denotes this hypothetical second solar Ne component in lunar samples, in contrast to solar energetic particles actually measured in situ in space.

SEP-Ne in lunar samples has remained enigmatic. Its inferred abundance relative to the implanted SW is about 25 to 50\% \((3)\), which is several orders of magnitude higher than expected from long-term averages of present-day fluxes of solar energetic particles in space, even postulating that part of the surface-sited SW got lost \((10)\). Thus, the high SEP-Ne abundance suggested a periodic increase in solar activity in the past \((10, 13)\). Furthermore, more recent observations of solar energetic particles on the Advanced Composition Explorer (ACE)
spacecraft indicate a highly variable Ne isotopic composition, making it doubtful whether the long-term average solar energetic particle composition differs significantly from the SW value (14). Other explanations for the apparent large abundance of SEP-Ne in lunar grains have included a strong depletion of the SW component due to grain-surface sputtering (10) and a galactic (15) rather than a solar source for the SEP-Ne component; none of these explanations was completely satisfactory.

To investigate this problem, a special bulk metallic glass (BMG) target (16, 17) was exposed to solar particles for 27 months on NASA’s Genesis Discovery Mission (18). The BMG target lends itself to high-resolution depth profiling of the amount and isotopic composition of implanted solar noble gases by in vacuo online etching (16), because it can be etched very homogeneously in nitric acid (19). It is one of the few Genesis targets recovered essentially intact.

The depth distribution of SW Ne isotopes was analyzed in two BMG samples of 0.11 and 0.37 cm² in 9 and 30 steps, respectively. The Ne isotopic composition became progressively heavier...
with increasing depth (Fig. 2, fig. S1, and table S1), following a mass-dependent fractionation line. The first minor steps showed $^{20}\text{Ne}/^{22}\text{Ne}$ ratios around 16, which is distinctly higher than the bulk SW average. With progressive etching, $^{20}\text{Ne}/^{22}\text{Ne}$ ratios decreased, with a large fraction of the gas displaying ratios around the bulk SW value (Fig. 2). The last steps releasing sufficient gas for precise analyses displayed $^{20}\text{Ne}/^{22}\text{Ne}$ ratios similar to the SEP-Ne value observed in lunar samples; however, the final point was below the SEP-Ne value. The total Ne released stepwise from the BMG had an average bulk $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.85 ± 0.11, which is similar within uncertainties to the bulk SW average of 13.75 ± 0.05 as measured by total extraction in three additional BMG samples (Tables S1 and S2). Both values are slightly corrected for backscatter losses (16).

Although three of the four largest high-energy solar flare events producing solar energetic particles in solar activity cycle 23 fell into the exposure period of Genesis (20), the fluence of these particles was at least three orders of magnitude too small (13) to possibly account for the Ne with a SEP-Ne–like isotopic composition (abundance ratio of solar energetic particles/SW < 0.001). Furthermore, in contrast to the much longer–exposed lunar samples, the abundance ratio of deeply and near-surface implanted solar Ne in the metallic glass was unaffected by sputter-induced losses of surface layers. Therefore, the data pattern of the metallic glass in Fig. 2 does not require a mixture of two energetically and isotopically different solar components but rather is due to isotopic fractionation effects upon implantation.

This conclusion is corroborated by ion implantation modeling using the SRIM (stopping and range of ions in matter) code (2, 16). Apart from the elevated $^{20}\text{Ne}/^{22}\text{Ne}$ ratios measured in the first few minor steps, the modeled Ne isotopic profile matches the measured profile very well (Fig. 2). In particular, the SEP-Ne value of 11.2 is reached in the modeled profile at around 94 to 97% gas release, which is similar within uncertainties to the measured profile. Progressively lower $^{20}\text{Ne}/^{22}\text{Ne}$ ratios well below 11.2 are expected in the remaining 3%, although this would be difficult to measure. Therefore, a single SW-Ne component with a uniform isotopic composition over the entire velocity range suffices to explain the depth profile in the BMG.

The data pattern of the BMG is very similar to those of many lunar samples, with two major simplifications. First, Genesis samples do not contain detectable amounts of Ne produced by spallation by galactic cosmic ray particles (GCR-Ne), because no appreciable concentrations of GCR-Ne accumulated during the approximately 3 years in space. This fact is important because a seemingly strong argument for the reality of SEP-Ne had been that in many in vacuo etch runs (such as lunar plagioclase grain separates), the data points of the later etch fractions fall onto
a straight line in a Ne three-isotope diagram (Fig. 1). This line passes through the extrapolated GCR point on the lower right and the SEP-Ne point on the upper left, apparently indicating a constant isotopic composition of the SEP-Ne end member over a large depth range. Second, the major gas fractions released early in the BMG etch runs showed higher $^{20}\text{Ne}/^{22}\text{Ne}$ ratios around 14.3, with the first fraction being as high as 16. In contrast, in the first etching steps, many lunar samples reveal $^{20}\text{Ne}/^{22}\text{Ne}$ ratios essentially identical to the SW value of ~13.7 determined by the Apollo SW composition experiments. As discussed quantitatively below, the SW-like isotopic composition of gases released from very close to the surface of lunar grains can be explained by the sputtering of grain surfaces due to SW irradiation or mechanical erosion (5, 10, 21).

To account for the differences between Genesis and lunar exposure conditions, we extended the SRIM simulations (Fig. 3) by implementing surface sputtering, leading to sputter-saturation equilibrium, and GCR-Ne production in a material with plagioclase composition (22), with amounts adjusted to match the solar-Ne/GCR-Ne abundance ratio in lunar plagioclase grains (4, 16). At the grain surface, the simulated data points show the true composition of the impinging SW. This corroborates the earlier interpretation that the outermost layers of lunar samples may in fact conserve the true isotopic composition of the SW (5, 10). At greater depths, the simulated mixing curve essentially reproduces the linear trend displayed by many lunar samples. This similarity between measured lunar and simulated data is a very strong indication that the SEP-Ne inferred earlier from the lunar samples actually does not exist. Obviously, fractionated SW-Ne close to the SEP-Ne composition is just abundant enough to be detected in the presence of GCR-Ne, whereas more heavily fractionated SW-Ne at even greater depth becomes too rare to be recognized.

Although discussed repeatedly [as in (4, 6, 23)], it was difficult for previous authors to appreciate the dominant role of fractionation during implantation because of complications in the exposure history of lunar samples. Thus, the existence of an SEP-Ne component was widely accepted. The realization that SEP-Ne is not needed has become possible primarily thanks to the excellent properties of the BMG target when etched in vacuo, allowing high-resolution depth profiling of implanted noble gases, plus the opportunity to see pure SW-Ne, not mixed with GCR-Ne. Furthermore, the exposure occurred in coordination with excellent monitoring of the SW velocity distributions by in situ measurements with spaceborne instruments. Therefore, exposure conditions were very well constrained, in contrast to the lunar samples. The new explanation of the lunar Ne record has the virtue of not requiring strongly increased solar activity and thus higher solar energetic particle flux, or alternatively an enhanced flux of galactic particles (15), in the past. Similar implications can be expected for the other four noble
iv. solar wind neon and the lunar noble gas record

Gases and oxygen, for which substantial solar energetic particle–related contributions in lunar samples have been suggested (10, 24). It has been proposed by (25) that SEP-nitrogen in lunar ilmenites was isotopically light, opposite to Ne. The interpretation of these data needs to be revisited in light of the present work. Measured isotopic compositions of solar gases heavier than pure SW, rather than implying a prominent contribution of solar energetic particles, now indicate a relative enrichment of (isotopically heavy) fractionated SW from greater depths due to, for example, the loss of outermost grain layers. Examples of this are $^{20}\text{Ne}/^{22}\text{Ne}$ ratios as low as $\sim 11$ in interplanetary dust particles (6). One may also speculate that the “Ne B” component in Earth’s mantle proposed by (26, 27) is possibly a result of a contribution of fractionated implanted SW-Ne during terrestrial accretion.
References and Notes

16. Material, methods and data are available as supporting material on Science Online.

30. Carrying out the BMG experiment was made possible by the support of the entire Genesis Project at JPL, LANL and Lockheed Martin, with special thanks to the JPL Genesis Canister design team, headed by D. Sevilla and to the Genesis curation team at JSC. We also thank I. Leya for constructive comments and suggestions, O. J. Homan for plasma cleaning the BMG and S. Tosatti for non-destructive XPS surface analyses. This study was financially supported by the Swiss National Science Foundation and the Genesis Project.

Supporting Online Material

www.sciencemag.org/cgi/content/full/314/5802/1133/DC1

Materials and Methods

Fig. S1

Tables S1 and S2

References and Notes

received 7 August 2006; accepted 4 October 2006

Materials and Methods

Material: The collector material called Bulk Metallic Glass (BMG, Genesis target #40598) is an amorphous metal with a composition of Zr$_{58.5}$Nb$_{2.8}$Cu$_{15.6}$Ni$_{12.8}$Al$_{10.3}$ (atomic %). The nucleation of crystalline metal has been largely suppressed by rapidly quenching the liquid. Because there is no crystal lattice, fractionation and loss of trapped solar ions due to diffusion along preferred paths on crystal planes is reduced. Similarly, there are virtually no grain boundaries to provide fast-diffusion pathways. Therefore, the glass retains noble gases as neon well. Moreover, it etches very uniformly in HNO$_3$ and is therefore well suited for a stepwise gas release (SI).

Stepwise gas release: Neon was released by stepwise etching of the BMG surface with HNO$_3$ vapor. The closed system stepwise etching (CSSE) device in Zurich is directly connected to a noble gas mass spectrometer (SI). Sample and acid were stored in separate volumes under ultra high vacuum. Total amounts of $^{20}$Ne released with CSSE and an achieved depth resolution of ~5-10 nm/step from sample 40958-03-F and 40958-03-B/04-K (Table S1) are in perfect agreement with amounts from total extraction given in Table S2.
**Total extraction:** Concentration and isotopic composition of the total Ne trapped in the BMG was determined by melting three samples of 7.7-8.7 mm² each at ~1500 °C, well above the BMG melting point. The released gas was analyzed in the same mass spectrometer as mentioned above. The values for all three samples are given in Table S2.

**Modeling:** Implantation of solar wind ions into the BMG was simulated using the SRIM code (S2). For each isotope the implantation depths and the fraction of backscattered particles lost from the target have been determined for 1.8×10⁶ particles, assuming implantation of SW ions with a uniform ²⁰Ne/²²Ne ratio of 13.75 (see Table S2) over the entire SW energy range, which is supported by (S3), and a velocity distribution as measured with instruments onboard the advanced composition explorer (ACE) during the exposure time of Genesis (S4). The calculated backscattering correction for Ne into BMG is relatively high compared to a target like the Al-foils used in the Apollo experiment (S5), because the BMG consists to ~90% of heavy transition-metals. The adopted correction factors for the BMG are 1.014 for the ²⁰Ne/²²Ne ratio, 1.007 for the ²¹Ne/²²Ne ratio and 1.13 for the ²⁰Ne amount.

To adapt this model to lunar grains two additional processes had to be introduced to account for the exposure conditions on the lunar surface. The first is sputtering of grain surfaces due to SW irradiation together with ongoing SW implantation on the freshly exposed surface. This was simulated until sputter-saturation equilibrium of the Ne isotopic composition was reached (after ~150 nm of surface sputtering). The second process considered was the production of Ne by spallation due to galactic cosmic rays (GCR). The isotopic composition of GCR-Ne was calculated using the model of (S6) for plagioclase chemistry and an assumed average shielding of <5 cm. The concentration of GCR-Ne added was chosen to yield a GCR-Ne/SW-Ne abundance ratio in the BMG similar to that found in lunar grains. In combination both processes shift SRIM results to SW-like values for surface-sited neon and to higher ²¹Ne/²²Ne ratios in greater depth where SW ions are less abundant, leading to a slightly curved fractionation line that is hardly discernible from the straight line fit through the measured lunar data points for later steps.

Note that the ²¹Ne/²²Ne ratio of the GCR point in Fig. 3 is slightly higher than in Fig. 1. Possibly this is partly due to the fact that the model by (S6) does not perfectly reproduce the isotopic composition of GCR-Ne or that the assumed mean shielding of <5 cm is not correct. However, it seems more likely that the linear extrapolation of the data points as indicated in Fig. 1 yields a slightly too low ²¹Ne/²²Ne -GCR ratio, since the underlying assumption of a two-component mixing has proven in this paper to be wrong.
**Supporting Figures**

![Figure S1](image)

**Figure S1.** Ne three-isotope diagram from stepwise in-vacuo etching of Genesis bulk metallic glass sample (BMG). Data are for two etching-runs on the BMG (40958-03-F and 40958-03-B/04-K) exposed to the solar wind throughout the entire ~2.5 year Genesis exposure (Table S1). Data from the first steps (representing most superficially implanted gas) plot at $^{20}\text{Ne}/^{22}\text{Ne}$ ratios somewhat higher than the plotted SW value measured in Al foils during the Apollo lunar missions (S3). Large $2\sigma$-error bars for some steps indicate very small amounts of gas released. Data from more deeply implanted Ne released in subsequent steps are aligned along a mass-dependent fractionation line. This trend is quantitatively described by fractionation during ion implantation, as shown by SRIM (S2) simulations plotted as its $2\sigma$-envelope assuming implantation of a SW component with a velocity-independent $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.75±0.05 and $^{21}\text{Ne}/^{22}\text{Ne}$ ratio of 0.0330±0.0004 (average Table S2). The lowest measured $^{20}\text{Ne}/^{22}\text{Ne}$ ratios are similar to the SEP-Ne value reported from lunar samples (S7) plotted for reference. Overall the release pattern is similar to that from the lunar ilmenite (Fig. 1), apart from the fact that many lunar samples also release minor amounts of GCR-Ne, shifting especially the data points from later steps towards higher $^{21}\text{Ne}/^{22}\text{Ne}$ ratios.

