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APPLICATION OF DROPLET DETECTORS TO ALPHA RADIATION DETECTION

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Superheated Droplet Detectors (SDDs) are traditionally employed in the detection of neutrons. In this work the focus is on the detection of alpha particles using C_2ClF_5 as the target liquid. The alpha-droplet interaction is examined via computational studies, and a geometric model developed to describe the anticipated detector response. Experiments with alpha-emitting uranium- and samarium-doped SDDs at temperatures of 5-12 ºC confirm that the event rate is related to the size of the droplets, and are in model agreement for temperatures below 8 ºC; above this temperature, the acoustic sensitivity is reduced by signal attenuation as a result of the increasing bubble population, for which the addition of an attenuation coefficient restores the agreement with experiment. The results suggest a SDD-based alpha spectrometer using mono-sized droplets.

INTRODUCTION

A superheated droplet detector (SDD) consists of a distribution of micrometric superheated liquid droplets in a gel-like medium that undergo a transition to the gas phase upon energy absorption from radiation. The thermodynamic operation conditions can be tuned to render the SDD sensitive to only neutrons and alpha particles and insensitive to all minimum ionizing radiation backgrounds. Traditional SDD applications have been centred on neutron dosimetry and spectrometry through the registration of nuclear recoils following neutron interactions with the liquid atoms [1]. In this work the focus is reversed to the detection of alpha particles, specifically at very low emission rates. Possible applications of this methodology may include the measurement of alpha particle emissivity from modern ultra-low-activity integrated circuits aimed for high safety purposes.

A soft error (SE) is a nondestructive functional error induced in electronic devices by an energetic ion strike. Electronic devices in aircraft, space missions and nuclear power plants are selected for their reduced intrinsic SE response and resistance to radiation-induced SEs. One of the primary sources of terrestrial single soft errors is alpha (α)-particles emitted from radioactive impurities in materials [2]. Industry roadmaps call for instruments with detection limits of 1 α/kh-cm^2 in the energy range 1-10 MeV for measurement times of less than 168 h (1 week) with sample sizes <1500 cm^2 [3], which cannot be fulfilled by any of the current commercial proportional counters since their backgrounds (5 cts/h) are a factor 5 too high; ionization chambers are currently being explored.

In this work, we have examined a new approach to the measurement of α-emission rates, based on SSDs developed in the context of the SIMPLE dark matter search [4] which have demonstrated intrinsic neutron- and α-background to be smaller than $1x10^{-4}$ and 3×10^{-3} cts/h respectively, a factor 100 less than ionization chambers [5]. The study focused on their response to low energy α's during temperature ramping. Diluted sources have been employed as a preliminary study in a simple set-up relative to the inclusion of a solid sample in the gel matrix.

The response of SDDs to α irradiations has been previously studied in Refs. [6,7], mostly however using either a uranium composite (U_3O_8) or ^{241}Am distributed in the gel matrix; the response to ^{226}Ra at various temperatures was examined in Ref. [8]. The focus of this study was on the SDD response to the α-emitting elements uranium and samarium, with dominant energies of 4.72 and 4.77 MeV for ^{234}U, 4.15 and 4.20 MeV for ^{238}U and 2.25 MeV for ^{147}Sm.

MATERIALS AND METHOD

The general physics of SDD operation is based on the "thermal spike" model of Seitz [9]. Each superheated droplet can undergo a phase transition when two nucleation conditions are fulfilled: the incident particle energy deposition must be higher than a thermodynamic critical energy, and the deposition energy density must be greater than a critical linear energy transfer (LET_c).

The SDDs were prepared following standard SIMPLE protocols using C_2ClF_5 [10]. The hot gel was outgassed to remove all air trapped during the

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fabrication process. A quantity of radioactive liquid source was then injected into the hot gel and agitated quickly before being placed inside a hyperbaric chamber. For the uranium solution (Uranium Standard solution in HNO_3), the quantities were 0.37 Bq; for the samarium (Sm_2O_3 in 5% HNO_3), 0.37 Bq. Alpha spectroscopy identified ^{234}U and ^{238}U, with 4.77, 4.72, 4.2 and 4.15 MeV α's; for samarium, only ^{147}Sm with its 2.25 MeV α. The contribution from α-decays of other natural isotopes can be neglected due to low natural abundance (^{235}U) or significantly longer half-lives (^{148}Sm, ^{149}Sm) [11].

