

DEUTERIUM CONTENT OF LUNAR MATERIAL, L. Merlivat, M. Lelu, G. Nief, E. Roth, Département Recherche et Analyse, C.E.N. Saclay, 91190 Gif-sur-Yvette, France.

Previous experiments had shown that extraction of hydrogen gas from lunar material was not complete when degassing under vacuum. It is limited by an oxydation reaction starting at 575°C (1). In order to overcome this difficulty we have built a new extraction line, based on a flow system, working at atmospheric pressure.

The extracted gases are carried by helium. The flow value is 50 cc/mn. A typical line includes two successive traps. In the first one, gases condensable at liquid nitrogen temperature are retained. The second one, is kept also at liquid nitrogen temperature and, in addition is filled with charcoal. In this last trap, hydrogen is collected. We have four lines of this type. Each of these is used in a separate range of extraction temperature. The samples are heated up to 1200°C in quartz holder by a resistor oven. A constant heating rate of 4°C/mn is adopted. Degassing between 150 and 200°C is performed for 90 minutes before the gas collection. A thirty minutes period at 1200°C completes the whole heating. When the extraction procedure is completed, helium is evacuated from the whole of the installation. Then the gas released from the traps react successively with vanadium at 800°C. Vanadium acts as a chemical trap for all compounds except rare gases and hydrogen-namely, H₂O is reduced in hydrogen. Then, hydrogen gas samples are transferred to ampoules filled with charcoal at liquid nitrogen temperature. The samples are ready for analysis by mass spectrometry. Before each experiment the quartz tube is degassed at 1200°C. Afterwards, at this same temperature at which pollution by grease should most likely occur, blanks are run with free helium. The amounts of hydrogen and water collected are always smaller than 0.3 mm³ S.T.P.

Experimental results obtained using the flow extraction system are reported in table I. In table II the results from experiments performed with the vacuum extraction line are given. Sample 14003 consists of fines. In this table, the experiments for which the maximum extraction temperature is 800°C are singled out with an, a or a'. During experiments labelled with b the range of temperature has been extended to 1200°C. To prevent pollution by grease craking, the experimental set up has had to be slightly modified in such a way that water extracted from the samples could not be collected.

Discrepancies are observed between the deuterium content of hydrogen extracted from a same sample in the two techniques. We always measure lower values when we use the vacuum technique. Experiments are underway to elucidate this problem. However the following conclusions are independant of this fact because of a different order of magnitude.

DEUTERIUM CONTENT

Merlivat L. et al.

1. In all runs except the first one, the amount of water extracted by the flow system is much smaller than it is under vacuum extraction conditions and the opposite is true for the hydrogen quantities. This correlates directly with the previous observations that under vacuum oxydation of hydrogen to water in presence of lunar material takes place. More precisely, the three experiments run with the fine sample 14003 have been made for purpose to check if the nature of the material used to wrap the moon sample could catalyse the oxydation reaction. Experiments run with gold (a') instead of platinum (a) don't lead to drastic changes. Conversely in the flow system, the pressure of 760 torrs of helium lowers the probability of reaction of the extracted hydrogen with the oxydes of the lunar sample. The deuterium content of water measured in both types of experiments is in general agreement with the previous observations. Water extracted from the samples show, in table I, values around 140 ppm. Values around 110 ppm in table II correspond to a mixture of water extracted as such and water formed by combustion of deuterium depleted hydrogen.

2. In both extraction procedures, the deuterium content of hydrogen extracted from fines and breccia displays a typical pattern. The second extracted fraction ($\sim 460^{\circ}$ - 725°) is always depleted in deuterium with respect to the first one and the third one (table I). We tentatively explain the relatively higher value observed for the first fraction as the result of isotopic exchange between hydrogen and deuterium rich water. The main part of the latter is extracted when heating in the low temperature range. The second fraction represents mainly solar wind hydrogen which is known to be very low in deuterium (1,2,3). In the third fraction, deuterium formed by nuclear reactions mainly by spallation processes, is the origin of the increase in values of D/H ratios. For instance, during experiments on a basalt sample, 75.035, a very high value of the deuterium content, 258 ppm, has been obtained in the hydrogen extracted between 725°C and 995°C . Such a value can in no case be explained by exchange or direct contamination with terrestrial water.

3. We are still confronted with the problem of the origin of the water extracted from the samples. Contamination cannot be ruled out as its deuterium content is very similar to terrestrial water, though relatively high. The values around 155 ppm could be due to an isotopic enrichment accompanying an adsorption process on the lunar material.

