

LOW-ENERGY ION IMPLANTATION: LARGE MASS FRACTIONATION OF ARGON.

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Trapped argon acquired by low-energy implantation (≤ 100 eV) into solids is strongly mass fractionated ($\geq 3\%$ /amu). This has potential implications for the origin and evolution of terrestrial planet atmospheres.

The isotopic signatures of noble gases in the atmospheres of the Earth and other planets are considerably evolved when compared to signatures observed in the solar wind. The mechanisms driving the evolution of planetary volatiles from original compositions in the solar accretion disk are currently poorly understood. As Pepin stated, "The field bristles with models of one type or another, but none are without problems." [1]

Modeling of noble-gas compositional histories requires knowledge of fractionating processes that may have operated through the evolutionary stages. Since these gases are chemically inert, information on noble-gas fractionation processes can be used as probes. The importance of understanding these processes extends well beyond "noble-gas planetology."

The central issue to the origin and evolution of terrestrial planets' atmospheres is how, when, and from what sources did the accreting planetesimals get their gaseous elements. Volatile sources may include the gas phase of the solar nebula, the solar wind, comets and impacts by other bodies. Several mechanisms for the incorporation of planetary atmospheric gases have been proposed; some have been modeled in detail. One class of mechanisms, termed "non-fractionating" processes, includes: gravitational capture of nebular gases, solar-wind loading of accreting matter, and impact degassing of projectiles (comets, meteorites, asteroids). Another class, termed "fractionating" processes, includes: adsorption of gases on accreting grains, implantation of solar wind with sticking coefficients < 1 , ion bombardment in plasma interactions before or during accretion, giant impacts, Jeans' escape and hydrodynamic escape, and losses by non-thermal processes. Some of these mechanisms, such as adsorption, will fractionate only the elements. [2][3][4] Others will involve also isotopic fractionation: ion implantation from plasmas [5][6][7][8], gas sputtering [9], Jeans' escape and hydrodynamic escape. [1][2]

Fractionation mechanisms involving ion implantation have received scant attention possibly because Bernatowicz and coworkers concluded that the isotopic fractionation was too small to account for the large isotopic shifts of Xe observed in the terrestrial atmosphere. [8] On the other hand, Tombrello and coworkers observed significant isotopic shifts in sputtering processes, which were not predicted by theoretical models. [10] It is clear from these results that further examination of an ion implantation model and its related processes is needed. Ion implantation and gas sputtering may prove to be a viable process for fractionation in the origin of planetary atmospheres.

Recently, we have examined the ion implantation of argon onto a tungsten wire using a modified Bayard-Alpert device, which is a refinement of the one Bernatowicz and Hagee [8] used. We can degas the ion collector directly by resistive heating. Both the grid and the walls of the chamber are kept at ground. The filament is at -30 V relative to the grid. The ion collector is varied from -40 V to -100 V relative to the grid at ground. A metal canister encased the filament, grid and ion collector instead of glass, although all the feedthroughs are embedded in glass.

A modified static VG 5400 mass spectrometer was used for isotopic analyses. This instrument is fitted with both a Daly detector and Faraday detectors. The mass spectrometer is connected to an all-metal vacuum line that includes two getters, one of titanium sponge and one of SAES NP10. There is also a stainless-steel mesh frit.

The walls of the chamber were heated to 325°C for at least 12 hours before the procedural blanks were run. The grid and the ion collector were degassed several times prior to use.

Procedural blanks were run before every experiment and blank corrections are only significant at the lowest voltages. All ion implantation experiments were done for one hour while the reaction chamber was exposed to a getter. The free phase was collected on a liquid-nitrogen-cooled, stainless steel frit. The reaction chamber was then closed. To collect the trapped phase, the ion collector was brought to a white-hot temperature and remained at this temperature until all of the trapped gas was released.

To test the effect of temperature on ion implantation, the reaction chamber body was stabilized at 200°C before the experiment. The ion collector potential was set at -100 eV and as before the experimental run time was one hour. This result was compared with a -100 V experiment done in the usual manner.

