Comment on "Observation of the Deexcitation of the ^{229m}Th Nuclear Isomer"

In Ref. [1], the photon emission from recoil nuclei originating from the α decay of ^{233}U, implanted into MgF_2 plates, is studied in the ultraviolet spectral range. The result is interpreted as the first direct observation of the deexcitation of the lowest-lying isomeric state in ^{229}Th and an isomer half-life of 6(1) h is deduced. We want to point out here that radioluminescence resulting from the decay of other nuclei of the ^{233}U decay chain produces signals with a similar time dependence, but unrelated to the isomeric state in ^{229}Th.

With the same motivation of an optical observation of this unusual nuclear transition, we [2] and others [3] have performed related experiments. We observed a luminescence signal in the same spectral range as in [1], decaying within a few hours. The intensity depended on the material and thickness of the recoil absorber plate. From a fit of the time dependence we concluded that the luminescence is largely due to Cherenkov radiation resulting from the β decay of ^{209}Pb and ^{213}Bi, whereas the decay of ^{229m}Th was either radiationless [4] or happened within less than 30 s, during a time when the samples could not be observed. Those experiments were performed 3 yr after preparation of the ^{233}U source and decay products had been building up.

The experiments reported in Ref. [1] used freshly produced ^{233}U sources that were then observed over a period of about 140 days. We performed a calculation of the activities from an initially pure ^{233}U source that provides a ^{229}Th flux of $5 \times 10^4/\mathrm{s}$. The source is allowed to age (from 1 up to 150 days). Recoil nuclei from the α decays are collected in an absorber for 3.5 h. The time evolution of the α and β activities in the absorber after separation from the source is shown in Fig. 1. For simplicity, the same recoil transfer efficiency of 0.25 is assumed for all α daughters. For a source age above 10 days, the temporal dependence of the β activity largely follows the decay of ^{209}Pb (half-life 3.2 h). The α activity shows a more complicated time dependence, first decreasing because of the fast decay of ^{221}Fr (half-life 4.9 min), then building up over several days (half-lives of ^{225}Ra and ^{225}Ac) towards an equilibrium determined by the implanted ^{229}Th activity.

Since the sources used in Ref. [1] also contain 8 ppm of ^{232}U, the ingrowth of ^{232}U decay products is discussed as the reason for a time-dependent photon emission [1]. In a similar calculation for the ^{232}U decay chain, the temporal dependence of the activities after implantation is mainly determined by the β decay of ^{212}Pb (half-life 10.6 h). For the parameters of Ref. [1], the calculations yield a peak implanted β activity of $20/\mathrm{s}$ for a source age of 150 d.

These rates are smaller than the expected initial ^{229m}Th activity. But while the decay of the isomer may happen radiationless or on a shorter time scale, a single β decay may produce more than 10 Cherenkov photons within the wavelength range detected by the photomultiplier. It therefore appears likely that the processes discussed here have contributed to the signals observed in Ref. [1]. In addition, phosphoresence in the MgF_2 plates has been observed. Differential measurements with a Mylar foil that blocks the recoil ions are reported. The influence of this foil on the inducement of phosphorescence may be manifold and therefore not completely specified by a single scaling factor as it is done in Ref. [1]. While we do not exclude that the decay of the ^{229m}Th isomer has contributed to the photon emission observed in [1], we conclude that the sought-after signal would be heavily masked by background from other nuclear decays and radioluminescence induced in the MgF_2 plates. More experimental work will be required to elucidate the effect.

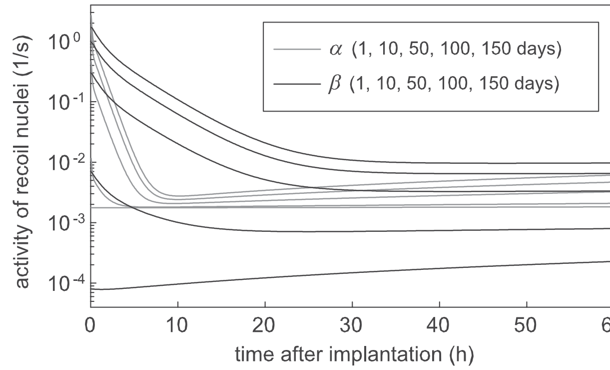

FIG. 1. Temporal dependence of the α (gray lines) and β (black lines) activities of recoil nuclei collected from a ^{233}U source of increasing age between 1 and 150 days (from bottom to top).

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