**Supporting Tables**

<table>
<thead>
<tr>
<th>Etching step</th>
<th>$^{20}\text{Ne}/^{22}\text{Ne}$ (×10$^{-9}$)</th>
<th>$^{20}\text{Ne}/^{22}\text{Ne}$ (×10$^{-9}$)</th>
<th>$^{21}\text{Ne}/^{22}\text{Ne}$ (×10$^{-9}$)</th>
<th>$^{21}\text{Ne}/^{22}\text{Ne}$ (×10$^{-9}$)</th>
<th>$^{21}\text{Ne}$ (atoms/cm$^2$)</th>
<th>$^{21}\text{Ne}$ (atoms/cm$^2$)</th>
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### IV. Solar Wind Neon and the Lunar Noble Gas Record

Sample 40598-03-B/04-K

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<th>Etching step</th>
<th>$^{20}$Ne/$^{22}$Ne ± 2σ</th>
<th>$^{21}$Ne/$^{22}$Ne ± 2σ ($\times$10$^{-2}$)</th>
<th>$^{20}$Ne ($\times$10$^9$ atoms/cm$^2$) ± 2σ</th>
<th>$^{21}$Ne ($\times$10$^9$ atoms/cm$^2$) ± 2σ</th>
</tr>
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<tr>
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<td>1311 ± 33</td>
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**Table S1.** Ne concentrations and isotopic ratios measured by stepwise etching of samples 40598-03-F and 40598-03-B/04-K. The data shown for each single step and the respective total value are not corrected for losses due to backscattering. 2σ-errors include ion statistics, variations in sensitivity, interference corrections and uncertainties in the absolute calibration. Factors for the backscattering corrected total values are similar as described above. The average of values corrected for backscattering of both etching-runs is 13.85±0.11 for the $^{20}$Ne/$^{22}$Ne ratio, 0.0330±0.0004 for the $^{21}$Ne/$^{22}$Ne ratio and (1.31±0.03)×10$^{12}$ for the $^{20}$Ne amount.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{20}$Ne/$^{22}$Ne ± 2σ</th>
<th>$^{21}$Ne/$^{22}$Ne ± 2σ ($\times$10$^{-2}$)</th>
<th>$^{20}$Ne ($\times$10$^9$ atoms/cm$^2$) ± 2σ</th>
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<td>13.75 ± 0.05</td>
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</table>

**Table S2.** Ne concentrations and isotopic ratios measured by total extraction of samples 40598-03-C, -03-D and -03-E, not corrected for losses due to backscattering. 2σ-errors include ion statistics, variations in sensitivity, interference corrections and uncertainties in the absolute calibration. The 2σ-error for the average values is given as the 2σ error of the mean and factors for the backscattering corrected average are similar as described above.
S1. V. Heber, PhD thesis #14579, ETH Zurich (2002),
V. Composition of Light Solar Wind Noble Gases
Composition of Light Solar Wind Noble Gases in the Bulk Metallic Glass flown on the Genesis Mission

A. Grimberg1*, D.S. Burnett2, P. Bochsler3, H. Baur1, R. Wieler1

1 Isotope Geology, ETH Zürich, CH-8092 Zürich, Switzerland
2 Calif. Inst. of Technology, GPS MC 100-23, Pasadena, CA 91125, USA
3 Physikalisches Institut, University of Bern, CH-3012 Bern, Switzerland

* Author for correspondence, Email: grimberg@erdw.ethz.ch

Abstract: We discuss data of light noble gases from the solar wind implanted into a metallic glass target flown on the Genesis mission. Helium and neon isotopic compositions of the bulk solar wind trapped in this target during 887 days of exposure to the solar wind do not deviate significantly from the values in foils of the Apollo Solar Wind Composition experiments, which have been exposed for hours to days. In general, the depth profile of the Ne isotopic composition is similar to those often found in lunar soils, and essentially very well reproduced by ion-implantation modelling, adopting the measured velocity distribution of solar particles during the Genesis exposure and assuming a uniform isotopic composition of solar wind neon. The results confirm that contributions from high-energy particles to the solar wind fluence are negligible, which is consistent with in-situ observations. This makes the enigmatic “SEP-Ne” component, apparently present in lunar grains at relatively large depth, obsolete. 20Ne/22Ne ratios in gas trapped very near the metallic glass surface are up to 10% higher than predicted by ion implantation simulations. We attribute this superficially trapped gas to very low-speed, current-sheet-related solar wind, which has been fractionated in the corona due to inefficient Coulomb drag.

Keywords: Sun: solar wind, isotopic abundance ratios, Methods: laboratory

1. INTRODUCTION

To infer the isotopic and elemental composition of the solar photosphere from that of the solar wind (SW), it is essential to have precise information on the fractionation mechanisms operating at the source, i.e. the sun and its outer convective zone, and in the solar wind itself. This is one of the major goals of NASA’s Genesis mission (Burnett et al., 2003), which collected SW particles for more than 2 ½ years at the Lagrangian point L1 for high-precision analysis on earth. Three SW regime collector panels sampled the low-speed interstream wind, high-speed wind from coronal holes, and SW related to coronal mass ejections, respectively (Reisenfeld
et al., 2007). Additional collectors sampled all SW regimes, including high-energy particles, to provide compositional information about the bulk SW and potential variations as a function of energy.

In the following we present noble gas data from a bulk metallic glass (BMG) target (Jurewicz et al., 2003) exposed to the solar corpuscular radiation during the entire exposure period. The main purpose of this experiment was to determine the dependence of the isotopic and elemental composition of light noble gases on implantation depth in order to study possible variations in their isotopic composition as a function of solar particle energy. We compare the BMG data with those from the Apollo Solar Wind Composition (SWC) experiments (Geiss et al., 2004), in-situ measurements (Kallenbach et al., 1997a and 1997b, Kallenbach et al., 1998) and depth profiles of the Ne isotopic composition measured in lunar samples (Black and Pepin, 1969, Etique et al., 1981, Wieler et al., 1986, Benkert et al., 1993, Wieler, 1998).

Noble gases are among the key elements to be studied in Genesis targets, since their composition in the sun and the solar nebula cannot be deduced from meteorites, where they are heavily depleted. Moreover, light noble gases are relatively easy to measure in Genesis targets, and therefore are particularly well suited to study possible fractionation processes in different solar wind regimes. Precise measurements of He, Ne and Ar isotopes in the SW have already been obtained from the SWC-foils (Geiss et al., 2004). Their short exposure duration of two days at most restricts the interpretation of the obtained data to specific SW conditions, especially to low-speed solar wind. In-situ measurements, e.g. with instruments onboard the Advanced Composition Explorer (ACE) (Gloeckler et al., 1998, Mason et al., 1998, McComas et al., 1998, Smith et al., 1998) or the Solar and Heliospheric Observatory (SOHO/CELIAS) (Hovestadt et al., 1995), provide the full range of energy and regime-dependent data (Kallenbach et al., 1998, Wimmer-Schweingruber et al., 1998), but often suffer from lack of precision, which is necessary to constrain theories on isotopic fractionation in the SW. Noble gases measured in lunar regolith samples that were exposed to the SW for several million years include all SW regimes. The complicated sample history, variable losses of solar wind gases (Wieler et al., 1986, Wieler, 1998, Pepin et al., 1999, Pepin et al., 2000, Burnett et al., 2003) and a non-ideal behaviour during stepwise noble gas extraction make it difficult to deduce the bulk SW composition from such samples and to obtain reliable constraints on further parameters such as the dependence of trapped noble gas composition on implantation depth and thus particle energy.

The BMG on Genesis is well suited to deduce the composition of SW noble gases as a function of implantation depth, and hence, to possibly provide information on fractionation effects
depending on the implantation energy. On the one hand, this is because the BMG etches very homogeneously in nitric acid (Heber, 2002). A sufficient number of solar wind Ne ions were collected to analyse released gases in several tens of steps, yielding a high depth resolution. At the same time, the exposure period was short enough to prevent loss of near-surface-sited gas due to surface sputtering, as is often the case in natural (meteoritic or lunar) samples, and to minimise the production of spallogenic isotopes by galactic cosmic rays (GCR). On the other hand, the trapping behaviour for He and Ne irradiation in the metallic glass and other target materials was carefully tested experimentally (Grimberg et al., 2005) and simulated with the SRIM-code (Ziegler, 2004). Furthermore, precise input parameters on SW conditions during the Genesis exposure for SRIM modelling were provided by instruments onboard ACE and the Genesis spacecraft itself (Reisenfeld et al., 2007).

2. BULK SOLAR WIND HELIUM AND NEON DATA

Although the BMG was primarily designed to study the dependence of trapped noble gas composition on implantation depth, we also report data on the composition of the bulk solar wind trapped by this target during the maximum activity of solar cycle 23. Such data are important for comparison with other target materials on Genesis as well as with earlier SW collection experiments, e.g. the SWC foils or with gases trapped in lunar samples. The bulk BMG data (Table I) were obtained by total noble gas extraction via melting of the glass or as the sum of the closed-system etching steps described below. The measured bulk isotopic ratios of He and Ne from both methods are in good agreement. To compare these data with those obtained on other target materials and to deduce absolute solar wind fluxes, one has to consider that the BMG mainly consists of transition metals ($\text{Zr}_{58.5}\text{Nb}_{28.8}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3}$, subscripts in atomic-%) with a relatively high atomic number. Since backscattering of impinging ions, the relevant process controlling the trapping efficiency of a material, depends on the atomic masses, of both, the projectile and the target, care has to be taken when comparing results from different target materials. Backscatter losses from the BMG are larger than for targets consisting of lighter elements, hence the correction is more important for the heavy BMG target than for, e.g., the SWC Al-foils. Correction factors for He and Ne isotopes listed in Table I have been determined with implantation experiments at the CASYMS facility in Bern (Ghielmetti et al., 1983, Grimberg et al., 2005) and SRIM. For Ne, the experimental and the modelled value are consistent and we choose the SRIM value because of its smaller uncertainty. For He we adopted the CASMYS-deduced correction factor noting that SRIM simulations for the BMG seem to overestimate backscattering of very light elements in the SW energy range.
Table 1  Bulk isotopic and elemental composition from the Genesis BMG and from SWC foils reported by Geiss et al. (2004)

<table>
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<tr>
<th>Sample</th>
<th>^3He/^4He (×10^{-4}) ± 2σ (×10^{-4})</th>
<th>^20Ne/^22Ne ± 2σ</th>
<th>^4He/^20Ne ± 2σ</th>
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<td>466 20</td>
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This work; mean ratios of three samples for both, CSSE and total extraction

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<th>Sample</th>
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<th>^20Ne/^22Ne ± 2σ</th>
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Corrected He and Ne isotopic ratios in the BMG agree well with the results of the SWC foils reported by Geiss et al. (2004). We note that the bulk SW isotopic and elemental composition of He and Ne does not vary significantly between the daily scale recorded by the SWC experiments and the 2.5 year average recorded by the Genesis BMG target. The ratios of ^4He/^20Ne and ^4He/^3He measured in different SWC experiments correlate with each other (Geiss et al., 2004), which can be explained by fractionation caused by Coulomb drag, affecting both, the elemental and the isotopic ratio simultaneously (Geiss et al., 1970, Bodmer and Bochsler, 1998). The BMG data point falls right on the Coulomb drag fractionation line which is consistent with the SWC foil data (Bochsler, 2007). This also confirms that even the more mobile He has been quantitatively retained in the BMG despite the estimated temperatures of ~180°C during exposure.

3. DEPTH DEPENDENCE OF TRAPPED SOLAR Ne
COMPOSITION IN THE BMG

The mean penetration distance of ions into a target material scales with their implantation energy (Ziegler, 2004). Therefore, the depth-dependent distribution of SW isotopes and elements in a material contains information about the energy-dependence of the SW composition. This information is smeared out by two effects, however. On the one hand, particles of a given
implantation energy have a relatively wide depth distribution. Changes in SW velocity over a collection period result in an overlap of different depth distributions for a given particle. On the other hand, at a given moment, all SW species have similar velocities, causing heavier particles to have higher kinetic energies and thus greater penetration depths. This leads to mass fractionation upon implantation.

The depth-dependent isotopic composition of neon obtained by closed-system stepwise etching (CSSE) analyses (Heber, 2002, Grimberg et al., 2005) in three BMG samples is shown in Figure 1. In the first steps from very close to the surface, $^{20}$Ne/$^{22}$Ne-ratios are distinctly higher than the measured bulk SW average in the BMG of 13.75±0.19 (Table I) or any value reported from depth-dependent analyses of lunar grains (Black and Pepin, 1969, Etique et al., 1981, Wieler et al., 1986, Benkert et al., 1993). With progressive etching the $^{20}$Ne/$^{22}$Ne-ratio slowly decreases and the final etching steps, releasing deeply sited particles, display $^{20}$Ne/$^{22}$Ne-ratios as low as 10.5±0.3.

