A LONG-TERM CHANGE OF THE AR/KR/XE FRACTIONATION IN THE SOLAR CORPUSCULAR RADIATION; R. Wieler, H. Baur, and P. Signer, ETH-Zürich, NO C61, CH-8092 Zürich, Switzerland

Abstract: Solar noble gases in an ilmenite separate from breccia 79035 (antiquity > 1 Ga) were analyzed by closed system stepped etching (CSSE). All five gases show the familiar two-component structure: first solar-wind (SW) gases are released, followed by gases from solar energetic particles (SEP). Element patterns in 79035 are similar to those of 71501 ilmenite (ref. 1; antiquity <100 Ma). SW-He-Ne were partly lost, but SEP-He-Ne-Ar are retained (nearly) unfractionated. Constant Ar/Kr/Xe ratios indicate that ilmenites contain an unfractionated sample of the heavy SW-SEP noble gases. Ar/Kr/Xe ratios in the solar corpuscular radiation are, however, different from "solar system" values, whereby the Kr/Xe difference in 79035 is about twice as large as in 71501. We propose that Xe is less fractionated than Kr and Ar, though its first ionisation potential (FIP) is higher than the "cutoff" at ~11.5eV, above which all elements in SEP are usually assumed to be depleted by a roughly constant factor. SW-Ne may be isotopically slightly heavier in the ancient SW trapped by 79035, as proposed earlier (2,3).

In this work we extend our previous CSSE studies (1) of solar noble gases *including Kr and Xe* to a lunar sample irradiated at least 1 Ga ago (breccia 79035, ilmenite separate, 42-64μm). This sample was particularly gently etched in the first steps. Surprisingly, the first three steps, each releasing ≤0.5‰ of the total 36-Ar, showed an SEP-like trapped component plus relatively large concentrations of cosmogenic gases. Steps 4ff contain much less cosmogenic and more solar gas with a SW-like isotope pattern. Thus, a very minor easily etchable phase that has completely lost its SW-gases must be responsible for steps 1-3. We will not discuss these steps here and refer to the actual step 4 as the "initial" etching step.

Element ratios: 4-He/36-Ar and 20-Ne/36-Ar patterns (Fig. 1) are very similar to earlier ones: ratios are below SW values (4) in the first steps but approach or reach SW ratios in the second half of the gas release. This indicates fractionating loss of solar wind He-Ne but (nearly) unfractionated retention of SEP-He-Ne-Ar in ilmenites (1). Unlike He/Ar and Ne/Ar, the Ar/Kr and Kr/Xe ratios are almost constant in 79035, as in 71501 (1). However, the average 84-Kr/132-Xe in 79035 is about 50% lower than in 71501 (4.75 vs. 8.97). Similar differences were previously reported (5) and interpreted to reflect possibly a variable Xe abundance in the SW (6,3). The present data offer strong additional support to this view: We argued (1) that the constant Ar/Kr/Xe ratios in CSSE runs, together with the conclusion that even the light gases He and Ne are not fractionated in the later steps, clearly suggest that the heavy gases in ilmenites are unfractionated samples both of the incoming SW and SEP. Ar/Kr/Xe ratios, however, do not agree with tabulated solar system values (ref.7, "SS" in Fig. 1). If ilmenite indeed retains unfractionated SW- and SEP-Ar-Kr-Xe, these gases seem thus to fractionate in the corona. The Kr/Xe fractionation factor thereby decreased about twofold since breccia 79035 was irradiated. Note that we postulate that Kr/Xe below SS in ilmenite is *not* due to a noble gas loss on the Moon. We find no evidence for SS-like Kr/Xe in the early release fractions as has been postulated for stepped combustion/pyrolysis runs (8). Our highest 4-He/36-Ar ratios in 79035 do not exceed the highest values in 71501, but the maximum is reached much earlier in the breccia than in the soil. We thus believe that the higher bulk 4-He/36-Ar of 79035 reflects a relatively smaller loss of SW-He from this sample rather than a higher 4-He/36-Ar ratio in the ancient SW (3).

Fig. 2 shows Ar-Kr-Xe fractionation factors relative to SS values versus their first ionisation potentials (FIP). All noble gases have FIP above 11.5 eV, the usually assumed "cutoff" value for FIP-related fractionation in SEP. Thus FIP may not be the only relevant parameter, or the cutoff seems to be larger for noble gases. To explain the different Kr/Xe ratios in ancient and recently exposed samples, Wiens et al. (9) proposed that the FIP cutoff might have been higher in the ancient sun.

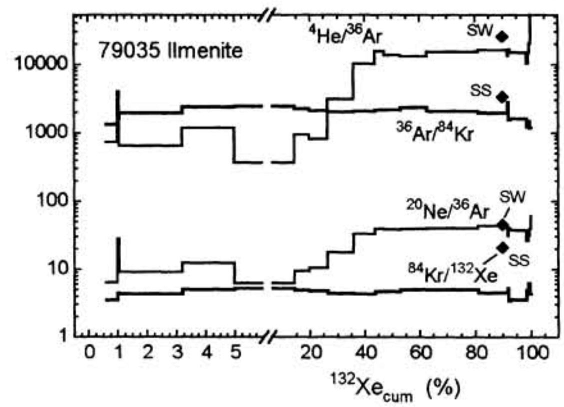

Fig. 1: 4-He/36-Ar and 20-Ne/36-Ar approach solar wind (SW, ref. 4) values in the last etching steps. This indicates that SEP-He-Ne-Ar - *and by inference also solar Kr and Xe* - is unfractionated in lunar ilmenites. 36-Ar/84-Kr and 84-Kr/132-Xe ratios are constant throughout the run but below "solar system" (SS, ref. 7) ratios, suggesting noble gas fractionation in the solar corpuscular radiation.

