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The objective of this research is an improved understanding of the early solar system environment and of the processes involved in the nebula and in the evolution of solid bodies. We present results of our studies on the isotopic signatures in selected primitive solar system objects and on the evaluation of the cosmic ray records and of inferred collisional events. Furthermore, we report data of trapped martian atmospheric gases in meteorites and the inferred early evolution of Mars' atmosphere.

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FINAL REPORT

## 1. STUDIES OF NITROGEN ISOTOPIC SIGNATURES

Nitrogen ranks sixth in the solar system abundance table and isotopic signatures vary by over a factor of two. Do these variations represent primordial solar system inhomogeneities or do they indicate surviving nucleosynthetic carriers?

In order to resolve the problem of heterogeneous N isotopic signatures in the solar nebula we carried out the study of the N isotopic composition of primitive achondrites. These meteorites are of particular interest because they represent the link between primitive material and more highly differentiated matter like basaltic achondrites. They experienced thermal processing on a planetary body but are still primitive enough to identify the chemical and isotopic characteristics of their source regions within the solar nebula.

The Acapulco meteorite indicated a high degree of equilibration for major and trace elements. Heating to temperatures of (~1200°C) led to partial melting in a closed system. Studies of radiogenic isotopes (Sm-Nd, Pb-Pb) reflect a closed and well behaved system. However, large scale isotopic disequilibrium in N among different mineral phases has been observed in this meteorite. The first N analyses on Acapulco, using a static mass spectrometer, were carried out by stepwise pyrolysis. Variable N signatures indicate the presence of isotopically distinct N carriers. A more detailed study of mineral separates clearly indicates a light N isotopic composition in metal ($\delta^{15}N$ = -150‰) and heavier N in silicates and troilite ($\delta^{15}N$ = 0 to + 14‰). Variable isotopic signatures in the metal separate itself indicate the presence of a distinct, but still unresolved, N carrier.

Graphites of known textural relationship were separated from the IAB iron meteorite El Taco, a part of the Campo del Cielo meteorite shower. Irons of the IAB type are known to have silicate inclusions with approximately chondritic mineralogy, but achondritic texture. Along the interface of matrix metal and silicate inclusions, graphites form broad layers up to 200 mm in thickness. Graphites within the silicate inclusions often form rims around metal cores and silicate phases. The morphologies of individual graphite grains indicate several populations. Graphite concentrates were obtained by density separation from the silicate inclusion and the interface. Graphite associated with the matrix metal was obtained from an acid residue. The results indicate a range of isotopic signatures for graphites. Graphites (grain sizes up to 100 μm) separated from the broad graphite layers at the interface have a well defined isotopic compositions of $\delta^{15}N$ = -35‰. Graphite separates (grain sizes < 20μm) from both the silicate inclusion and the matrix metal have variable isotopic compositions with $\delta^{15}N$ values between -22 and -50‰. The N concentrations of El Taco graphite range from 5 ppm (interface) to 15 ppm (silicate inclusion and matrix metal residue). We determined the N isotopic composition of the matrix metal ($\delta^{15}N$ = -68).

Nitrogen was also studied in metal phases of IIE and IVA irons in a search for suggested possible links with H- and L- chondrites, respectively. Extraterrestrial samples exhibit a large range of bulk δ15N of unknown origins. The distribution, isotopic composition, and source of N in the metal and separated non-metallic phases of iron meteorites are likewise not well understood. Samples of IIE irons Colomera, Seymchan, Techado and Tobychan, and IVA irons Woods Mountain and Mart, were prepared. Stepped pyrolysis was employed to extract the gases, first in a quartz furnace with a resistance heater (up to 1120 °C), then up to the melting temperature in a molybdenum crucible with RF induction heating. Nitrogen was analyzed as $N_2$ by static mass spectrometry. In a few cases both large and small volume fractions were analyzed, with resulting isotopic signatures obtained being identical within experimental uncertainty. Excesses observed at mass 30 are due mainly to CO and were used to correct for CO at masses 28 and 29. N fractions were cleaned by exposure to CuO, converting background gases to $CO_2$ and $H_2O$ which were collected on an $LN_2$ trap and later discarded. Xenon abundances and compositions were measured as a means of detecting possible contributions from graphite and/or silicate inclusions.

The high temperature release patterns indicate well defines isotopic signatures, but small deviations may be due to spallation effects.

The indigenous N isotopic signatures from the metal phase of the IIE and IVA iron meteorites, in comparison to the signatures from H and L/LL chondrites provides a diagnostic tool for distinguishing genetic relationships among these groups. With the data at present, it appears that the trapped N composition in both classes of irons is approximately the same ($\delta^{15}N$ = -5%), despite significant $\Delta^{17}O$ differences in their silicates.

## 2. COSMIC RAY RECORDS

The Peekskill H6 meteorite presents an unusual opportunity to compare a photometric estimate of preatmospheric size with one inferred from cosmic-ray-produced nuclide concentrations. A consortium approach was organized to measure the stable and radioactive nuclides as well as tracks in order to study Peekskill's history of exposure to cosmic rays. The shielding corrected (via the $^{22}Ne/^{21}Ne$ ratio) average $^{3}He$, $^{21}Ne$, and $^{38}Ar$ exposure age of 25 Ma is considered to represent a lower limit. The $^{10}Be/^{21}Ne$ age is 32 My, and falls onto the peak in the H-chondrite exposure age distribution. The nuclide abundances $^{26}Al$, $^{14}C$, $^{36}Cl$, and $^{10}Be$ are all close to the maximum values expected for H-chondrites, but in contrast, the $^{60}Co$ activity requires a near-surface location and/or a much smaller body. A two-stage model can account for the data. We estimate an upper bound of 70 cm on the radius of the earlier stage of irradiation and conclude that Peekskill's radius was less than 70 cm when it entered the Earth's atmosphere. These size limits are comparable, but somewhat smaller than the photometric determinations.

