

RADIOHALOS, TEKTITES AND LUNAR RADIOACTIVE CHRONOLOGY, R. V. Gentry, Chemistry Division, Oak Ridge National Laboratory*, Oak Ridge, Tenn., 37830.

The rationale for the lunar halo search is that halos in terrestrial minerals serve as pointers to localized radioactivity and thereby make possible analytical studies that bear importantly on the general problems of isotopic dating and mode of crystallization of the host mineral. For example, several minerals such as mica, fluorite and cordierite often contain unusual halos originating with fairly short half-life alpha radioactivity, e.g., ^{218}Po ($t_{1/2} = 3$ min), and it is not clear how the presence of these halos can be reconciled with a hydrothermal, magmatic, or metamorphic origin of the host minerals (1). Therefore, a further aim was to pursue such ancillary studies on terrestrial halos and on certain samples of special origin (e.g., tektites, meteorites).

With respect to the first aim, the MSC thin section collection from the Apollo 11-15 suite of rocks has been personally scanned for halos on several occasions with completely negative results. It is difficult to estimate the probability that this means a real absence of halos because the only basis for such an estimate, their terrestrial occurrence, is exceedingly spotty. Further, terrestrial halos are most numerous in rock types that are generally conspicuously absent from the lunar samples thus far available for study. The absence of halos is not due to a lack of radioactive inclusions in lunar samples, since such have been well identified in rocks such as 12013. Perhaps the high radiation environment indigenous to the lunar surface effectively anneals the halos as they are being formed.

In parallel with the search for halos in lunar material, certain types of terrestrial halos have been examined for information on the general field of radioactive chronology. In particular, we have used the ion-microprobe mass spectrometer to determine lead isotope ratios in the microscopic inclusions within certain terrestrial radiohalos that were expected to possess unusual Pb isotope abundances (1). Specifically, one inclusion showed Pb/U and Pb/Th ratios > 5000 with $^{206}\text{Pb}/^{204}\text{Pb} > 1000$, i.e., no U, Th or ^{204}Pb detected. In another case, the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio was ≈ 60 , an impossibly high value if the Pb isotopes had been derived from uranium decay. Clearly, any attempt to date this sample in the usual way would be impossible, and it appears desirable to establish the extent of such anomalies in lunar and other terrestrial samples.

While their exact origin and history is uncertain, iron meteorites are important in radioactive chronology as reference material for defining primordial Pb isotope ratios. Over the last decade, several analyses have shown modern radiogenic lead in addition to the primordial mixture in the troilite phase of a number of iron meteorites (2). These anomalous results have usually been attributed to terrestrial contamination (3), but in order to settle the question unambiguously, we have initiated ion-probe Pb isotope studies of this class of samples. To evaluate the potential of the ion-probe to perform accurate Pb isotope ratios on the meteorites, NBS common lead was utilized as a

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standard in test runs. A comparison of ion-probe (IP) runs at Oak Ridge with the NBS reference data for their sample yielded the following results (4); $^{204}\text{Pb}/^{206}\text{Pb} = 0.0591 \pm 0.0074$ (IP), 0.059042 ± 0.000037 (NBS); $^{207}\text{Pb}/^{206}\text{Pb} = 0.909 \pm 0.025$ (IP), 0.91464 ± 0.00033 (NBS); $^{208}\text{Pb}/^{206}\text{Pb} = 2.174 \pm 0.035$ (IP), 2.1681 ± 0.0008 (NBS).

Another part of the study was to have been a comparison of lunar and terrestrial U and Th halo sizes in relation to the fundamental question treated more than 30 years ago by Dirac (5) and more recently by Hoyle and Narlikar (6), i.e., that of the possible time variation of the fundamental physical constants and in particular of the radioactive transformation rate over geological time. Due to the absence of lunar specimens comparative studies were not possible, yet terrestrial halo investigations have yielded some data that may bear considerably on lunar radioactive chronology. As background information I note that in the context of the Dirac hypothesis, Gerling and Ovchinnokova (7) have recently reported differences in rock ages as measured by various age-dating techniques, a situation which they attribute to a variation in the alpha-beta branching ratio in the Precambrian era. A few years ago Gamow (8) proposed using the ring structure of uranium halos to check branching ratios in the Precambrian, not realizing that Wilkinson (9) had earlier interpreted the same data (somewhat erroneously) in favor of confirming presently accepted values. Interestingly, the same data which for decades have been utilized to establish decay rate invariance and branching ratio constancy are now being interpreted to imply a variation in the radioactive decay rate (10). Clearly the same data cannot support both conclusions!

