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A study of the radioactive decay law

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Abstract. The decay of radon has been followed in effect over approximately 40 halflives in an effort to detect any deviation from the exponential decay law. The results were fitted to a decay of the form $N = N_0 \exp\{-\lambda t(1+bt)\}$ and b was calculated to be $(1{\cdot}9 \pm 1{\cdot}0) \times 10^{-6}\,\mathrm{d}^{-1}$; the halflife was determined to be $3{\cdot}8235_1 \pm 0{\cdot}0003_4$ d.

1. Introduction

In several papers (Hellund 1953, Namiki and Mugebayashi 1953, Khalfin 1958, Winter 1961 and Osborne 1966) theoretical attention has been drawn to nonexponential components of α decay. According to present theory, the nonexponential component does not become significant until after the passage of many halflives. However, Winter (1962) has suggested that the presence of hidden variables associated with the nucleus might be indicated by nonexponential decay. He reported measurements on the β decay of ^{54}Mn over 34 halflives. No nonexponential effect was detected, although the statistics at the end of the measurement were necessarily poor. Bohm (1971, private communication) has also suggested that hidden variables might be manifest in nuclear decay although he felt that nuclear decay might be too complex a process for ideal study.

2. Method

This paper describes a measurement of the α decay of radon extending effectively over 40 halflives, from which a limit on any deviation from exponential decay has been calculated. Radon was chosen for several important reasons. The presence of impurities in the radioactive source will, in general, set a limit to the length of time over which the decay can be measured and also the precision. Radon emanation, when condensed, is virtually free from all radioactive impurities except its own decay products. The lifetimes of radium A, B and C are short compared with radon so they follow the radon decay in transient equilibrium. In this experiment the γ radiation of radium C was measured in preference to the radon α rays themselves. Radium D is the first long-lived product with a halflife of 22 years; its radiations are soft. Radium E is virtually a pure β emitter and in the case of γ ray counting only contributes via bremsstrahlung; radium F (polonium) is an α emitter. In the present high energy γ ray measurements none of the radiations from these decay products was detected.

The influence of variations in long term instrumental stability was minimized in the present experiment by comparing the decay of an 'aged' radon source with that of a 'fresh' source of approximately the same activity. The 'aged' source was obtained by

allowing one curie of radon to decay to approximately one microcurie; this microcurie was then condensed and sealed into a small gold tube leaving the decay products of the curie behind on the walls of the original container. A source of one microcurie of 'fresh' radon was prepared in a second tube. The γ activity of each source was measured daily for the first 55 days and then less frequently as the activities became very small and counting times of up to one day were required to achieve good statistics. The sources were counted in the well of a 3 in $\times$ 3 in sodium iodide crystal contained in a 1 in thick lead castle and the activity of the radon was measured as the counts in the energy range 1·58 MeV to 2·45 MeV spanning the full energy peaks of the 1·76 MeV and 2·20 MeV γ rays of radium C. As the two sources decayed so the background count became increasingly significant. The background was monitored every two or three days. Small variations in it of up to $\pm 0{\cdot}3$ cpm (in approximately 28 cpm) were detected and these were attributed to varying levels of radon decay products in the environment.

3. Results and discussion

Figure 1 shows the decay curves for the two sources on a logarithmic scale expressed in terms of the time from which the first measurements were made. At that time, the 'aged' source had already decayed over 75 days so that the 76 days over which the decay was followed represents the decay over the second 20 halflives of a 40 halflife period. The two straight lines are weighted least squares fits of an exponential to the data and correspond to halflives of $3{\cdot}8232_5 \pm 0{\cdot}0004_7$ d and $3{\cdot}8237_8 \pm 0{\cdot}0004_8$ d for the 'aged' and 'fresh' radon sources respectively. The mean value of the halflives of the two sources of $3{\cdot}8235_1 \pm 0{\cdot}0003_4$ d agrees with other determinations (Marin 1956, Tobailem 1955).