In a collaborative study with K. Nishiizumi, R. Finkel and M. Caffee to characterize the 7 My stochastic event better, metal separates were prepared to determine $^{36}Cl$-$^{36}Ar$ exposure ages for six H4 afternoon falls and ten H5 morning falls. This dating method uses production rate ratios P ($^{36}Cl$)/P($^{36}Ar$) and is independent of the shielding-sensitive absolute production rates. It is also known that for protons the production rate ratio is rather insensitive to changes in the energy spectrum; the dependence of this ratio for secondary neutrons is at present less understood.
Interestingly, the cosmic-ray-produced $^{3}He/^{38}Ar$ ratios show a bimodal distribution with two clusters at ~15 and ~9. About half of the $^{3}He$ is produced via $^{3}H$ which is known to

diffuse in metal at relatively low temperatures. Therefore, these results provide evidence for a quasi-continuous loss of $^{3}He$ from such metals. If this loss mechanism is due to solar heating, perihelia <1 AU are indicated for these meteorites. Losses are prominent for H5 a.m. falls, but not for H4 p.m. falls. The orbital implications are consistent with those already known from the time-of-fall parameter (p.m. falls/total falls) which was used in the selection of the H4,H5 sample sets (Graf and Marti, 1995). The exposure age histograms of both H groups show the well known clusters at 7 Ma. The width of the exposure age peaks differ, however, and the collisional break-up event can be further constrained. Five out of six members of the H4 p.m. group cluster at $7.0\pm0.3$ My.

The asteroid belt is the ultimate source of iron meteorites and it is of considerable interest to obtain a chronology of break-ups of asteroidal objects. Meteorites which were fragmented in more than one collisional event have recorded integral effects of cosmic ray interactions in varying geometrical configuration and complex histories need to be evaluated. Exposure age histograms based on potassium ages indicated that irons of groups IIIA and IIIB reveal similar histograms and probably were derived from the same parent body, and a cluster for group IVA is also possible. In collaboration with B. Lavielle, we are using published and new noble gas data to re-evaluate collisional histories of the various groups, as well as, the evidence for a change in the cosmic ray flux. Unlike potassium ages which show large uncertainties for ages <300 Ma, argon (T38) ages are obtained also for short exposure times of iron meteorites. We confirm the evidence for stochastic events for IIIAB and IVA irons. The statistics are improved because of the larger data base. Recent reports of H-chondritic inclusions in IIE irons, whose exposure ages are consistent with H-chondrite clusters, may support the oxygen isotopic evidence for genetic link. There is good evidence that the collisional evolution of the IVA and IVB groups differ, as in group IVB there is a single exposure age at the position of the major peak of group IVA. It is interesting to note that the collisional event of the IVA group coincides with the time of the inferred collisional event of the L-chondrite parent object as recorded in $^{39}Ar$-$^{40}Ar$ chronologies of shocked L6 chondrites and that N isotopic signatures of group IVA members are consistent with those of L-chondrites.

Similarities in oxygen isotopic signatures between silicates in IIE irons and H-chondrites, as well as between IVA irons and L/LL - chondrites were noted and nitrogen measurements on samples of the same groups revealed similarities also for N isotopic signatures. These isotopic signatures play a role in the spectroscopic data interpretation leading to the suggestion that asteroid 6Hebe may be the parent body of both the H-chondrites and of IIE irons. In collaboration with B. Lavielle and K. Nishiizumi, we obtained a new cosmic-ray-exposure data, both noble gases and radionuclides, of group IIE and IVA members. Seven new $^{36}$Cl-$^{36}$Ar exposure ages of 260±20Ma differ from the $^{40}$K-$^{41}$K ages and need to be reconciled.

The noble gas records in IIE irons permit the recognition of the two subgroups, termed "old" and "young" in which reflect distinct thermal and irradiation histories. Irons in the "old" clan have radiometric ages >4Ga and exposure ages >50Ma, shorter than those of IVA irons. The "young" clan has not only young (>4Ga) radiometric ages, but very short exposure ages (<15Ma) which are close to those of the major collisional peak (7-8 Ma) of the H-chondrites although the exposure age distribution of this clan shows fine-structure.

## 3. XENON COMPONENTS IN MARTIAN METEORITES

Most SNC meteorites have young solidification ages which imply large-scale outgassing of the parent body during magmatic activity. Xe isotopic signatures observed in these meteorites not only have the potential of tracing the evolution of a planet's atmosphere but also might help to establish relationships among the solar system reservoirs. New measurements of N and Xe in ALH84001, EET79001 and Zagami provide additional constraints. Our results show varying $^{129}$Xe/$^{132}$Xe ratios due to component mixtures, but suggests that the Martian atmospheric Xe component evolved early on. Shifts exist in the heavy Xe isotopes due to additional fission components. The atmospheric component may be characterized as solar-type Xe, but strongly mass fractionated by 3.7% per amu.

Noble gas and N measurement in glass separates of EET79001 and Zagami provide strong supporting evidence for a Martian origin of these meteorites. Measured isotopic and elemental ratios of noble gases and N in these members of the SNC meteorites are strikingly similar to the Viking data of the Martian atmosphere. We carried out measurements on specific lithologies of Zagami, EET79001 and bulk ALH84001, and the Xe isotopic records reveal the presence of similar mass fractionated Martian atmospheric components in both samples, as well as a solar-type Xe component in ALH84001, released in varying mixing ratios in stepwise extractions.