In conclusion, though the origin of water collected is still uncertain, evidence for deuterium formed by nuclear reaction on the moon has been given.

REFERENCES : (1) Merlivat L., Nief G., Roth E. (1972) Proc. 3rd Lunar Sci. Conf. Vol.2, 1473-1477 ; (2) Epstein S. and Taylor H.P., Jr. (1972) Proc. 3rd Lunar Sci. Conf. Vol.2, 1429-1454 ; (3) Friedman I., Hardcastle K.G., Gleason J.D. (1972) Apollo 15 samples 302-306, L.S.I Publ.

DEUTERIUM CONTENT

Merlivat L. et al.

Table I

| Sample | H ₂ μmoles/g | (D/H) H ₂ ppm | H ₂ O μmoles/g | (D/H) H ₂ O ppm |
|---------------------------------|----------------------------|--------------------------|------------------------------|----------------------------|
| 15299, m=0.755 g - rock breccia | | | | |
| a. 195 - 455°C | 1.1 | 71.5 | 8.5 | 151.8 |
| b. 455 - 690°C | 16.8 | 42.7 | 3.5 | 144.6 |
| c. 690 - 995°C | 1.3 | 70.7 | 0.3 | 120.3 |
| d. 995 - 1200°C | 0.2 | 105.4 | 0.4 | 109.4 |
| whole sample | 19.4 | 46.8 | 12.7 | 147.7 |
| 15299, m=0.827 g - rock breccia | | | | |
| a. 200 - 455°C | 1.1 | 59.5 | n.d | n.d |
| b. 455 - 725°C | 14.8 | 37.2 | n.d | n.d |
| c. 725 - 990°C | 1.2 | 71.2 | 0.3 | 119.6 |
| d. 990 - 1200°C | 0.03 | 110.8 | 0.1 | 116.4 |
| whole sample | 17.1 | 41.2 | - | - |
| 15600, m=0.588 g - fines | | | | |
| a. 200 - 450°C | 3.9 | 39.3 | 2.1 | 144.3 |
| b. 450 - 725°C | 12.8 | 27.0 | 0.5 | 155.6 |
| c. 725 - 990°C | 1.0 | 122.5 | <0.01 | - |
| d. 990 - 1200°C | 0.2 | 112.6 | <0.01 | - |
| whole sample | 17.9 | 36.0 | 2.6 | 146.5 |
| 15600, m=0.536 g - fines | | | | |
| a. 160 - 380°C | 0.9 | 67.6 | 3.2 | 140.4 |
| a'. 380 - 460°C | 2.0 | 33.3 | <0.01 | - |
| b. 460 - 720°C | 10.3 | 27.5 | 0.6 | 143.6 |
| c. 720 - 1000°C | 0.7 | 124.3 | <0.01 | - |
| whole sample | 13.9 | 35.8 | 3.8 | 140.9 |
| 68501, m=0.546 g - fines | | | | |
| a. 160 - 460°C | 3.2 | 43.7 | 4.1 | 140.3 |
| b. 460 - 735°C | 12.0 | 36.9 | 1.1 | 129.2 |
| c. 735 - 985°C | 1.6 | 95.5 | <0.01 | - |
| d. 985 - 1200°C | 0.3 | 87.5 | <0.01 | - |
| whole sample | 17.1 | 44.5 | 5.2 | 137.9 |
| 75035, m=0.653 g - basalt | | | | |
| a. 170 - 470°C | 0.04 | 120.3 | 2.22 | 140.2 |
| b. 470 - 725°C | 0.19 | 170.5 | 0.26 | 151.7 |
| c. 725 - 995°C | 0.07 | 258.5 | <0.01 | - |
| d. 995 - 1200°C | 0.02 | 129.6 | <0.01 | - |
| whole sample | 0.32 | 180.9 | 2.48 | 141.4 |

Table II

| | | | | |
|-----------------------|------|------|------|-------|
| 14003, m=0.230 g (a') | 18.8 | 31.3 | 12.3 | 96.5 |
| 14003, m=0.242 g (a') | 20.1 | 30.7 | 16.7 | 115.1 |
| 14003, m=0.209 g (a) | 21.6 | 31.9 | 19.8 | 112.5 |
| 15600, m=0.223 g (a) | 14.6 | n.d | 10.0 | 110.5 |
| 15600, m=0.254 g (a) | 12.0 | 24.8 | 14.6 | 114.6 |
| 15299, m=1.925 g (b) | 8.1 | 29.3 | - | - |
| 15299, m=0.759 g (b) | 8.5 | 29.1 | - | - |
| 68501, m=0.372 g (b) | 12.1 | 27.0 | - | - |