Figure 1. Depth-dependent neon isotopic composition of implanted SW derived from three Genesis BMG samples (white, grey and black boxes) by CSSE together with SRIM simulation data (light grey shaded area in between the two solid black lines displaying the 2σ uncertainty envelope including the statistical error and the error of the bulk measurement by total extraction, assuming a velocity-independent $^{20}$Ne/$^{22}$Ne ratio of 13.75±0.19), versus the cumulative $^{20}$Ne fraction. Length of boxes in horizontal direction indicates gas amounts released per etching step, vertical extension of boxes indicates 2σ uncertainties of $^{20}$Ne/$^{22}$Ne ratios including ion statistics, extraction blank variability, interferences and mass discrimination. Step 14 of sample 40598-04-E/-F/-G/-J had to be corrected for ~23% of Ne contribution from a gas inclusion in the BMG opened during the etching process. $^{20}$Ne/$^{22}$Ne ratios of all three BMG samples follow a trend that gets progressively heavier with depth. The implantation depth of the measured gas given on the upper abscissa is estimated from the simulated depth according to SRIM. Simulated and measured $^{20}$Ne/$^{22}$Ne profiles agree very well with each other. Thus the data pattern of the BMG can be explained by a fractionation of an isotopically uniform solar wind (SW = BMG bulk average from total extraction) upon implantation. Remarkably, ratios in the first 8% of the gas from very close to the surface show clearly higher $^{20}$Ne/$^{22}$Ne ratios than predicted by the SRIM simulations. This suggests this fraction to be very low-speed, current-sheet-related SW. The putative “SEP-Ne” data point is only for reference and should not be considered any longer.
The generally very good agreement of the measured depth distribution of Ne isotopes with SRIM predictions corroborates mass-dependent fractionation of SW ions upon implantation to be the responsible process for the observed Ne data pattern in the BMG (Grimberg et al., 2006). This process has been discussed earlier (Tamhane and Agrawal, 1979, Pepin et al., 2000, Mewaldt et al., 2001) but the solar Ne in mineral grains from lunar soils has widely been interpreted to be a mixture of two isotopically distinct components in the solar corpuscular radiation, SW-Ne near the surface, and “SEP-Ne” with a $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 11.2±0.2 (Etique et al., 1981, Wieler et al., 1986, Benkert et al., 1993) at larger depth. The main problem with this interpretation has always been the very high required fluence of solar energetic particles (Wieler, 1998, Mewaldt et al., 2001, Wimmer-Schweingruber and Bochsler, 2001) since in-situ analyses measured SEP/SW ratios <0.001, being orders of magnitude smaller than those reported from lunar samples (SEP-Ne/SW-Ne ~0.1-0.4). The BMG data now show unambiguously that the putative “SEP-Ne” component is not needed to explain the lunar soil data. The interpretation of the BMG data here is also fully consistent with the solar energetic particles fluences measured during the exposure period of Genesis with the ACE instruments SIS, CRIS and ULEIS (Mason et al., 1998, McComas et al., 1998, Stone, 1998a and 1998b). The integrated Ne fluence ($\pi$) of suprathermal ions in the range from 5.0 keV/nucleon to 1000 keV/nucleon is four orders of magnitude lower than the SW fluence (SEP/SW~0.0001) for the Genesis period (Mewaldt R.A., private communication). The corresponding Ne concentrations in the BMG are much too low to be resolved from the SW contribution. The data reported here show that fractionation upon implantation is capable to produce $^{20}\text{Ne}/^{22}\text{Ne}$ ratios as low and even lower than the putative “SEP-Ne” value formerly inferred from lunar data. The “SEP”-noble gas component is thus an artefact and should not be considered any longer.

4. ISOTOPIC FRACTIONATION OF THE SOLAR WIND

As discussed above, the depth distribution of noble gas isotopes also contains, in principle, information about the fractionation operating on the SW prior to implantation, though this is complicated by implantation-induced fractionation overprinting the original SW signature. However, the initial etch steps, representing the first 8% of the Ne from very close to the BMG surface (Figure 1), show $^{20}\text{Ne}/^{22}\text{Ne}$ ratios up to 10% higher than implied by SRIM simulations (Figure 2). We note that losses of superficially implanted neon by diffusion would produce the opposite trend. The same statement applies for the effect of a possible superficial contamination of the BMG with air neon.
The depletion of the heavy isotope relative to SW implanted with higher energies supports the model of isotopic fractionation due to inefficient Coulomb drag in low-speed, current-sheet-related solar wind (e.g. Wimmer-Schweingruber, 1994, Bodmer and Bochsler, 1998). The inefficiency of Coulomb drag in low-speed wind is corroborated by a coincident depletion of \(\alpha\)-particles represented by small He/H ratios <0.015 (cf. Geiss et al., 1970) at low SW velocities as measured with the Genesis ion monitor. Adopting the correlation of \(^4\text{He}/^{20}\text{Ne}\) and \(^{20}\text{Ne}/^{22}\text{Ne}\) observed with the Apollo Foil experiments the Coulomb drag model (Bochsler, 2007) predicts a fractionation of Ne towards a lighter isotopic composition in the low-speed SW relative to the SW average. Since the surface-near neon composition in the BMG and the Coulomb drag model are, in general, consistent we attribute the superficially trapped gas to very low-speed, current-sheet-related solar wind.

In greater depths (\(^{20}\text{Ne}\) fraction >0.1), however, the BMG data show a slightly less steep gradient towards heavier Ne composition than predicted by the SRIM model (Figure 1). This pattern might be explained by an enhancement of the light isotope with increasing SW energy relative to the bulk SW (Figure 2). However, this is in contrast to in-situ measurements from SOHO/CELIAS/MTOF reported by Kallenbach et al. (1997a, 1997b, and 1998), that display a
marginally significant enrichment of the heavy isotope by ~2% over the range of 350 km/s to 650 km/s, and it is also contradicted by any solar wind fractionation concept. The reproducibility of the three different CSSE measurements at a given implantation depth in turn limits the determination of SW fractionation prior to implantation. Hence, the significance of the trend found in greater depths (20Ne fraction >0.1) of the BMG needs further investigation; for instance some smearing of the isotopic distribution due to somewhat inhomogeneous etching cannot be ruled out at this point.

5. CONCLUSION

The isotopic composition of Ne as well as the He/Ne ratio of trapped solar wind in Genesis’ Bulk Metallic Glass (BMG) target is fully consistent with data from the previous Apollo Solar Wind Composition experiment (Geiss et al., 2004). This shows that the BMG target quantitatively trapped and retained light noble gases from the solar wind, which is also encouraging for other Genesis targets. Thanks to the homogeneous etching of the BMG, allowing high resolution depth profiling of the Ne isotopic composition, and the detailed monitoring of SW conditions during the Genesis exposure, we have been able to show that the observed depth distribution of Ne isotopes is, in general, consistent with fractionation of a SW with uniform isotopic composition upon implantation. Since the measured depth profile is also similar to the distribution of solar Ne in lunar soils, an important consequence of this finding is that the putative “SEP-Ne” component is obsolete. While our data are thus consistent with a uniform SW Ne isotopic composition over most of the SW energy range, isotopically very light Ne representing the most shallowly implanted gas indicates that very low-speed, current-sheet related SW is fractionated relative to the bulk SW composition due to inefficient Coulomb drag. Further measurements of the He and Ar isotopic depth distribution in this target have the potential to examine the dependence of the elemental composition on SW energy.

Acknowledgements

We would like to thank R. A. Mewaldt, G. M. Mason, C. M. S. Cohen, R. A. Leske and, M. E. Wiedenbeck for providing suprathermal fluence data from ACE, F. Bühler for valuable discussions, and S. Tosatti and O. J. Homan for XPS and plasma cleaning of the BMG. We also would like to thank the entire Genesis team. This work was supported by the Swiss National Science Foundation and NASA.
References


VI. Helium, Neon, and Argon
Isotopic and Elemental Composition
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**Solar Wind Helium, Neon, and Argon Isotopic and Elemental Composition: Data from the Metallic Glass flown on NASA’s Genesis Mission**

Ansgar Grimberg¹,*, Heinrich Baur¹, Fritz Bühler², Peter Bochsler², and Rainer Wieler¹

¹Isotope Geology, ETH Zürich, CH-8092 Zürich, Switzerland
²Physikalisches Institut, University of Bern, CH-3012 Bern, Switzerland

*Current address of the author for correspondence: Physikalisches Institut, University of Bern, CH-3012 Bern, Switzerland.
Email: grimberg@space.unibe.ch

**Abstract**

Solar wind (SW) helium, neon and argon trapped in a metallic glass (BMG) target flown on NASA’s Genesis mission were analyzed for their bulk composition and depth dependent distribution. The bulk isotopic and elemental composition for all three elements is in good agreement with the mean values observed in the Apollo Solar Wind Composition (SWC) experiment. Conversely, the He fluence derived from the BMG is relatively low compared to values reported from other Genesis bulk targets or in-situ measurements during the exposure period. SRIM implantation simulations using a uniform isotopic composition and the observed bulk velocity histogram during exposures reproduces the Ne and Ar isotopic variations with depth within the BMG in a way, which is generally consistent with observations. The similarity of the BMG release patterns with the depth-dependent distributions of trapped solar He, Ne, and Ar found in lunar and asteroidal regolith samples shows that also the solar noble-gas record of extraterrestrial samples can be explained by mass separation of implanted SW ions with depth. Consequently, we conclude that a second solar noble gas component in lunar samples, referred to as the “SEP” component, is not needed, and we confirm that contributions from high-energy particles to the solar wind fluence are minor, consistent with in-situ flux observations. **On the other hand, a small fraction of the total solar gas in the BMG released from shallow depths is markedly enriched in the light isotopes relative to predictions from implantation simulations with a uniform isotopic composition. Contributions from a neutral solar or interstellar component are too small to explain this shallow sited gas. We tentatively attribute this superficially implanted gas to low-speed, current-sheet related SW, which was fractionated in the corona due to inefficient Coulomb drag. This fractionation process could also explain relatively high Ne/Ar elemental ratios in the same initial gas fraction.**

1. **INTRODUCTION**

Solar noble gases are an important diagnostic to study the fractionation mechanisms acting on elements and isotopes in the solar wind (SW). Information about these mechanisms is
VI. Helium, Neon, and Argon Isotopic and Elemental Composition

essential to draw inferences on the isotopic and elemental composition of the solar photosphere from the SW record. The elemental and isotopic composition of the photosphere and the outer convective zone of the sun are believed to have remained largely unchanged over the entire solar lifespan relative to the composition of the early solar nebula. They are fundamental for the understanding of the geochemical evolution of the solar system, and they provide a benchmark for the composition of the galaxy. NASA’s Genesis mission (Burnett et al., 2003) followed the footsteps of the Apollo solar wind composition (SWC) experiment performed during the lunar landings (Geiss et al., 1972; 2004) by trapping SW ions in solid target materials in space and by returning them back to Earth for laboratory analysis.

The main solar wind fractionation processes are related to the first ionization potential (FIP) or first ionization time (FIT) of elements (Von Steiger and Geiss, 1989; Feldman, 1998; Von Steiger et al., 2000) and inefficient Coulomb drag, respectively (cf. Geiss et al. 1970, Bodmer and Bochsler, 1998). The FIP/FIT effect leads to an enrichment of elements with first ionization potentials <10 eV but is not expected to sensibly affect the isotopic composition of elements. Conversely, inefficient Coulomb drag and gravitational settling in the corona influence both the elemental and the isotopic composition and depend on the SW flux in the inner corona (Geiss et al., 1970; Bürgi 1992, Bodmer and Bochsler, 1998). With Genesis samples two different approaches can be used to study the energy dependence of fractionation effects in the SW. The first are SW regime collector panels that separately sampled the low-speed interstream wind, high-speed wind from coronal holes, and SW related to coronal mass ejections, respectively (Reisenfeld et al., 2007). The second approach is to study the energy dependence of SW particles by depth-dependent analyses of implanted ions trapped in a target. Both approaches have their advantages and disadvantages, as discussed below. In this work we study the depth dependence of elemental and isotopic composition of solar He, Ne, and Ar in a special purpose target from Genesis.

SW noble gases have already been studied by several means. Precise He, Ne, and Ar measurements were obtained by the SWC foil experiments (Geiss et al., 2004) with a good time resolution. However, unfortunately no information is available on the He/H ratio in the solar wind during the exposures. Also, because of the solar conditions prevailing during exposures, the SWC foils sampled only low-speed SW. Noble gases measured in lunar regolith samples that were exposed to the SW for several million years include all SW regimes (Eberhardt et al., 1970; Pepin et al., 1970; Reynolds et al., 1970; Wieler et al., 1986; Wieler, 1998). The complicated sample histories, variable losses of solar wind gases, and a non-ideal behavior during stepwise noble gas extraction make it difficult to obtain reliable constraints on the
dependence of the trapped noble gas composition on implantation depth and thus on particle energy. In-situ data from ACE, Ulysses, WIND, and SOHO missions (Wiens et al., 2004 and refs. therein) provide the full energy- and regime-dependent data, but often suffer from lack of the required precision.

In this work we report data of He, Ne, and Ar in a bulk metallic glass (BMG) target (Hays et al., 2001; Jurewicz et al., 2003). The BMG was especially developed for analysis by closed system stepwise etching (CSSE) to provide information on the depth distribution of implanted ions with a very high depth resolution (Signer et al., 1993). CSSE releases gases at room temperature in order to minimize fractionation of trapped atoms due to diffusion during the gas release. This is an advantage compared to stepwise heating experiments and thermal ablation techniques. The BMG was exposed to the solar radiation for the entire exposure period of Genesis not distinguishing different SW regimes, which complements collection in other targets on Genesis that alternately sampled distinct SW regimes (Neugebauer et al., 2003).

The elemental and isotopic composition of implanted noble gases may vary with depth in the target as a result of the implantation process or because of an energy-dependent variability of the composition of impinging solar ions. We will use the term “mass separation” for variations ascribed to implantation effects and will reserve the term “fractionation” for processes thought to reflect a real variability of the SW composition. A tool to distinguish between “mass separation” and “mass fractionation” is to compare measured depth profiles of the noble gas composition with profiles simulated by the ion implantation code SRIM, Stopping and Range of Ions in Matter, by Ziegler (2004). We will also briefly report on comparisons of SRIM predictions with artificial irradiations of He and Ne into various targets materials (Grimberg et al., 2005; Grimberg, 2007).

The results of this paper represent the first complete bulk and depth-dependent data set of He, Ne, and Ar measured in one single Genesis target. Our data will be compared to results from lunar and meteoritic samples, results from the SWC foil experiment, and from in-situ measurements and thus can serve as a benchmark for all other elements to be studied in Genesis samples and their interpretation with respect to the composition of the Sun.

2. SAMPLE

We studied a bulk metallic glass (BMG) target (Jurewicz et al., 2003) that was exposed to the
solar corpuscular radiation during the entire Genesis exposure period. The glass mainly consists of transition metals with relatively high atomic numbers (Zr\textsubscript{58.5}Nb\textsubscript{2.8}Cu\textsubscript{15.6}Ni\textsubscript{12.8}Al\textsubscript{10.3}, subscripts in atom percent). The BMG was custom-made at the California Institute of Technology (Hays et al., 2001). It was chosen because it can be etched homogeneously in nitric acid, allowing thus a high depth resolution of the composition of implanted noble gases. Furthermore, the BMG was expected to be very retentive for implanted light noble gases. The final composition of the BMG was selected after etching tests by Heber (2002). To prevent oxidation during manufacturing, the BMG was casted in an Ar atmosphere. This lead to substantial atmospheric Ar blank contributions, compromising some of the Ar analyses, as will be discussed later.