In all experiments the amount of argon that initially entered the reaction chamber was known. Within error

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limits all the gas was accounted for in either the trapped phase or the free phase.

Results for one-hour runs with -40 V to -100 V potentials on the ion collector are shown in Figure 1, and have been corrected for experimental blanks and mass discrimination.

Figure 1 reveals a large, uniform, $\geq 3\%$ per amu, isotopic fractionation for implanted argon at the -50 eV potential. The trapped fraction here is about 6.6% of the original aliquot. At -100 eV, the trapping efficiency for argon increases to 38% while the isotopic fractionation for the trapped phase drops to 1% per amu. Isotopic mass balance was achieved between the trapped and free phases in all cases. Of course, mass balance assignments require that the isotopic fractionation of the trapped phase will drop to zero as trapping efficiency approaches zero.

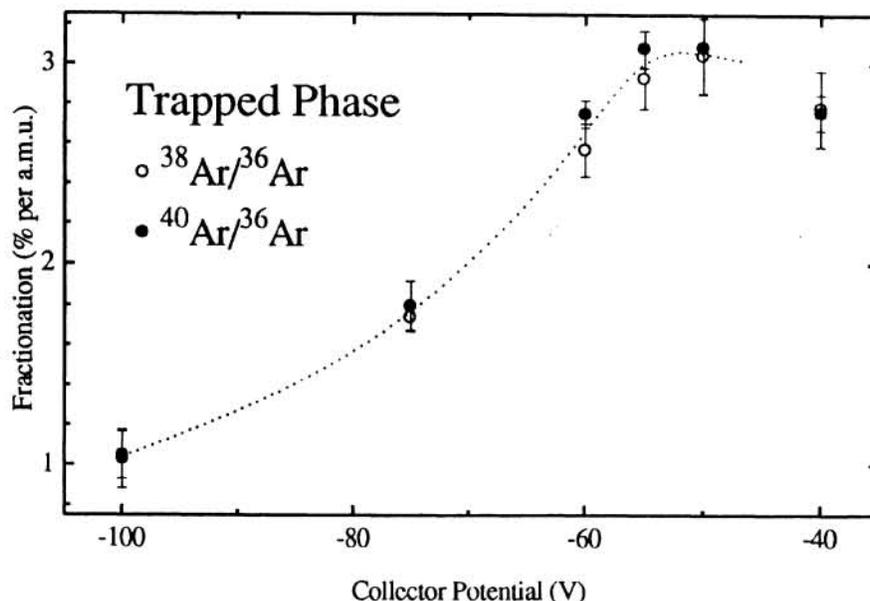


Figure 1: Mass fractionation of trapped argon as a function of ion collector potential with the filament at -30 eV and both the grid and walls at ground. At -50 eV, the trapped argon is 6.6% and at -100 eV, 38%. Mass fractionation is 3% per amu for -50 eV and 1% per amu for -100 eV.

The temperature dependence of argon ion implantation at -100 eV was studied in two experiments, with the ionization chamber initially at ambient temperature and at 200°C respectively before the ionization filament was turned on. The final temperature at the glass support for the feedthroughs was 73 °C in the former and 155 °C in the latter experiment. At 200°C the extent of argon trapping drops by a factor of five to $(5.58 \pm 0.05)\%$ while the isotopic fractionation is at most only marginally increased to $(1.32 \pm 0.09)\%$ /amu for $^{38}\text{Ar}/^{36}\text{Ar}$ and $(1.26 \pm 0.03)\%$ /amu for $^{40}\text{Ar}/^{36}\text{Ar}$.

Although it is difficult to assign fractionation effects to either ion implantation, gas sputtering or diffusive loss after ion implantation in these experiments, it is obvious that significant isotopic fractionation is produced by low-energy ion implantation. One of these processes or a combination of them has potential implication for the isotopic evolution of planetary atmospheres. The magnitude of fractionation in other gases needs to be assessed.

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