From α -decay theory it has been argued previously that the near agreement between uranium and thorium halo ring radii and computed alpha ranges in the same mineral was proof of an invariant decay rate (11). To see this is not exactly true we use the α -decay theory notation given in (12) to compute the fractional change in the decay constant (λ), arising from the fractional change in ring radius (r). In this case $\ln T \approx 3Z^2R^2 - 4ZE^{-2}$; $\lambda = \nu T$ where T is the transmission probability, E the energy of the emitted α -particle, Z the atomic number of the daughter nucleus, ν is the frequency with which the α particle strikes the barrier, and R is the nuclear radius. It then follows that in the appropriate units $d\lambda/\lambda \approx [(3/2)Z^2R^2 - 1](dR/R) + [\frac{1}{2} + 2Z/E^2]X(dE/E)$. A particle of mass m_1 and charge z_1 has a range r (halo radius) given approximately by the expression $r = \text{const. } E^2/Z_1^2 m_1$. Subsequent calculations lead to the expression $d\lambda/\lambda \approx 43(dR/R) + 46(dr/r)$. Since the minimum uncertainty in measuring halo radii is about $\Delta r = 0.1 \mu\text{m}$, then the resulting fractional uncertainty in λ for ^{238}U (halo radius $12.5 \mu\text{m}$) is $d\lambda/\lambda \approx 46(0.2/12.5) \approx 1/2$ assuming $dR = 0$. Since some uncertainties are involved in the conversion of air to mineral ranges, Van de Graaff He^{++} ion beams of varying energy were utilized to induce coloration bands in the mineral containing the halos. In examining scores of U and Th halos and in attempting to match their ring radii with the corresponding ranges obtained from the experimentally determined range-energy curve, I have found occasional rather than systematic differences in certain halo ring radii. Since the Van de Graaff induced ranges correlated closely with halo radii at 4.2 Mev ($13 \pm 0.5 \mu\text{m}$ V.d.G. vs $12.7 \pm 0.3 \mu\text{m}$ for halo radii) and 5.3 Mev ($19 \pm 0.5 \mu\text{m}$ V.d.G. vs $18.9 \pm 0.5 \mu\text{m}$ for halo radii), at present I do not attribute such differences to an actual change in E_α .

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An unanswered question is whether λ variations might occur without changes in halo radius ($dE = 0$). Wilkinson (9) has suggested that the physical constants might vary in some unknown fashion without affecting λ . The correlative of this suggestion would imply the correlation of measured α -ranges and halo radii is not a test of λ invariance. Variations in λ might then be detected by noting age differences in samples determined by radiometric, geologic, or archeologic methods. As a case in point, there is a wide discrepancy between the radiometric (13,14) ages (0.7 my and 4 my) and the geologic and stratigraphic (15,16) age (several thousand years) of the australites. That two cosmic events separated by several million years would result in similar stream-field tektite patterns is in itself most unusual irrespective of whether the tektites are of lunar or terrestrial origin (17). Evidence suggesting a common link between the H/Na (4 my) australites and the general australite population comes from recent Th/U determinations at ORNL (18), i.e., Th/U = 5.9 (AN-87) and 8.3 (P-192) in the H/Na group compared to Th/U values of 4.8 - 8.5 previously reported for the general group (19).

It has been remarked that the fission track age of the new H/Na group of australites may be high for some reason (17). This is quite significant in that it is universally agreed that fission tracks can form only after the tektites have cooled. Unless the australite fossil fission tracks originated with a spontaneous fissioning nuclide besides ^{238}U , the only remaining possibility for a "high" fission track age is a hiatus in the decay rate due to causes presently unknown. While this explanation is unusual, I do not think it can be definitely ruled out by simply appealing to the regularity of radihalo data.

Acceptance of this hypothesis would not necessarily have implied significant changes in the other fundamental constants but would affect age dating.

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