During exposure in space a molecular film mainly consisting of Si, O, C, and F formed on many targets, including the BMG (Allton et al., 2006). One source of this film (informally labelled as “Brown Stain”) appears to be evaporated and recondensed glue, which had been used to fix the three screws with which the BMG was mounted. Traces of this glue were found on the edges of the BMG’s screw holes. To test this hypothesis and to assess whether the molecular film may have reduced the trapping probability of SW ions in the BMG we estimated its thickness by X-ray photoelectron spectroscopy (XPS). As shown in Fig. 1, we measured ratios of Zr/Si and Zr/C (in arbitrary units) along six profiles, three near the outer edge of the BMG (represented by the large circle), shown in the left panel, and three between the mounting holes (black circles) and the center of the BMG, right panel. Small ratios indicate relatively large contamination. All profiles show measurable amounts of Zr, indicating a maximum contamination thickness of 8 nm. The contamination is concentrated around the mounting holes, suggesting glue to fix the screws as the contamination source.

Fig. 1. XPS analyses of Zr, stemming from the BMG, and Si and C from the molecular contamination. Shown are Zr/Si and Zr/C ratios (in arbitrary units) along six profiles, three near the outer edge of the BMG (represented by the large circle), shown in the left panel, and three between the mounting holes (black circles) and the center of the BMG, right panel. Small ratios indicate relatively large contamination. All profiles show measurable amounts of Zr, indicating a maximum contamination thickness of 8 nm. The contamination is concentrated around the mounting holes, suggesting glue to fix the screws as the contamination source.
that it was caused by glue condensates. Nevertheless, the Zr signal is visible everywhere along the six profiles. Hence, the film thickness nowhere exceeds about 8 nm, the maximum depth of detection of the XPS system used. According to implantation simulations using the SRIM code (Ziegler, 2004), only a very small fraction of SW ions was stopped in the contamination. The corresponding reduction of the gas amounts trapped in the BMG is smaller than the statistical uncertainties of our mass spectrometric analyses. We will therefore not correct for effects caused by the molecular film. The samples analyzed in this work all were taken at least 1 cm away from screw holes.

Though the trapping of ions was not substantially affected by the molecular film, it caused another problem. Because it was polymerized by solar UV radiation it became difficult to remove. In particular, the film is resistant to nitric acid, the solvent used for stepwise etching of the BMG. Therefore a cleaning procedure to gently remove the film without affecting the BMG surface had to be developed. A first attempt using plasma ashing with pure oxygen as the reactive species was not successful as subsequent stepwise etching of a BMG aliquot treated by this procedure did hardly release any noble gases. We therefore tried a two-step procedure. Carbon originating from the contamination was first oxidized by gentle oxygen plasma ashing, followed by a plasma etching procedure with SF$_6$ gas as the reactant to transform Si-compounds into volatile SiF$_4$. The procedure was accompanied by sputtering with SiF$_6$ ions additionally removing the molecular film. This procedure was entirely successful. Samples cleaned this way could be analysed by CSSE and the recovered total gas amounts were always very close to the values obtained from complete pyrolysis. This confirms an efficient removal of the contaminating film without any substantial loss of SW gases from the BMG near-surface layers.

3. EXPERIMENTAL PROCEDURE

3.1. Pyrolytic Analysis

To determine the bulk SW composition of the trapped He, Ne, and Ar, three BMG aliquots of 0.08-0.09 cm$^2$ were extracted in a Mo crucible at ~1500 °C, well above the BMG melting temperature of 830 °C.

3.2. Closed System Etching Technique

To determine the depth distribution of He, Ne, and Ar isotopes in the BMG, the main
VI. HELIUM, NEON, AND ARGON ISOTOPIC AND ELEMENTAL COMPOSITION

experiment reported here, we used the CSSE line whose acid-exposed parts consist of Au or Pt only (Busemann et al., 2000). This line has been widely used to analyse SW noble gases in lunar samples and gas-rich meteorites (e.g. Heber et al., 2003), primordial noble gases in meteorites (Busemann et al., 2000) and interstellar He trapped in a BeO layer on BeCu-foils in the COLLISA experiment (Busemann et al., 2006). Sample and acid containers were held at 23 °C during etching steps, while the sample was exposed to nitric acid vapour for some prescribed time. At the end of each step, before the gas was expanded into the cleaning line, the partial pressure of the nitric acid was reduced by cooling the acid container for 65 min to -90 °C. Compared to previous studies we used a larger sample container to allow horizontal storage of the BMG samples, reducing the possibility of inhomogeneous etching. The gases of three aliquots, 0.11-0.51 cm$^2$ in size, were released in 12, 30, and 24 steps respectively, with etching steps lasting from hours to several days.

For both extraction methods the released gases were exposed to a series of getters in the stainless steel part of the line to remove interfering and chemically reactive gas species: first CaO, then TiZr, and finally AlZr (SAES®). The released Ar was separated from He and Ne with activated charcoal held at -196 °C. Subsequent mass spectrometric analyses of helium, neon, and argon were carried out according to procedures described by Graf et al. (1990) and Heber (2002).

3.3. Blanks

Blank corrections for pyrolysis were < 2% for Ne and always negligible for He. Analyses of bulk SW Ar were compromised due to high blank Ar contributions from the furnace and especially the BMG itself. Therefore, no precise bulk SW-Ar values have been obtained by pyrolysis. The He and Ne blanks in the CSSE runs mainly stem from the nitric acid. They depend on step duration but do not scale linearly with time. On the other hand, blank Ar contributions in etching runs mostly stem from Ar contained in the BMG itself. Blank values for a typical etch step of ~20 hours are as follows (in 10$^{-12}$ cm$^3$STP): $^4$He = 3, $^{20}$Ne = 2 and $^{36}$Ar = 5, with the Ne blank showing atmospheric composition. These blank contributions were negligible for all helium and neon measurements. However, Ar blanks were substantial, mainly due to the fact that the BMG had been manufactured in an Ar atmosphere. Fortunately, $^{36,38}$Ar blanks could be corrected for by assuming $^{40}$Ar to be entirely atmospheric, which is reasonable since $^{40}$Ar does essentially not exist in the SW. Nevertheless, this correction may be somewhat problematic when it becomes very large, because the atmospheric Ar in the BMG might actually be slightly enriched towards heavier isotopes in its composition. This is indeed suggested by the first
etching step releasing negligible amounts of solar Ar ($^{40}$Ar/$^{36}$Ar = 290), with an $^{36}$Ar/$^{38}$Ar ratio of about 5.27, slightly lower than the atmospheric value of 5.30 (Lee et al., 2006). Blank gas contributions from the BMG itself were significant for each step, indicated by the $^{40}$Ar amount, which is always 2-10 times higher than the $^{40}$Ar amount in a typical acid blank. Amounts of $^{40}$Ar correlate only weakly with etching time, showing that Ar incorporated during the manufacturing process is inhomogeneously distributed within the BMG. The measured $^{40}$Ar/$^{36}$Ar ratio is close to atmospheric for the first etching steps and never drops to values below 49.

All Ne data were corrected for interferences from $^{40}$Ar$^{++}$ and CO$_2^{++}$ and blank Ne. However, these corrections were negligible in all steps. Due to the low electron voltage in the ion source of only 45 V, doubly ionised species were hardly formed.

### 3.4. Uncertainties

Stated uncertainties (2σ) in the data tables include ion statistics, extraction blank uncertainty, interferences, mass discrimination and the uncertainty of backscatter correction. He, Ne and Ar isotopic ratios of calibration analyses are reproducible to within 0.35% and elemental ratios to within 0.75%. For elemental ratios, the calibration of absolute amounts was the most relevant source of uncertainty. The errors of absolute amounts are estimated to be about ~3%. For bulk analyses with pyrolysis He and Ne isotopic ratios are reproducible to within 2%, elemental ratios to within 7%, and element concentrations to within 12%.

<table>
<thead>
<tr>
<th>Target</th>
<th>$^3$He</th>
<th>$^4$He</th>
<th>$^{20}$Ne</th>
<th>$^{21}$Ne</th>
<th>$^{22}$Ne</th>
<th>$^{36}$Ar</th>
<th>$^{38}$Ar</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al$^a$</td>
<td>0.87±0.02</td>
<td>0.89±0.02</td>
<td>1.00±0.02</td>
<td>1.00</td>
<td>1.00±0.02</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>SRIM Al$^b$</td>
<td>0.899</td>
<td>0.919</td>
<td>0.992</td>
<td>0.993</td>
<td>0.994</td>
<td>1.000</td>
<td>1.000</td>
</tr>
<tr>
<td>CASYMS BMG$^c$</td>
<td>0.81±0.02</td>
<td>0.85±0.03</td>
<td>0.89±0.04</td>
<td>-</td>
<td>0.90±0.05</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>SRIM BMG$^b$</td>
<td>0.760</td>
<td>0.775</td>
<td>0.885</td>
<td>0.892</td>
<td>0.898</td>
<td>0.948</td>
<td>0.952</td>
</tr>
<tr>
<td>SRIM BMG$^d$</td>
<td>0.785</td>
<td>0.801</td>
<td>0.895</td>
<td>0.901</td>
<td>0.907</td>
<td>0.954</td>
<td>0.958</td>
</tr>
</tbody>
</table>

$^a$ Experimental He and Ne trapping efficiencies adopted from Meister (1969), given with the 2σ uncertainty of the mean for He and Ne isotopes from the CASYMS irradiation results for 6 Al samples.

$^b$ SRIM values modeled for the implantation energy of 0.83 keV/nucleon.

$^c$ Experimental He and Ne trapping efficiencies determined at CASYMS with 0.83 keV/nucleon for 3 BMG samples, normalized to Al. Values are given with the 2σ uncertainty of the mean.

$^d$ SRIM values modeled over the entire energy range of the SW velocity distribution from 0.3 to 4.0 keV/nucleon as measured for the Genesis exposure period. Trapping efficiencies adopted for the BMG are plotted in bold. Uncertainties for values modeled with SRIM were estimated from uncertainties of the CASYMS experiments.
3.5. Artificial Irradiations

To study the relative trapping efficiencies of different target materials for noble gas isotopes, we carried out artificial irradiation experiments at the CASYMS facility of the University of Bern (Ghielmetti et al., 1983; Steinacher et al., 1995; Grimberg et al., 2005). He and Ne were implanted into flight-like BMG targets and Al-foils from the same batch as the foils exposed during the Apollo SWC experiments. The He and Ne ions were accelerated to an energy of 0.83 keV/amu, corresponding to a SW speed of ~400 km/s. The results are given in Table 1.

The trapping efficiency is controlled by backscattering of impinging ions, which depends on the atomic masses of both the projectile and the target. Backscatter losses from the BMG are higher than from targets consisting of lighter elements; hence the CASYMS calibration shows that the correction is somewhat more important for the BMG than for an Al target, e.g., as used for the SWC experiments. In order to remain consistent with the results of the Apollo foils, we do not use absolute trapping efficiencies as derived from a simultaneous measurement of the total ion fluence in the CASYMS calibration chamber and an absolute determination of the resulting noble gas contents in the target. Rather, we determined correction factors for all He and Ne isotopes implanted into the BMG at 0.83 keV/amu by comparing them with the result of simultaneously irradiated witness pieces of unflown Apollo-type Al-foils. We then normalized our results to the absolute trapping efficiencies determined for the Apollo foils by Meister (1969). Thus we use Al foils as standards for the fluence calibration at the CASYMS and, hence, the trapping efficiencies of BMG targets.

Furthermore, the irradiation experiments were important to check the validity of SRIM irradiation simulations in the keV/amu energy range. These simulations are used to determine the trapping efficiency for all Genesis targets and SW species. Thus we modelled the implantation and trapping of SW He and Ne isotopes in the BMG and Al adopting the irradiation parameters used in the CASYMS experiments. Additionally, we modelled the trapping efficiency for He, Ne, and Ar isotopes in the SW energy range (0.3-4.0 keV/amu).

For Ne isotopes implanted into the BMG, experimental and SRIM-modelled trapping efficiencies were consistent with each other. For Ar SRIM factors were adopted as well because SRIM predictions become more reliable with increasing total implantation energy, i.e. with higher projectile mass. Conversely, for He we used the CASMYS-deduced correction factors, noting that SRIM simulations for the BMG seem to overestimate the backscattering of He isotopes in the SW energy range compared to the CASYMS derived values. All correction
factors are given in Table 1 and the adopted uncertainties given in the footnote of Table 2.

### 3.6 SRIM Simulation of Depth Distributions of SW Atoms

We modelled the depth distribution of implanted noble gas ions with the SRIM code. For this we adopted the SW velocity histogram for He, Ne, and Ar as obtained with instruments onboard the Advanced Composition Explorer (ACE) for the exposure period of Genesis (Reisenfeld et al., 2007) and the average bulk SW composition of the BMG measured via pyrolysis for He and Ne, and via CSSE for Ar. The SRIM simulations predict a mass separation of SW ions towards an enrichment of the heavier isotope with increasing depth. Since correction factors for backscattering of He modelled with SRIM differ significantly from experimental values, we consider the modelled depth distribution of He to be questionable as well. All SRIM depth distributions were calculated with a depth resolution of 2.5 nm and are plotted in the respective figure as the 2σ uncertainty envelope that includes the statistical error.

### 4. RESULTS

The noble gas results of three BMG samples analysed by pyrolysis and the three CSSE runs are given in Table 2 and Table 3a-3c, respectively. All BMG data given in the manuscript are measured values, i.e. not corrected for backscatter losses, unless noted otherwise. In the following we adopt the average He and Ne composition determined by total extraction as the bulk BMG values, as the summation of amounts over all CSSE steps leads to a higher uncertainty. The He, Ne, and Ar average bulk composition values obtained from the BMG are given in each figure as open diamonds labelled SW.