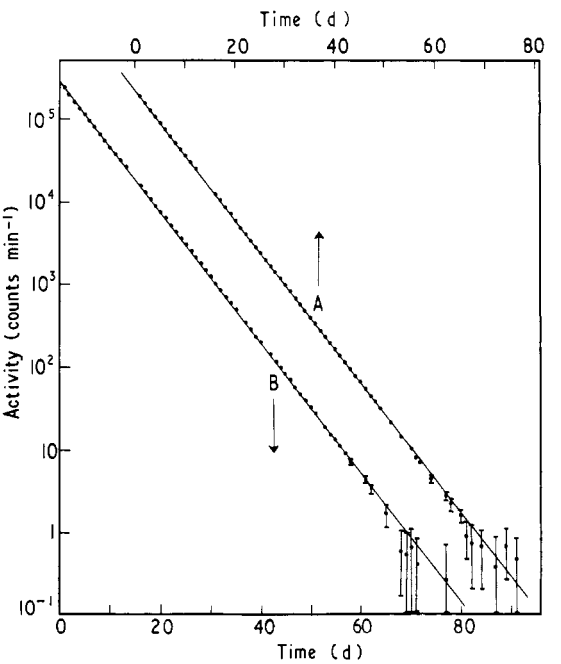

Figure 1. Decay curves for the 'aged' (A) and 'fresh' (B) radon sources.

In the tail of the decay the statistics are poor and the limit to which exponential decay can be verified depends upon the criterion adopted to signify a deviation. There is a suggestion of a deviation in the tails of the decay curves for both the 'aged' and the 'fresh' sources. This characteristic, common to both decay curves, must be due to a small systematic error in counting, rather than to a significant deviation from exponential decay. Figure 2 shows the ratio: activity of 'fresh' source to activity of 'aged' source as a function of time; the lower data cover the time up to 70 days and the upper data are for the first 50 days on an expanded ordinate scale. The straight lines correspond to the weighted mean ratio of activities of $1{\cdot}2657_8 \pm 0{\cdot}0007_0$ for the first 10 days.

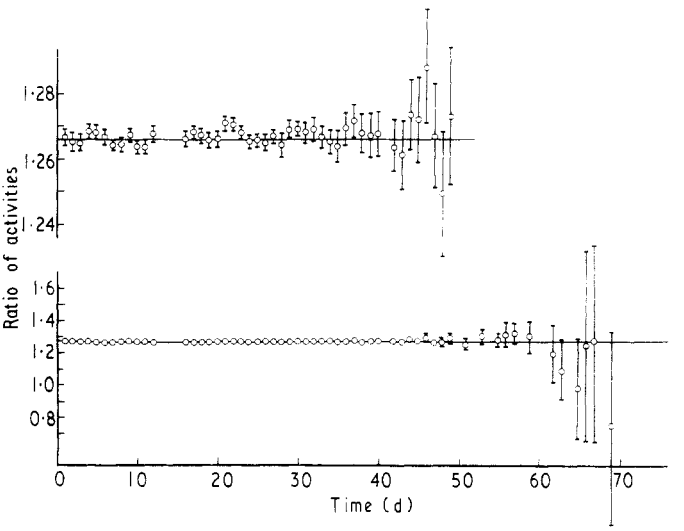

Figure 2. Ratio: activity of 'fresh' source to activity of 'aged' source as a function of time. The lower data are the ratios over 70 days; the upper data are the same ratios for the first 50 days shown on an expanded ordinate scale. The straight lines at $1{\cdot}2657_8$ represent the mean of the ratios for the first 10 days.

If one wishes to look empirically into the possibility of a slow variation of the decay constant with time, a decay of the form $N = N_0 \exp\{-\lambda t(1+bt)\}$, where b is a constant, is convenient. This expression has been fitted to the data shown in figure 2 by a least squares method and an estimate made of the constant b. A value of $1{\cdot}9 \times 10^{-6}$ d^{-1} was obtained with a standard deviation of $1{\cdot}0 \times 10^{-6}$ d^{-1}.

It is difficult to see how the present method could be developed to cover a radically greater time. It might be possible to start with, say, 10 Ci of radon but even so, with the existing equipment, this would only extend the investigation by approximately 3 halflives. A larger crystal would increase the detection efficiency and if this were coupled with more efficient shielding from background radiation it would be possible to extend the measurements over a few additional halflives.

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References

Hellund E J 1953 *Phys. Rev.* **89** 919–22
Khalfin L A 1958 *Sov. Phys.-JETP.* **6** 1053–63
Marin P C 1956 *Br. J. appl. Phys.* **7** 188–90
Namiki M and Mugebayashi N 1953 *Prog. theor. Phys.* **10** 474–6
Osborne H 1966 *Phys. Rev.* **145** 1272–86
Tobailem J 1955 *J. Phys. Radium* **16** 48–53
Winter R G 1961 *Phys. Rev.* **123** 1503–7
—— 1962 *Phys. Rev.* **126** 1152–3