NITROGEN, XENON AND ARGON IN LUNAR REGOLITH BRECCIA 60016: A STUDY OF SOLAR-WIND NITROGEN AND MEASURES OF ANTIQUITY

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If the ^{15}N content of solar-wind N has increased over the lifetime of the lunar regolith [1], ancient regolith breccias rich in "excess" fission Xe (from extinct ^{244}Pu) should contain some of the most ^{15}N-poor N available. However, a pioneering study by Fourcade & Clayton [2] of 14047, 14055 and 14307 yielded N with unexceptional $\delta^{15}N$ values, ranging from -51 to +5.8^o/oo, reducing to -78 to +0.2^o/oo after correction for spallogenic ^{15}N. Those values greatly exceed the lowest values (around -200^o/oo[3]) observed in some Apollo 11 and 17 breccias not noticeably rich in fission Xe. Fourcade & Clayton noted several possible explanations for these unexpected results, including the likelihood that their samples could have experienced complex irradiation histories resulting in solar wind and fissiogenic gas having been acquired at different epochs. The complex make-up and histories of individual constituents of those regolith breccias preclude a straightforward interpretation of results on such bulk samples. We are therefore searching within apparently ancient regolith breccias for lithic components with relatively simple irradiation histories: samples of these lithologies are then isotopically analysed for N, Xe and Ar, because both fission Xe and parentless ^{40}Ar can serve as measures of antiquity of surface exposure [1].

At this time we report on two key preliminary steps in this study. First, we are examining regolith breccia 60016 (shown by McKay et al. [4] to contain excess fission Xe) for fractions with comparatively simple irradiation histories. Specifically, we are focusing on plagioclase separates free of complex particles such as agglutinates and microbreccias [5]. Second, we are assessing the feasibility of disaggregating samples in an atmosphere of pure O_2 in order to minimise uptake of terrestrial N and noble gases, particularly Xe. We have developed experimental procedures which permit us to study N isotopic abundances in temperature steps that yield nanogram quantities and to study Xe and Ar isotopes in volume splits of the same gas fractions, see also [6]. The sample is placed within a tungsten basket contained in an extraction vessel which is surrounded by a separate vacuum jacket. The basket is then resistively heated to the appropriate extraction temperature.

Light-colored, presumably plagioclase-rich, clast 60016.240 was derinded in pure O_2. The rind, which has experienced curatorial N_2, will be compared with interior material, exposed only to O_2: analysis of that interior fraction is reported here. The sample was outgassed overnight at 200^{o}C and then extracted in three temperature steps: 400^{o}C in an externally heated sidearm, and then at 1400^{o}C and 1750^{o}C (well above its melting point) in the tungsten basket. Resulting gases were split, with the larger fraction used for Xe and Ar isotopes by standard procedures, and the other for $^{15}N/^{14}N$ analysis using the noble-gas mass spectrometer. Nitrogen data were corrected for CO background.

A very low N yield, 0.021ppm, at 400^{o}C suggests that disaggregation in pure O_2 has been quite successful in reducing atmospheric N_2 contamination. Our first attempt at identifying an appropriate lithology has been less successful. The low N and noble-gas concentrations at 1400^{o}C (N=1.85ppm; ^{36}Ar= $12x10^{-8}$ccSTP/g; $^{132}Xe=66x10^{-12}$ccSTP/g) suggest that this sample contains little record of solar-wind exposure, simple or otherwise. Not only is the apparent solar-wind fluence very low, its elemental systematics are abnormal. Compared with solar values, the noble-gas/N ratios (^{36}Ar/N=1.1% of solar;

^{132}Xe/N=3.4% of solar) are exaggeratedly low, even by lunar regolith standards [e.g.,6]. As a probe of the solar wind, ancient or modern, this lithology is clearly unsuitable. Compared with bulk 60016 [4], the noble-gas content of clast 240 is lower by a factor of about 5.

The modest N content is nonetheless significantly above estimates of indigenous lunar abundance [7], showing that some extralunar, presumably solar-wind, N is present. Correcting our observed δ^{15}N value of +3.5^o/oo for spallogenic ^{15}N, calculated from the ^{38}Ar-derived cosmic-ray exposure age, see below, leads to a trapped δ^{15}N value of +1.9^o/oo. Thus, this N resembles that found by Fourcade & Clayton [2]. It is possible that our observed ^{15}N/^{14}N ratio may have been enhanced by diffusive loss of ^{14}N during a thermal degassing episode on the moon, though little evidence for such mass fractionation has been observed. The N yield on melting was not sensibly different from the estimated blank level at that temperature: a higher-resolution stepwise extraction is in progress.

The Ar released above 400^{o}C shows clear evidence for excess ^{40}Ar (^{40}Ar/^{36}Ar=83.94 at 1400^{o}C and about 270 at 1750^{o}C). Release of ^{38}Ar at 1400^{o}C (^{38}Ar/^{36}Ar=0.2157) and 1750^{o}C (^{38}Ar/^{36}Ar=0.312) corresponds to a few Myr cosmic-ray exposure, possibly a combination of South Ray Crater ejecta exposure with some preirradiation. The ratios ^{40}Ar/^{38}Ar$_{spall}$ in the 1400^{o}C and 1750^{o}C temperature steps are similar, indicating that both the radiogenic and spallogenic Ar components are released by volume diffusion from the same mineral. This evidence suggests that ^{40}Ar represents predominantly a radiogenic component produced *in situ*.

The ^{136}Xe/^{132}Xe ratio of clast 240 is within 15% of that measured for bulk 60016 [4], but given the small quantity of trapped Xe in the clast, the absolute amount of fission Xe in 240 is also much lower than that in the bulk. The amount of ^{136}Xe (1x10^{-12}cc/g) released at 1750^{o}C would be consistent with *in situ* decay of U over about 4Gyr, using the U content of bulk 60016. However, the U content of clast 240 is almost certainly less than the bulk value [H.Wänke, pers. comm.], so that some Xe from ^{244}Pu decay may be present, presumably from *in situ* decay in view of its high release temperature. "Excess" fission Xe may also be present in the 1400^{o}C fraction (^{136}Xe/^{132}Xe= 0.3266) but we first need to assess the isotopic signature of trapped Xe. No evidence for excess radiogenic ^{129}Xe was observed in clast 240.

Following assessment of the disaggregation procedure in pure O_2, we shall continue our search for a lithology that has experienced a solar-wind fluence somewhat greater than that of clast 240 but still sufficiently modest to make a simple irradiation history plausible.

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