#### 4.1. Isotopic Composition

##### 4.1.1. Neon

The average neon isotopic composition of all total extractions and the weighted average of the three CSSE runs is given in Table 2. Also given are backscatter-loss corrected values for all data sets. The corrected bulk SW $^{20}$Ne/$^{22}$Ne and $^{21}$Ne/$^{22}$Ne ratios deduced by both methods agree well with each other (13.75±0.19, 0.0330±0.0009 for pyrolysis and 13.81±0.24, 0.0330±0.0004 for CSSE) and are also in agreement with the average $^{20}$Ne/$^{22}$Ne and $^{21}$Ne/$^{22}$Ne ratios of the bulk SW determined with the SWC foils (Table 2, (Geiss et al., 2004). However, the values are
slightly lower than the backscatter-corrected bulk ratios given by Mabry et al. (2007) measured in aluminium on sapphire. The fluence of $^{20}$Ne released via CSSE agrees within 2.5% with the total extraction average of $(1.14\pm0.13)\times10^{12}$ atoms/cm$^2$ and the resulting flux in turn is equal to the $^{20}$Ne flux obtained from a bulk SW carbon target exposed on Genesis (V.S. Heber, private communication), but is 15% lower than that reported by Mabry et al. (2007).

The $^{20}$Ne/$^{22}$Ne ratios of CSSE steps monotonously decrease from values around 15.5, well above the bulk SW ratio, to values around 11 (Figs. 2, 3). Fig. 2 also shows a similar trend for $^{21}$Ne/$^{22}$Ne ratios. They decrease as well with increasing depth of etching, with all data points plotting around a mass fractionation line (a “mass separation line” in the terminology used here) passing through the bulk SW point. Uncertainties of first and last steps are relatively large because of very small amounts of implanted gases. Step 14 of sample 40598-04-E/-F/-G/-J released unexpectedly large amounts of Ar and to a lesser extent also Ne, but normal amounts of He with a composition only slightly different from that of the adjacent steps. Almost certainly, the etching process opened a Ne and Ar containing gas inclusion produced during the manufacturing of the BMG. Such inclusions were observed previously in a test BMG studied by Heber (2002). We corrected the Ne amount of this step by ~42%, estimated via the $^4$He/$^{20}$Ne ratios of the adjacent steps 13 and 15. Over almost the entire depth range the measured Ne isotopic composition agrees well with SRIM simulation results assuming implantation of SW-
Ne with uniform isotopic composition (using the bulk SW composition deduced from total extraction analyses). One minor difference between measured and simulated data is that the measured $^{20}\text{Ne}/^{22}\text{Ne}$ ratios decrease slightly less steeply with depth than the simulated values. A more important difference is that the first few percent of released gas show clearly higher $^{20}\text{Ne}/^{22}\text{Ne}$ ratios than predicted by SRIM. For this fraction, a superficial contamination with atmospheric Ne can be excluded as this would enrich the heavy isotope and thus lower the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio. We also simulated the effects of potential diffusive losses of implanted Ne. The resulting Ne isotopic composition at the target surface would remain essentially unchanged or be even heavier than the starting composition at the surface. Similarly, a preferential stopping of light isotopes in the most superficial layers to produce a gradient much steeper than predicted by the SRIM simulation seems also unlikely. A simulation of the depth distribution with SRIM, which also accounts for the temperature distribution of SW ions for each velocity bin, confirmed the depth distributions calculated here (C. Olinger, personal communication).

Apart from the isotopically light surface-near gas fraction, similar dependencies of the isotopic composition of solar Ne on implantation depth have been observed earlier in numerous stepwise
etching experiments on lunar plagioclase and ilmenite separates (Etique et al., 1981; Wieler et al., 1986; Benkert et al., 1993; Wieler, 1998; Heber et al., 2003) and - less distinctly - in stepwise combustion and pyrolysis experiments of lunar soils and meteorites (Black and Pepin, 1969; Black, 1972; Becker and Pepin, 1994). These data patterns differ from those reported here in two aspects only. First, in the first gas release fractions the lunar and meteoritic data show lower $^{20}$Ne/$^{22}$Ne ratios than the BMG, sometimes close to SW values. Second, the lunar and meteorite samples also contain cosmogenic Ne such that the data paths defined by the latest gas release fractions point towards $^{20}$Ne/$^{22}$Ne and $^{21}$Ne/$^{22}$Ne ratios close to ~1 in a Ne three isotope plot.

4.1.2. Helium

The bulk isotopic compositions of SW-He deduced from both extraction methods are in very good agreement with each other, as already observed for Ne. We determined a bulk $^3$He/$^4$He ratio of $(4.41\pm 0.23)\times 10^{-4}$ as the average value of the three pyrolysis experiments and $(4.44\pm 0.26)\times 10^{-4}$ as the CSSE average, respectively (values corrected for backscatter losses). Our values are also in excellent agreement with the average SWC $^3$He/$^4$He ratio of $(4.26\pm 0.22)\times 10^{-4}$ (Geiss et al., 2004). The total amount of gas released by CSSE is 5.4 % lower than that released by pyrolysis, but both values agree within uncertainties. The resulting backscatter-corrected bulk $^4$He/$^20$Ne ratio of 527±39 also compares well with the average value of 570±70 reported for the SWC foils. However, the resulting $^4$He flux derived from the BMG by total extraction of $(9.13\pm 0.75)\times 10^6$ atoms/cm$^2$s (backscatter-loss corrected) is 21% lower than that obtained

![Approximate implantation depth of $^{20}$Ne according to SRIM (nm)](image-url)

**Fig. 4. Same as Fig. 3 for the ratio $^3$He/$^4$He.** SRIM simulations assume a velocity-independent bulk $^3$He/$^4$He ratio of $(4.41\pm 0.23)\times 10^{-4}$. The distribution simulated by SRIM strongly overestimates the mass separation of He isotopes with depth. The pattern of measured data resembles those observed in lunar ilmenites, showing a small variation of ratios with depth only. Similar to Ne, the first gas fraction shows enhanced ratios relative to subsequent steps.
VI. Helium, Neon, and Argon Isotopic and Elemental Composition

Table 2. Total He, Ne, and Ar in the BMG and the SWC foils

<table>
<thead>
<tr>
<th>Sample</th>
<th>⁴He Flux a (×10⁶)</th>
<th>³⁴He/⁴He (×10⁻⁶)</th>
<th>²⁰Ne b (×10¹¹)</th>
<th>²⁰Ne/²²Ne</th>
<th>³⁶Ar/³⁸Ar</th>
<th>⁴He/²⁰Ne</th>
<th>²⁰Ne/³⁶Ar (×10⁻⁵)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMG</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CSSE</td>
<td>7.36±0.23</td>
<td>4.34±0.12</td>
<td>11.6±0.1</td>
<td>13.61±0.17</td>
<td>3.27±0.02</td>
<td>466±19</td>
<td>5.28±0.23</td>
</tr>
<tr>
<td>Total extraction</td>
<td>7.76±0.58</td>
<td>4.31±0.02</td>
<td>11.4±1.3</td>
<td>13.56±0.09</td>
<td>3.28±0.06</td>
<td>504±36</td>
<td>-</td>
</tr>
<tr>
<td>CSSE, corrected c</td>
<td>8.66±0.40</td>
<td>4.44±0.26</td>
<td>13.1±0.6</td>
<td>13.81±0.24</td>
<td>3.30±0.04</td>
<td>488±23</td>
<td>5.30±0.24</td>
</tr>
<tr>
<td>Total extraction, corrected c</td>
<td>9.13±0.75</td>
<td>4.41±0.23</td>
<td>12.8±1.6</td>
<td>13.75±0.19</td>
<td>3.30±0.09</td>
<td>527±39</td>
<td>-</td>
</tr>
<tr>
<td>SWC Foils d</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Time weighted average</td>
<td>12.0</td>
<td>4.26±0.22</td>
<td>-</td>
<td>13.7±0.3</td>
<td>3.33±0.33</td>
<td>570±70</td>
<td>5.4±0.3</td>
</tr>
</tbody>
</table>

a The ⁴He flux is given in atoms/cm²s, calculated for an exposure duration for the BMG of 852.83 days.
b Gas concentration of Ne in (atoms/cm²).
c Corrected for backscatter losses (see text).
d Geiss et al. (2004).

He and Ne BMG data are mean values of three samples each for total extraction and CSSE. Ar data are values from one CSSE run. Uncertainties are given as 2σ error. The adopted uncertainties for backscatter correction are: ⁴He: 3.5%, ²⁰Ne: 4.8%, ³⁴He/⁴He: 5.1%, ²⁰Ne/²²Ne, ²¹Ne/²²Ne, and ³⁶Ar/³⁸Ar: 1.2%, ⁴He/²⁰Ne and ²⁰Ne/³⁶Ar: 2.3%.

from the bulk SW carbon target exposed on Genesis (V.S. Heber, private communication) and about 30% lower than the flux derived with ACE instruments of (13.0±2.6)×10⁶ atoms/cm²s for the exposure period of Genesis (Reisenfeld et al., 2006). This might suggest a substantial He deficit in the BMG, although the ACE data have a large uncertainty of 20%. Correspondingly, preliminary results from the bulk SW carbon target indicate a high backscatter-corrected ³He/²⁰Ne ratio of 673 (V.S. Heber, private communication), which is also substantially above the Apollo foil value.

The ³He/⁴He ratios are shown as a function of the cumulative ²⁰Ne fraction in Fig. 4. The data patterns of all three runs agree well with each other. Similar to the ²⁰Ne/²²Ne ratios, ³He/⁴He ratios in the first ~10% of the gas released are considerably higher than in the subsequent steps. The peak value of (5.37±0.06)×10⁻⁴ at 3.5% of ²⁰Ne released is also substantially above the backscatter-loss corrected bulk value of (4.41±0.23)×10⁻⁴. We will comment on this in Section 5.3 but mention at this place that a superficial contamination with atmospheric He would lower the ³He/⁴He, contrary to what is observed. At larger depths, ³He/⁴He ratios first continue to decrease slightly but the ³He/⁴He pattern is flatter than the corresponding ²⁰Ne/²²Ne pattern. Near the very end, the ³He/⁴He ratios even tend to slightly increase again, contrary to ²⁰Ne/²²Ne ratios.

A possible process to explain the potential He deficit would be a diffusional loss of He caused by target temperatures that were expected to reach 180 °C due to solar irradiation in space. However, this seems unlikely because the ³He and ⁴He distributions plotted as a function of
VI. HELIUM, NEON, AND ARGON ISOTOPIC AND ELEMENTAL COMPOSITION

Table 3a. He and Ne in BMG sample 40598-03-F (0.107 cm²) measured by CSSE

<table>
<thead>
<tr>
<th>Step</th>
<th>Etching time (min)</th>
<th>⁴He (x10¹⁴)</th>
<th>³He/⁴He (x10⁻⁴)</th>
<th>²⁰Ne (x10¹¹)</th>
<th>²⁰Ne/²²Ne</th>
<th>²¹Ne/²²Ne</th>
<th>⁴He/²⁰Ne</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>120</td>
<td>0.05</td>
<td>4.87</td>
<td>0.05</td>
<td>16.08</td>
<td>0.0373</td>
<td>986</td>
</tr>
<tr>
<td>2</td>
<td>500</td>
<td>0.56</td>
<td>4.79</td>
<td>0.90</td>
<td>14.72</td>
<td>0.0359</td>
<td>625</td>
</tr>
<tr>
<td>3</td>
<td>480</td>
<td>0.80</td>
<td>4.38</td>
<td>1.50</td>
<td>14.62</td>
<td>0.0332</td>
<td>531</td>
</tr>
<tr>
<td>4</td>
<td>480</td>
<td>1.76</td>
<td>4.32</td>
<td>2.86</td>
<td>14.19</td>
<td>0.0332</td>
<td>615</td>
</tr>
<tr>
<td>5</td>
<td>300</td>
<td>0.06</td>
<td>4.43</td>
<td>0.20</td>
<td>13.56</td>
<td>0.0330</td>
<td>322</td>
</tr>
<tr>
<td>6</td>
<td>450</td>
<td>1.31</td>
<td>4.24</td>
<td>2.82</td>
<td>13.52</td>
<td>0.0324</td>
<td>463</td>
</tr>
<tr>
<td>7</td>
<td>300</td>
<td>0.17</td>
<td>4.16</td>
<td>0.52</td>
<td>13.51</td>
<td>0.0329</td>
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<tr>
<td>8</td>
<td>400</td>
<td>0.42</td>
<td>4.16</td>
<td>1.58</td>
<td>13.11</td>
<td>0.0316</td>
<td>263</td>
</tr>
<tr>
<td>9</td>
<td>480</td>
<td>0.08</td>
<td>4.20</td>
<td>0.45</td>
<td>11.87</td>
<td>0.0313</td>
<td>176</td>
</tr>
<tr>
<td>10</td>
<td>1000</td>
<td>0.12</td>
<td>4.43</td>
<td>0.47</td>
<td>11.59</td>
<td>0.0315</td>
<td>249</td>
</tr>
<tr>
<td>11</td>
<td>1800</td>
<td>0.07</td>
<td>4.53</td>
<td>0.17</td>
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<tr>
<td>12</td>
<td>5400</td>
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<td>0.01</td>
<td>11.16</td>
<td>0.0398</td>
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<tr>
<td>Σ/mean</td>
<td>11710</td>
<td>5.42</td>
<td>4.39</td>
<td>11.60</td>
<td>13.67</td>
<td>0.0327</td>
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</tr>
<tr>
<td>Σ/mean corr.</td>
<td>6.38</td>
<td>4.50</td>
<td>13.03</td>
<td>13.87</td>
<td>0.0330</td>
<td>494</td>
<td></td>
</tr>
</tbody>
</table>

* corrected for backscatter losses (see text). See Table 2 for uncertainties of the backscatter correction. Gas concentrations in (atoms/cm²). Uncertainties (2σ) are: Gas concentrations: 4%, ³He/⁴He and ²⁰Ne/²²Ne: <1.5%, ²¹Ne/²²Ne:<5% (italicized values <10%, step 12 <30%), ⁴He/²⁰Ne: 5%.

etch time per step are similarly narrow as the Ne and Ar distributions, unlike the broadened distributions that would be expected as a result of He diffusion. Another explanation might be underestimated He backscatter losses from the BMG. The irradiation experiment at the CASYMS was carried out with a single velocity of 400 km/s. Thus, for velocities <400 km/s backscatter losses of He might be higher than expected. Lower-than-predicted He trapping efficiencies were indeed observed in irradiation experiments for the COLLISA experiment at velocities <100 km/s (Filleux et al. 1980). On the other hand, unaccounted backscatter losses from low target depths would even increase the contribution of the light helium component, and furthermore, since the backscatter losses of ³He are always higher than for ⁴He, they would also increase the ³He/⁴He ratio of this light component.