He-Ne-Ar isotopes: We concluded (10, 1) that ilmenites release in the first CSSE steps isotopically unfractionated SW noble gases, because recently irradiated 71501 ilmenite shows in the first steps isotopic compositions identical to modern SW. The more ancient 79035 ilmenite thus should allow us to rule on the long term constancy/variability of the isotopic composition of SW noble gases (3). Both samples show essentially the same 20-Ne/22-Ne value of 13.8 in their first step. However, in 79035 this first step comprises a mere 3.8‰ of the total 132-Xe, compared to 13% in the case of 71501. Steps 2-4 of 79035 (accounting for 4% of the total Xe) form a "plateau" at 20-Ne/22-Ne = 13.5. This value may thus represent SW-Ne in 79035 ilmenite, possibly showing a secular decrease of this ratio in the past >1Ga, as has often been postulated (e. g. 2, 3). The high ratio of 13.8 in step 1 may either be due to a minor contamination with recently irradiated soil (R. H. Becker, pers. comm.) or reflect a lower penetration depth of the lighter Ne isotope. We prefer the first explanation, since ratios light/heavy element are not enhanced in step 1.

Measured 3-He/4-He in the first 79035 steps is within ±8% of the SW value derived from 71501 ilmenite (1,10). 3-He/4-He in the SW to be inferred from these data is, however, rather uncertain due to cosmogenic 3-He. A possible secular increase of this ratio (2,3) is confined by these and earlier 79035 CSSE data (1,10) to less than about 7%. 36-Ar/38-Ar in the SW in 71501 and 79035 are indistinguishable.

20-Ne/22-Ne of SEP-Ne in 79035 is ~11.1, slightly lower than the value of 11.2 in 71501. Both, 3-He/4-He and 36-Ar/38-Ar in SEP in 79035 are similar to the 71501 values (1,10) but less well determined due to rather large corrections for cosmogenic gases.

Kr-Xe isotopes: The CSSE steps with pure SW-Ne should also allow us to determine the isotopic composition of SW-Kr-Xe. SW-Kr in 71501 ilmenite is very slightly heavier than atmospheric Kr (86-Kr/84-Kr = 0.3041, ref. 1), i. e. heavier than most current estimates for SW-Kr (e. g. 11). In 79035 we now determine 86-Kr/84-Kr = 0.3018, lighter by ~0.4‰ per amu than the 71501 value and close to the range preferred in (11). Hence, the isotopic composition of SW-Kr possibly also changed since the time 79035 was irradiated. Surprisingly, however, SW-Kr would have become isotopically heavier, unlike SW-He and -Ne. Both, Kr and Xe in 79035 get isotopically heavier with increasing step number, analogous to He-Ne-Ar and similar to 71501. This corroborates the existence of (heavy) SEP-Kr-Xe inferred in (1).

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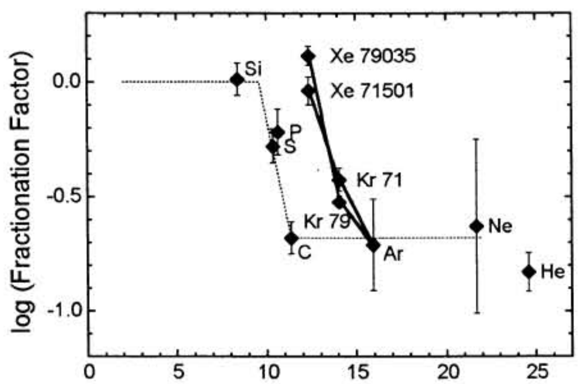

Fig. 2: Fractionation factors of solar Ar, Kr, and Xe in two lunar ilmenites, relative to solar system values (7). Kr and Xe points anchored to Ar as given in (7), data of other elements also from (7). Kr and Xe errors reflect 2σ of mean of 36-Ar/84-Kr and 36-Ar/132-Xe in main CSSE steps, but not uncertainty of Ar/Si fractionation. Xe/Kr in the ancient solar corpuscular radiation sampled by 79035 is twice as high as in recently irradiated soil 71501. The indicated FIP "cutoff" seems too low to explain the noble gas data.

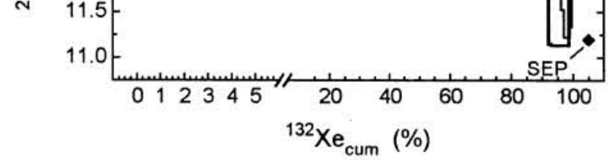

Fig. 3: 20-Ne/22-Ne of solar component versus cumulative fraction of 132-Xe released. Breccia 79035 indicates a ~2% lower 20-Ne/22-Ne in the ancient solar wind, compared to recently irradiated soil 71501 (the first step of 79035 may show contamination with recently irradiated fine dust). Both samples show the presence of SEP-Ne with 20-Ne/22-Ne = 11.1-11.2.