Even though it appears that the BMG data do not allow to derive an accurate SW He fluence, it is fair to note that this is the only target so far for which the He trapping efficiency has been determined experimentally. Further investigations on other Genesis bulk SW targets will be required to obtain a reliable He fluence during the Genesis exposure period. Additionally, the fact that the Ne fluence obtained with the ACE instruments is almost two times as high as all values reported from Genesis bulk samples and also as measured by instruments onboard the ULYSSES satellite (Reisenfeld et al., 2006) reduces the significance of the putative difference of fluences between ACE and the ones derived from the BMG.
Unlike for Ne, the observed variability of \(^{3}\text{He}/^{4}\text{He}\) ratios with depth is very different from that predicted by SRIM. The observed \(^{3}\text{He}/^{4}\text{He}\) ratios vary orders of magnitude less than those predicted (SRIM values are only partly shown in Fig. 4). However, \(^{3}\text{He}/^{4}\text{He}\) ratios in lunar ilmenites (Heber et al., 2003) also show little variation with depth. Hence, we conclude that SRIM does not only fail in predicting isotopic fractionation due to backscattering of the very light element He from the BMG as has been shown in the irradiation experiments, but in particular also overestimates the depth-dependent isotopic separation of the implanted He.

Table 3b. He and Ne in BMG sample 40598-03-B/04-K (0.371 cm\(^2\)) measured by CSSE

<table>
<thead>
<tr>
<th>Step</th>
<th>Etching time (min)</th>
<th>(^{4}\text{He}) ((\times 10^{14}))</th>
<th>(^{3}\text{He}/^{4}\text{He}) ((\times 10^{-4}))</th>
<th>(^{20}\text{Ne}) ((\times 10^{11}))</th>
<th>(^{20}\text{Ne}/^{22}\text{Ne})</th>
<th>(^{21}\text{Ne}/^{22}\text{Ne})</th>
<th>(^{4}\text{He}/^{20}\text{Ne})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>240</td>
<td>0.09</td>
<td>4.90</td>
<td>0.10</td>
<td>15.58</td>
<td>0.0351</td>
<td>901</td>
</tr>
<tr>
<td>2</td>
<td>180</td>
<td>0.03</td>
<td>4.94</td>
<td>0.05</td>
<td>15.52</td>
<td>0.0321</td>
<td>712</td>
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*a* corrected for backscatter losses (see text). See Table 2 for uncertainties of the backscatter correction.

Gas concentrations in (atoms/cm\(^2\)).

Uncertainties (2\(σ\)) are: Gas concentrations: 4\%, \(^{3}\text{He}/^{4}\text{He}\): <1\%, \(^{20}\text{Ne}/^{22}\text{Ne}\): <2\% (italicized values <3\%), \(^{21}\text{Ne}/^{22}\text{Ne}\): <5\% (italicized values <15\%, steps 3 and 30 <30\%), \(^{4}\text{He}/^{20}\text{Ne}\): 4\%.
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* Gas bubble opened during etching. Ne values are corrected for 42% of blank gas contribution; Ar values discarded.

* The Ar phase had to be discarded before measurement because of too large amounts of active gases.

* Corrected for backscatter losses (see text). See Table 2 for uncertainties of the backscatter correction.

* Corrected for blank contributions of atmospheric Ar via 40Ar

Gas concentrations in (atoms/cm²).

Uncertainties (2σ) are: 4% 3He/4He and 36Ne/22Ne: <1% (italicized values <1.5%), 21Ne/22Ne: <4% (italicized values <4%), 36Ar/38Ar: <0.5%, 40Ar/38Ar: 2%, 36Ar/20Ne: 4%, 36Ar/36Ar: 2%, 36Ar/38Ar: 2%.

### 4.1.3. Argon

Two samples were analyzed by pyrolysis for bulk SW Ar. In these two samples the average 40Ar/36Ar ratio of 277±2 indicates large blank Ar contributions of atmospheric isotopic composition from gas contained in the BMG itself. Determination of 36,38Ar via 40Ar, the latter assumed to be entirely blank derived, would have led to corrections of more than 92%. We therefore discard the pyrolytic measurements.

Ar was also measured in one CSSE run. The measured data are given in Table 3c, the blank-
corrected $^{36}\text{Ar}^{38}\text{Ar}$ ratios are shown in Fig. 5 and also in Table 3c. As discussed in Section 3.2, blank corrections are often very substantial, as indicated by $^{40}\text{Ar}^{36}\text{Ar}$ ratios between 290 and 49. Step 14 had to be discarded because etching opened a gas bubble and released large amounts of gas. The general pattern is well explained by mass separation of a uniform SW component upon implantation. Ratios within the first 10% of Ar released are enhanced relative to subsequent steps, discussed in Section 5.3.

Despite of the uncertainties due to blank corrections, the Ar data from the CSSE runs provide valuable information. The basic data pattern shown in Fig. 5 is similar to the measured Ne pattern. First, $^{36}\text{Ar}^{38}\text{Ar}$ ratios in the etch steps representing the first 10% of the total $^{36}\text{Ar}$ released are markedly higher than the values in the subsequent steps and higher than predicted by SRIM for a uniform isotopic ratio in the entire SW velocity range. Second, after these initial steps, $^{36}\text{Ar}^{38}\text{Ar}$ ratios slowly decrease with increasing etching depths and the measured data are well reproduced by the SRIM simulations for mass separation of an isotopically uniform SW with a
$^{36}\text{Ar}/^{38}\text{Ar}$ ratio of 5.3. We conclude that the isotopic variation with depth for the gas representing the last 90% of total $^{36}\text{Ar}$ released are a result of mass separation upon implantation. As for the $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{4}\text{He}/^{4}\text{He}$ ratios, the high $^{36}\text{Ar}/^{38}\text{Ar}$ ratios in the first steps cannot be explained by atmospheric contamination on the BMG surface, as a superficially dissolved atmospheric component would rather lead to a depletion of the light isotope.

The “best” $^{36}\text{Ar}/^{38}\text{Ar}$ ratio for the bulk SW to be derived from our CSSE data is 5.30±0.24 (corrected for backscattering). However, due to the large blank correction this value needs to be taken with a grain of salt. It is consistent with the average SWC foil ratio of 5.4±0.3 (Geiss et al., 2004) but lower than the bulk value reported by Mabry et al. (2007) of 5.5 in the Genesis ALoS target or values of 5.5-5.6 derived from lunar soils and solar-gas rich meteorites (Benkert et al., 1993; Becker et al., 1998). It is also lower but within the stated uncertainties of the values for coronal hole- and inter-stream-related SW observed with SOHO/CELIAS/MTOF by Weygand et al. (2001).

4.2. Elemental Composition

The bulk $^{20}\text{Ne}/^{36}\text{Ar}$ ratio derived from a single CSSE run is given in Table 2. The single $^{20}\text{Ne}/^{36}\text{Ar}$ ratios for each step of this run as a function of the cumulative fraction of $^{36}\text{Ar}$ released

![Fig. 6. Measured depth profile of the elemental ratio $^{20}\text{Ne}/^{36}\text{Ar}$ of the third CSSE run versus the cumulative $^{36}\text{Ar}$ fraction. SRIM simulations assume a velocity-independent $^{20}\text{Ne}/^{36}\text{Ar}$ ratio of 44.1±2.2 (bulk value determined by CSSE). Similar to Fig. 5 some steps suffering from large blank gas contributions are plotted in parentheses and the first step is not plotted. Step 14 was discarded. The general pattern is similar to that predicted by SRIM. This confirms that mass separation of a solar wind with uniform composition does essentially explain the $^{20}\text{Ne}/^{36}\text{Ar}$ elemental pattern. An exception to this is again the slight enrichment of the light element Ne relative to Ar in the first few steps (Section 5.3).](image-url)
are plotted in Figure 6, and Figure 7 shows in the same format as Figure 6 the $^4\text{He}/^{20}\text{Ne}$ ratio as a function of the cumulative fraction of $^{20}\text{Ne}$ for all three runs (Tables 2, 3a-c).

We first discuss the Ne/Ar ratios. The backscatter-corrected bulk $^{20}\text{Ne}/^{36}\text{Ar}$ ratio derived from a single CSSE run of $44.1\pm2.2$ agrees within uncertainties with the value reported from the SWC foils (Tab. 2). As already noted in Section 4.1.3, some of the steps in this run show particularly large blank Ar contributions compared to adjacent steps. The blank corrected $^{20}\text{Ne}/^{36}\text{Ar}$ ratios of these steps are significantly lower than the values of neighbouring steps and are given in parentheses (Steps 11 and 22) or are not shown at all in Figure 6 (Steps 1 and 14). Despite the uncertainties caused by the blank correction, the $^{20}\text{Ne}/^{36}\text{Ar}$ profile (Fig. 6) shows striking similarities to the isotopic profiles. In the gas fractions released from shallow depths $^{20}\text{Ne}/^{36}\text{Ar}$ ratios are enriched in the light species and plot well above the backscatter-corrected bulk $^{20}\text{Ne}/^{36}\text{Ar}$ ratio. Additionally, $^{20}\text{Ne}/^{36}\text{Ar}$ ratios in these steps are above SRIM predictions that are based on the bulk $^{20}\text{Ne}/^{36}\text{Ar}$ ratio deduced from the BMG. At larger depths the pattern of $^{20}\text{Ne}/^{36}\text{Ar}$ ratios flattens out, although the measured ratios slightly increase with depth, somewhat in conflict to the SRIM predictions for an elementally uniform SW composition.

All three measured $^4\text{He}/^{20}\text{Ne}$ profiles (Fig. 7) agree quite well with each other and show very high ratios in the first steps. A second maximum with $^4\text{He}/^{20}\text{Ne}$ values up to 900 is seen in all runs at about 28% of $^{20}\text{Ne}$ is released. This maximum is followed by a broad minimum at 80-100% of released Ne and another strong increase in the very final fractions. Besides the high

![Graph showing approximate implantation depth of $^{20}\text{Ne}$ according to SRIM (nm). The graph includes data points and lines indicating measured and simulated ratios.](image)

**Fig. 7.** Measured depth-dependent elemental ratio $^4\text{He}/^{20}\text{Ne}$ derived from three CSSE runs versus the cumulative $^{20}\text{Ne}$ fraction. SRIM simulations assume a velocity-independent $^4\text{He}/^{20}\text{Ne}$ ratio of $527\pm39$ derived from total extractions. The distribution shows three peaks, a pattern that is completely different from SRIM simulations, the $^{20}\text{Ne}/^{36}\text{Ar}$ distribution, and all isotopic patterns.
ratios in the first steps, these are all features that are not predicted by SRIM for a uniform SW composition and have also no counterparts in the Ne/Ar or in the isotopic ratio profiles.

In summary, the similarity of the Ne/Ar depth distribution to that of the isotopic ratios is supportive evidence for a mass separation of atoms within the BMG upon implantation. The enhancement of the light element Ne over Ar in the first etching steps coincides with the enrichment of light isotopes over the heavier ones in these first steps. The interpretation of the He/Ne ratios with depth is hampered by their complex distribution.

5. DISCUSSION

5.1. Mass Separation upon Implantation

The very good agreement of the observed depth distribution of Ne isotopes in the BMG with SRIM predictions for a SW with uniform composition strongly indicates that the observed depth distribution is primarily a result of mass separation upon implantation. This has recently been discussed by Grimberg et al. (2006). The small deviations in the Ne pattern from SRIM predictions for very shallowly sited gas will be discussed separately in Section 5.3.

Similar depth distributions of solar Ne isotopes are observed in lunar and meteoritic samples exposed to the solar wind (Wieler et al., 1986; Benkert et al., 1993; Pepin et al., 1999). These data have widely been interpreted to result from a mixture of two isotopically distinct solar sources, namely SW particles and ions from solar energetic particle (SEP) events (Etique et al., 1981; Wieler et al., 1986; Benkert et al., 1993; Pepin et al., 1999). As has been discussed by Grimberg et al. (2006), the good agreement of the Ne profiles in the BMG with profiles in the extraterrestrial samples clearly shows that the former interpretation has become obsolete. The elimination of the “SEP” noble gas component solves a long-standing enigma. “SEP” gases would have represented up to several 10% of the total solar gases retained in lunar samples. This is many orders of magnitude higher than the SEP/SW abundance ratio for Ne of ~0.0001 as measured for the exposure time of Genesis with ACE instruments in the energy range of 5 to 1000 keV/nucleon (R.A. Mewaldt, private communication).

Now we address the question whether the He and Ar data support the interpretation of the Ne profiles by Grimberg et al. (2006) who argue that these are the result of mass separation upon implantation. Although the apparent discrepancy between He/Ne ratios found in different
Genesis targets renders the discussion about helium profiles somewhat difficult, we nevertheless conclude that the rather flat pattern of measured $^{3}\text{He}/^{4}\text{He}$ ratios reflects the true distribution of the He isotopes much better than the SRIM simulations. This is particularly so because the artificial irradiations indicate that SRIM predictions for the absolute amount of backscattering of He isotopes are unreliable. This conclusion is supported by CSSE data on lunar ilmenites (Heber et al., 2003), which also show only a modest mass separation of the He isotopes with depth. Thus, it seems reasonable to conclude that in all likelihood also the distribution of He isotopes in the BMG and in lunar and asteroidal regolith samples is mainly a result of mass separation upon implantation.

In contrast to the somewhat ambiguous He data, the $^{36}\text{Ar}/^{38}\text{Ar}$ depth profile in the BMG clearly yields additional support to the conclusion drawn from the Ne isotopes. The measured profile shows similar variations with depth as that simulated with SRIM for an isotopically uniform SW. We conclude that the general $^{36}\text{Ar}/^{38}\text{Ar}$ pattern is also essentially explained by mass separation upon implantation. Similar patterns as observed in the BMG are also reported for solar Ar in lunar samples (Benkert et al., 1993; Murer et al., 1997; Pepin et al., 1999). The conclusion by Grimberg et al. (2006) that the former “SEP” Ne component is obsolete can therefore be extended to Ar.

In general the measured $^{20}\text{Ne}/^{36}\text{Ar}$ ratios show a similar consistency with SRIM predictions as is observed for the Ne and Ar isotopic composition. We therefore conclude that the $^{20}\text{Ne}/^{36}\text{Ar}$ distribution at greater depths is generally also well explained by mass separation upon implantation. Because meteorites and lunar samples often show similar distributions of solar $^{20}\text{Ne}/^{36}\text{Ar}$ ratios with depths as the BMG (e.g. (Murer et al., 1997), also the Ne and Ar elemental depth distribution in lunar samples is mostly governed by mass separation of a uniform SW; a further argument that a second solar component “SEP” Ar needs no longer to be invoked.

In summary, the consistency of measured depth profiles of Ne and Ar isotopic and elemental abundances with SRIM simulations for a uniform SW leaves no doubt that mass separation is the major process controlling the distribution of solar noble gases in the BMG. On the other hand, the question may arise whether the fact that at cumulative Ne fractions $>$0.1 measured $^{20}\text{Ne}/^{22}\text{Ne}$ ratios in Figure 3 show a slightly less steep slope than the SRIM prediction may suggest an isotopic fractionation of the SW favouring the light isotopes at higher SW velocities. We do not think that our data supports this notion. Such a fractionation trend would be contrary to the Coulomb drag theory (Bodmer and Bochsler, 1998), which is the only known process which might significantly influence isotopic compositions. The enrichment of light isotopes
with increasing SW velocity would also be opposite to the possible fractionation of Mg and Ne isotopes as a function of SW speed observed by in-situ measurements from SOHO/CELIAS/MTOF (Kallenbach et al., 1997a; 1997b; 1998). We therefore conclude that the slightly different slopes of measured data and SRIM-predicted profiles in Fig. 3 is due to a not perfectly homogeneous etching of the BMG.

5.2. Bulk Solar Wind Composition

The bulk values of He, Ne, and Ar concentrations as well as isotopic and elemental ratios determined from three pyrolysis and three CSSE experiments of different BMG aliquots all agree very well with each other. Only the \( ^4\text{He} / \text{Ne} \) bulk values from the CSSE runs differ somewhat from the pyrolysis average, which is probably caused by the accumulation of uncertainties of absolute amounts over many etching steps. Thus we prefer the total extraction data to represent the bulk SW noble gas composition reported here.

Backscatter-corrected He, Ne and Ar isotopic ratios are all consistent with the bulk SW average values from the SWC foil data (Geiss et al., 2004). We therefore conclude that the bulk SW isotopic compositions of He, Ne, and Ar do not vary significantly on hourly and daily scales in low-speed solar wind regimes recorded by the SWC experiments and do not differ from the 2.5 year average recorded by the Genesis BMG target. Again we note that our Ar data have to be taken with some care as a result of the relatively high blank Ar contributions. Because backscatter corrections for the BMG were presumably relatively large compared to most other target materials flown on Genesis, all bulk values reported here should not be considered as final bulk SW compositions derived from the Genesis mission.

Previously we noted that besides the \( ^3\text{He}/^4\text{He} \) ratio also the bulk \( ^4\text{He} / \text{Ne} \) value is in very good agreement with data from the different SWC experiments, which would show that He was almost quantitatively trapped in the BMG (Grimberg et al., 2007). As discussed above, this statement may have to be reconsidered if the low He fluence measured in the BMG is due to He losses from this target. In this case the fact that the BMG bulk data point in a \( ^3\text{He}/^4\text{He} \) vs. \( ^4\text{He}/\text{Ne} \) diagram falls on a fractionation line defined by the Coulomb drag model (Bodmer and Bochsler, 1998), which reproduces the individual SWC foil data well (Geiss et al., 2004; Bochsler, 2007), would be coincidental. We reiterate, however, that so far the BMG is the only Genesis sample providing He concentration data experimentally calibrated for backscatter losses. At this point, we thus regard the agreement between BMG and SWC data as evidence supporting the hypothesis that the SW composition becomes fractionated by Coulomb-drag, although a final
assessment will have to await a conclusive intercalibration of trapping efficiencies and absolute measurement procedures of the different target materials.

5.3. Fractionation of Low Speed Solar Wind

From the enhancement of light isotopes of He, Ne, and Ar in the very first gas fractions released from the BMG we concluded in Section 4 that this fraction represents very low-speed SW, isotopically lighter than the bulk SW. Grimberg et al. (2007) already proposed that the most plausible explanation of the Ne data from the first gas release fractions is a depletion of heavy isotopes in very low-speed SW by inefficient Coulomb drag (Bodmer and Bochsler, 1998; Bochsler, 2007). This suggestion was supported by the fact that the $^3\text{He}/^4\text{He}$ and $^4\text{He}/^{20}\text{Ne}$ bulk ratios from the BMG fell on the Coulomb drag fractionation line. As discussed in the previous section, this argument might need reconsideration now. Nevertheless, the effect of inefficient Coulomb drag and gravitational settling in the corona on the composition of low-speed SW is corroborated by a depletion of $\alpha$-particles at low SW velocities during the exposure time of Genesis as revealed by small He/H ratios recorded with the Genesis ion monitor.

We now focus on the $^{20}\text{Ne}/^{22}\text{Ne}$, $^{20}\text{Ne}/^{36}\text{Ar}$, and $^{36}\text{Ar}/^{38}\text{Ar}$ ratios in the first few steps, which are enhanced relative to the subsequent steps. We use the approximations of Bodmer and Bochsler (1998) and Bochsler (2000) to determine the fractionation of Ne and Ar isotopes as expected from inefficient Coulomb drag. The formalism is given in the Appendix, and Figures 8 and 9 show the resulting correlation of $^{20}\text{Ne}/^{22}\text{Ne}$ with $^{20}\text{Ne}/^{36}\text{Ar}$ on the one hand and $^{36}\text{Ar}/^{38}\text{Ar}$ on the other hand. With increasing SW velocity the isotopic and elemental composition changes towards a heavier composition. Highest ratios are predicted for the very low-speed, current-sheet related SW. The current-sheet is theoretically defined by a strong superradial expansion of magnetic fields in the inner corona, lowering the flux of protons that exert the drag on the heavier

![Fig. 8. $^{20}\text{Ne}/^{22}\text{Ne}$ vs. $^{20}\text{Ne}/^{36}\text{Ar}$ diagram of raw data as given in Table 3c for the first ten etching steps of sample 40598-04-E/-F/-G/-J. Note that the data have not been corrected for some mass-dependent separation effects during implantation into the target. To position the grey line in Figure 1 we have therefore not taken “unfractionated” solar wind values but an average of uncorrected data from etching steps no 8 to 10, i.e. $(^{20}\text{Ne}/^{22}\text{Ne})_{\text{reference}} = 14.5$ and $(^{20}\text{Ne}/^{36}\text{Ar})_{\text{reference}} = 48.$](image-url)
ions. The data points for the first ten etching steps are also plotted in Figures 8 and 9. They are almost perfectly matched by the theoretical fractionation line predicted for inefficient Coulomb drag. On the other hand, this explanation is not easily reconciled with the very pronounced decrease of isotopic ratios as well as $^{20}\text{Ne}/^{36}\text{Ar}$ ratios over the first 10% of gas released. These initial gas fractions presumably were trapped in the outermost ~10 nm, but according to SRIM, ions with typical for current-sheet related SW velocities of ~260-320 km/s exhibit considerably stronger longitudinal straggling comparable to the penetration depths, and, correspondingly, a wider range distribution within the target.

The very low depths in which the light component resides implies an implantation with very low energies on the order of 50 eV/amu, corresponding to a velocity of ~100 km/s. Thus, neutral atoms have to be considered as another possible source for the low-speed component in the BMG. This neutral component could be either of interstellar origin or neutral SW (Mabry et al., 2007) or recycled SW particles previously trapped by the Genesis satellite itself.

However, all three possibilities seem unable to account for the amounts observed. Interstellar neutral He at 1 AU has been measured by Salerno et al. (2003) and Busemann et al. (2006) with the COLLISA experiment on the MIR space station. The exposure was carried out during a period of maximum relative velocities between interstellar gas and trapping device. The interstellar $^4\text{He}$ flux trapped in the BeO layer corrected for backscattering is more than three orders of magnitude lower than the flux of the doubly charged solar wind $^4\text{He}$ (Busemann et al., 2006). Hence, even assuming that the trapping probability for very low-energy particles in the BMG would not be lower than that in the light-element target BeO, e.g. 24% for $^4\text{He}$ in BeO (Salerno et al., 2003), interstellar He cannot account for the isotopically light component. Furthermore, interstellar He is isotopically much heavier ($^4\text{He}/^3\text{He} = (1.62±0.29)\times10^{-3}$; Busemann, 2006) than SW He, opposite to the shallowly sited gas fraction in the BMG and the Al target.

![Fig. 9. $^{20}\text{Ne}/^{22}\text{Ne}$ vs. $^{36}\text{Ar}/^{38}\text{Ar}$ diagram of raw data as given in Table 3c for the first ten etching steps of sample 40598-04. Note that the data have not been corrected. To position the grey line we have used the second of equations 7 and again taken “unfractionated” solar wind values but an average of uncorrected data from etching steps no 8 to 10, i.e. $(^{20}\text{Ne}/^{22}\text{Ne})_{\text{reference}} = 14.5$ and $(^{36}\text{Ar}/^{38}\text{Ar})_{\text{reference}} = 5.54.$]
VI. helium, neon, and argon isotopic and elemental composition

A neutral component in the SW is discussed, e.g., by Collier et al. (2001) or Bochsler et al. (2007). Collier et al. (2001) report an SW$_{\text{neutral}}$/SW ratio for hydrogen on the order of $10^{-3}$. Only with the assumption that all neutral H reported by Collier et al. (2001) would in fact be neutral He in the right magnitude as for the SW He/H composition, the resulting amounts of neutral He would approximately account for the isotopically light gas fraction observed in the Al and BMG target.

Yet other conceivable explanations for neutral atoms might be recycling of SW particles lost from the Genesis targets. Re-trapping might occur in the same way as it was proposed for $^{40}$Ar on the lunar surface (Manka and Michel, 1971) or perhaps by knock-on of atoms temporarily residing in the molecular film. All these possibilities fail to quantitatively explain the isotopically light component, however.

In summary, the enrichment of the light isotopes and elements for He, Ne, and Ar sited in shallow depth cannot stem from atmospheric contamination or be caused by diffusive fractionation. Therefore, the light component must in all probability represent particles implanted with low energy. The source of this particle population, however, is difficult to assess. A neutral solar or interstellar component may have velocities low enough to explain the very shallow implantation depth. However, abundances are too small to account for the gas observed in the Genesis targets. Interstellar He also has a lower $^3$He/$^4$He ratio than the SW, contrary to the observations. On the other hand, inefficient Coulomb drag would produce the right degree of fractionation and can explain the correlation of Ne and Ar compositions observed in the BMG. However, this model can not easily account for the small penetration depth and the steep decrease of isotopic and elemental ratios observed. Nevertheless, we believe that fractionation by inefficient Coulomb drag of current sheet related SW so far is the most feasible explanation for the pattern observed. This interpretation is also supported by measurements of the He and, to a minor degree, the Ne isotopic composition in different SW regime targets from Genesis, which also show an enrichment of the light isotope in the slow SW relative to the bulk SW (Heber et al., 2007).

6. CONCLUSIONS

The main objective, which led to the selection of the bulk metallic glass as a special purpose target for the Genesis mission has entirely been fulfilled. This sample has allowed us to determine high-resolution depth profiles of isotopic and elemental composition of implanted solar He, Ne, and Ar by the closed system stepwise etching technique. The Ne and Ar profiles in particular can be perfectly explained by a mass separation upon implantation of solar wind noble gases.
with an essentially uniform, energy-independent composition. This conclusion can be extended to solar noble gases implanted into lunar regolith samples and meteorites rich in solar gas. As a consequence, the enigmatic “SEP” noble gas component thought to represent solar energetic particles with a composition isotopically heavier than the solar wind can be eliminated from the “noble gas alphabet”. This solves the long-standing problem of an unreasonably high apparent flux of solar energetic particles in the past. Further consequences of this finding for noble gas geo- and cosmochemistry are discussed by Wieler et al. (2007).

The bulk metallic glass is not perfectly suited for an accurate determination of the isotopic and elemental composition of bulk solar wind noble gases. The high proportion of relatively heavy elements in the glass leads to substantial corrections for backscatter losses of He and also, but to a minor degree, of Ne. Ar measurements are compromised by large blank contributions from Ar incorporated into the glass during manufacturing. Nevertheless, our isotopic and elemental He, Ne and Ar values, representing an exposure average over ~2.5 years, are in good agreement with the mean values from the Apollo Solar Wind Composition experiment (Geiss et al., 2004) for much shorter exposure times and values from lunar samples and meteorites, representing a very long-term SW record. Up to date our results thus also support the hypothesis that the SW composition becomes fractionated by Coulomb-drag.

An interesting observation is a gas fraction sited in very shallow depths in the bulk metallic glass which is lighter in its isotopic and elemental composition than the bulk solar wind. Because this composition cannot be explained by contamination, diffusion, or mass separation upon implantation, it must represent a component with a composition lighter than normal solar wind, presumably also with a low He/H abundance ratio, which was implanted at low speeds. It is unlikely that this gas fraction represents a low-energy neutral component of solar or interstellar origin, because such sources cannot account for several percent of the total gas in the BMG, and interstellar He is also heavier than the He composition in the SW. In our view, the most feasible explanation is a very low-speed, current sheet related solar wind fractionated relative to bulk SW by inefficient Coulomb drag, although this explanation cannot straightforwardly account for the rapid change in composition with depth within the shallowly sited gas fraction.
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REFERENCES


VI. Helium, Neon, and Argon Isotopic and Elemental Composition


APPENDIX: Elemental and Isotopic Fractionation by Inefficient Coulomb Drag in the Solar Wind

Using the approximations of Bodmer and Bochsler (1998) and Bochsler (2000) we discuss fractionation patterns observed in the Genesis bulk metallic glass (BMG) target. The theoretical framework of Bodmer and Bochsler (1998) rests on the work of Bürgi (1992) who argued that the superradial expansion of magnetic fields below the Alfvén point in the inner corona regulates the coupling of helium to the proton flux by Coulomb friction, and that the proton flux itself is constrained by the coronal helium abundance. The assessment of the importance of Coulomb drag on the abundance of helium and the heavy ions in the solar wind is rendered difficult by the fact that the so-called FIP-effect (i.e., depletion of elements with high first ionization potentials) might also lead to a systematic depletion of helium in the solar wind. Bochsler et al. (2006) made an attempt to distinguish between these two possible causes and concluded that indeed the main responsible for the systematic depletion of helium in the solar wind is inefficient Coulomb drag.

In the formalism of Bodmer and Bochsler (1998) the fractionation factor of \(^4\text{He}\), which describes the depletion of helium relative to hydrogen and relative to the source composition in a given type of wind flow relative to the source composition, is assumed to follow the approximation

\[
f_{4H} = 1 - C^* H_4 ,
\]

where \(C^*H_4\) denotes the friction factor of \(^4\text{He}\), which causes the depletion of this species relative to its abundance at the source. The H-factor of a species with atomic mass \(A\) and charge state \(Q\) in the inner corona for a given species is (Bodmer and Bochsler, 1998)

\[
H = \frac{2A - Q - 1}{Q^2} \sqrt{\frac{A+1}{A}} .
\]

The H-factors for some species relevant in this paper are listed in Table A1. In analogy to equation 1, it is possible to express the depletion of any other species, relative to hydrogen, e.g.:

\[
f_{20H} = 1 - C^* H_{20} ,
\]

\[
f_{22H} = 1 - C^* H_{22} ,
\]

\[
f_{36H} = 1 - C^* H_{36} .
\]
Eliminating $C^*$ using equation 1 one finds:

$$f_{20H} = 1 - (1 - f_{4H}) \frac{H_{20}}{H_4}$$

$$f_{22H} = 1 - (1 - f_{4H}) \frac{H_{22}}{H_4}$$

$$f_{36H} = 1 - (1 - f_{4H}) \frac{H_{36}}{H_4}$$

$$f_{38H} = 1 - (1 - f_{4H}) \frac{H_{38}}{H_4}.$$  \hspace{1cm} (4)

Since hydrogen is not measured in the Genesis targets, we need to define isotopic and elemental ratios without reference to hydrogen. $f_{ij}$ denotes the fractionation factor of an elemental or isotopic abundance ratio compared to its ratio at the source.

$$f_{ij} = \frac{[z]}{[\overline{H}]} = \frac{[z]}{[\overline{H}]_{source}}.$$  \hspace{1cm} (5)

As is common use, brackets denote the abundance of a species.

Table A1. H factors for relevant species

<table>
<thead>
<tr>
<th>Species</th>
<th>H factor ($)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^4\text{He}^+$</td>
<td>1.389</td>
</tr>
<tr>
<td>$^{20}\text{Ne}^+$</td>
<td>0.496</td>
</tr>
<tr>
<td>$^{22}\text{Ne}^+$</td>
<td>0.559</td>
</tr>
<tr>
<td>$^{36}\text{Ar}^+$</td>
<td>0.998</td>
</tr>
<tr>
<td>$^{38}\text{Ar}^+$</td>
<td>1.092</td>
</tr>
</tbody>
</table>

Adopted charge states: $\text{He}^+$, $\text{Ne}^+$, $\text{Ar}^+$

Dividing two fractionation factors relative to hydrogen as given in equation 4 through each other yields isotopic and elemental fractionation factors

$$f_{202} = \frac{(H_4 - H_{20}) + f_{4H} \cdot H_{20}}{(H_4 - H_{22}) + f_{4H} \cdot H_{22}},$$

$$f_{2036} = \frac{(H_4 - H_{20}) + f_{4H} \cdot H_{20}}{(H_4 - H_{36}) + f_{4H} \cdot H_{36}}.$$  \hspace{1cm} (6)
Relating two ratios we eliminate $f_{41h}$, the depletion factor of helium relative to hydrogen:

$$f_{2036} = \frac{f_{2022} \cdot (H_{22} - H_{20})}{(H_{36} - H_{20}) - f_{2022} \cdot (H_{36} - H_{22})},$$

and

$$f_{3638} = \frac{(H_{36} - H_{20}) - f_{2022} \cdot (H_{36} - H_{22})}{(H_{38} - H_{20}) - f_{2022} \cdot (H_{38} - H_{22})}. \tag{7}$$

The fractionation factors as given in equation 7 can be used to describe the composition of a sample under the influence of a varying degree of fractionation relative to its source or a reference point. In our case we apply the equations to bulk solar wind as measured in the etching steps 8 to 10. We do not correct for mass dependent separation effects during implantation into the target as discussed in Section 4. The isotopic or elemental ratio $([i/j]_{\text{sample}})$ is thus

$$(\frac{i}{j})_{\text{sample}} = f_{ij} \cdot (\frac{i}{j})_{\text{reference}}. \tag{8}$$

To plot a fractionation line as shown in Figure 8 one simply has to replace the factors $f_{2036}$ and $f_{2022}$ by the respective elemental or isotopic ratios as stated in equation 8. Figure 8 shows a three-isotope plot for raw data as given in Table 3c for the first ten etching steps of sample 40598-04. Note that the data have not been corrected for some mass-dependent separation effects during implantation into the target. To position the line in Figure 8 we have therefore not taken “unfractionated” solar wind values for $x_{\text{reference}}$ and $y_{\text{reference}}$, but an average from etching steps no 8 to 10, i.e. $^{20}\text{Ne}^{32}\text{Ne}_{\text{reference}} = 14.5$ and $^{20}\text{Ne}^{36}\text{Ar}_{\text{reference}} = 48$. The trend of the curve is consistent with the observations, lending some credibility to our hypothesis attributing the enhancement of light isotopes to the effect of inefficient Coulomb drag in low-speed, helium-depleted solar wind.

Figure 9 shows a four-isotope plot produced with the second of equations 7. Again we have used the uncorrected results of etching steps 8 to 10 as a reference. The match of the calculated curve with the measurements is good. However, the variability of the measured argon isotopic ratios from the first few etching steps is marginally significant and therefore the consistency of the data with the Coulomb drag model should not be overemphasized.
REFERENCES


VII. CONCLUSIONS AND OUTLOOK
1. CONCLUSIONS

This thesis discusses noble gas analyses on a bulk metallic glass (BMG) sample that was exposed to the solar corpuscular radiation for 853 days on NASA’s Genesis mission. I used the two different gas extraction methods, total extraction via pyrolysis and depth dependent extraction via close system stepwise etching (CSSE), to analyse the trapped solar noble gases regarding their bulk composition and their compositional dependence on variations with implantation energy.

In a first part of this study, I carried out extensive irradiation experiments using the calibration system for mass spectrometers at the University of Bern to examine the trapping behaviour for noble gases of different target materials, including the BMG. These experiments were compared to irradiation simulations I performed with the open-access computer code SRIM (stopping and range of ions in matter), which is a widely used tool to predict the trapping behaviour of ions in target materials. The results of the artificial irradiations revealed problems of the SRIM code for projectiles implanted with low energies that are typical for the solar wind. For most of the target materials studied, SRIM predicted too small losses of He isotopes due to backscattering, a fact that is of great importance for the interpretation of many targets exposed on Genesis. On the other hand, the probability of He to be trapped in the BMG was underestimated by SRIM. The latter observation now has to be reconsidered regarding the data from the actually flown BMG. Nevertheless, because of its good reproducibility and no better calibrations, the experimentally derived correction factors were preferred for He in the interpretation of data obtained from the target flown on Genesis, whereas SRIM factors have been adopted for Ne and Ar. Similar problems were discovered with the simulation of depth distribution and isotope separation with depth of noble gas isotopes implanted at solar wind conditions in the BMG. While SRIM simulated the behaviour of Ne excellently, the model overestimates the separation of He isotopes with increasing implantation depth.

The main part of this PhD thesis focussed on analyses of the BMG, which was exposed on the Genesis satellite, that I run in the noble gas laboratories at ETH. Because CSSE is a unique extraction method that allows determination of the depth distribution of implanted isotopes with high depth resolution, the BMG got especially designed for this method to distinguish between solar noble gases implanted with different energies. This main expectation was totally fulfilled. The Ne and Ar data deduced from the BMG in three different CSSE runs are in perfect agreement with SRIM simulations for an implantation of solar wind noble gases with an essentially uniform, energy-independent composition. Similarities of the depth dependent
isotopic and elemental distribution with patterns measured in lunar and meteoritic samples led to the conclusion that the solar noble gas composition in these extraterrestrial samples is also mainly controlled by mass separation of a uniform solar wind upon implantation. As a consequence of this, the enigmatic “SEP” noble gas component thought to represent solar energetic particles with a composition isotopically heavier than the solar wind got eliminated from the “noble gas alphabet”. This solves the long-standing problem of an unreasonable high apparent flux of solar energetic particles in the past. Due to the fact that SRIM overestimates the mass separation of He in the BMG, the conclusions drawn from the Ne and Ar compositions can only be assumed to be valid for He as well. This, however, is reasonable because also the He depth distribution observed in the BMG finds its counterparts in lunar and meteoritic samples. The mass separation of He isotopes in lunar ilmenites is similar to what is observed in the BMG, a fact which supports that SRIM simulations are not correct for He at low energies.

In comparison to other solar wind experiments, e.g. the Apollo foils from the solar wind composition experiment, or to in-situ solar wind measurements in space, the isotopic bulk composition of He, Ne, and Ar and the elemental Ne/Ar ratio measured in the BMG mostly agrees with established data. Only the measured He concentration is several percent lower compared to other bulk solar wind samples from Genesis.

The most unexpected observation made is a fractionation of isotopic and, for Ne and Ar, elemental composition favouring the light species in gas that was implanted to very shallow depths only. Because atmospheric contamination on the BMG surface or fractionation by diffusion would shift the gas to even heavier composition, this fraction doubtlessly represents noble gases which were implanted at low speeds and that were fractionated at the source; an observation that got recently confirmed by colleagues in St Louis. The interpretation of this fractionation, however, turned out to be rather difficult as the small implantation depth requires very low particle velocities. Suggestions that this fraction could represent a neutral solar or interstellar component are wrong in all likelihood, as both sources conflict with the abundance or composition observed. The most feasible interpretation is a fractionation of very low-speed, current sheet related solar wind due to inefficient Coulomb drag in the corona, though it collides with the distinct compositional variation over a very small implantation range. However, inefficient Coulomb drag influences both the elemental and the isotopic compositions. It suffices to explain the light composition relative to the bulk solar wind as well as the correlation of compositional variation for Ne and Ar. This conclusion is a cornerstone for the interpretation of other Genesis targets in respect to a variation of the solar wind composition with speed.
2. OUTLOOK

The results deduced from the BMG helped to understand the nature of solar wind implantation and fractionation and solved the long-term “SEP”-noble gas mystery. One important future task will be to clarify whether the conclusions drawn from Ne and Ar are also valid for He. To address this, other target materials consisting of elements much lighter than the BMG are better suited as this would lower the importance of backscatter correction. A second point of large interest is the extent of solar wind noble gas fractionation as a function of solar wind speed. First, to further study the importance of inefficient Coulomb drag on the acceleration of very low-speed solar wind, and second, to examine fractionation of solar wind noble gases at higher velocities; information which could not be deduced with the CSSE method because of limits in reproducibility and precision set by the restrictions in etching behaviour of the BMG and the amount of ions trapped.

During the four years of study it became obvious that the fundamentals of ion trapping at low implantation energies are not fully understood. Therefore, to assist the interpretation of other Genesis targets as well as of future experiments and to help improving predictions of SRIM simulations at low energies, extensive artificial noble gas irradiations would be of great importance, not only for cosmochemistry and solar physics. However, the use of other elements or even molecules in addition to noble gases would widen the implications.
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… and so many thanks to Claudia! Baci…
 CURRICULUM VITAE

Ansgar Grimberg

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<tr>
<th>Date of Birth</th>
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1980 – 1984  Primary school (Grundschule) in Schmallenberg, NRW
1984 – 1993  High school (Gymnasium) in Schmallenberg, NRW
1994 – 1995  Studies of Chemistry at the Ruhr-Universität Bochum
1995 – 2002  Studies of Geology at the Rheinische Friedrich-Wilhelms-Universität zu Bonn;
             Diploma in Geology at the Rheinische Friedrich-Wilhelms-Universität zu Bonn;
             Diploma thesis at the Geologisches Institut at the Rheinische Friedrich-Wilhelms-Universität zu Bonn, under supervision of Prof. Dr. B. Reichert and Prof. Dr. J. Thein:
             *Hydrogeochemische Charakterisierung der einzelnen Grundwasserstockwerke in der südlichen Niederrheinischen Bucht*
2003-2007  Doctoral Work under direction of Prof. Dr. Rainer Wieler at the Institute for Isotope Geochemistry and Mineral Resources, ETH Zürich
From 2007 on  Post Doc at the Physikalisches Institut, Universität Bern