The droplet size distributions (DSD) from randomly-selected sites in the SSD volumes were measured using an Olympus Model Bx 60M optical microscope; the results in each slice were similar, following a Lorentzian distribution (expected value = 4 μm, Γ = 4 μm). For the α-response measurements each SDD was placed inside a temperature-controlled, circulating water bath, surrounded by a 20 cm thick radiation shielding of concrete blocks, paraffin and polyethylene. A 5 cm acoustic foam was installed inside the shielding to reduce the ambient noise (without acoustic foam, only events with amplitudes higher than 2 mV were detected; with acoustic foam, events with amplitudes lower than 0.4 mV could be detected). The bath temperature was monitored with an undoped SDD containing a temperature probe, which also provided background measurements.

The SDD signals were measured in atmospheric pressure at temperatures between 5–12 °C in 1 ºC steps (above 13 °C, γ-ray nucleation sensitivity begins). The time required for thermalization between adjacent steps was of order 1-2 h. Signal acquisition began after thermalization, for 20-60 min depending on the event rate. The acoustic instrumentation employed was the same as the SIMPLE experiment [4].

A visual examination of the experiments before and after each run noted many more bubbles than recorded by the microphone. Moreover, both the amplitudes and main frequency of the acoustic signal were observed to vary with temperature and the accumulated SDD exposure, most likely reflecting the smaller DSD of the measurements. Re-tuning of the fixed frequency filter to a 100-300 Hz window yielded a decrease in the acoustic noise level and generally higher event rates, but failed to record observed decreasing event rates above 8 ºC.

Previous studies [12,13] suggest an acoustic signal attenuation caused by the increasing bubble population. A separate set of experiments was performed to examine the event rate decrease with time, in which a detector with 3 Bq U was left to count for 6-24 h at fixed temperatures of 10 °C, 11 °C and 12 °C; after each temperature run, the SDD was recompressed. The exponentially-decreasing signal attenuation became evident after 10^3 bubbles.

SURFACE MODEL

In the case of the α-emitter doping, the U_3O_8 and Sm_2O_3 have an electrochemical affinity for both C_2ClF_5 [14]. In consequence, they should migrate to preferentially populate the larger droplet surfaces. The ions have moreover an affinity to the hydrophobic surface of the droplets, hence do not penetrate and in fact stabilize the emulsion by acting as a surfactant [6]. The results were analyzed on the basis of α Bragg curves computed for C_2ClF_5 at the experimental temperatures, as shown in Fig. 1, As seen, α's originating on a droplet surface would generally achieve LET > LET_c in C_2ClF_5 over distances of several tens of microns in the liquid, following several tens of micron penetrations with LET < LET_c. Droplets with diameters corresponding to LET ≤ LET_c ($p_<$) cannot contribute since the α transits the droplet without achieving LET_c; droplets with diameters much beyond the Bragg peak ($p_>$) continue to contribute despite the LET < LET_c since the bubble nucleation has already been initiated.

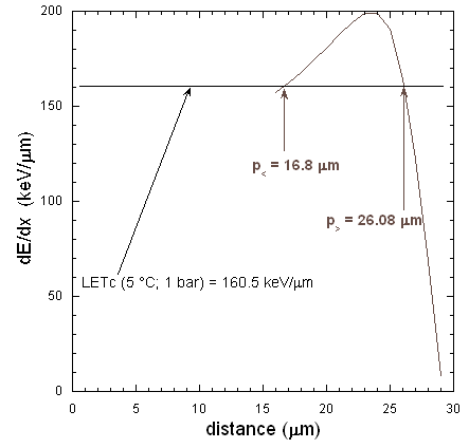

Figure 1. SRIM-computed 4.20 MeV α Bragg curve in C_2ClF_5 at 5 °C and ambient pressure. The intersection with the LET_c (161 keV/μm) gives $p_<$ (16.8 μm) and $p_>$ (26.08 μm), respectively.

From Fig. 2, for a fixed droplet size, the ^{234}U α emitters should begin triggering the droplets at 2 °C higher temperature than the ^{238}U α emitters, yielding an increase in the event rate with temperature ramping. The ^{147} Sm α should in contrast be able to trigger droplets of diameters > 1.5 μm, and a flat response in the counting rate while ramping the temperature from 5-12 °C at ambient pressure could be expected.

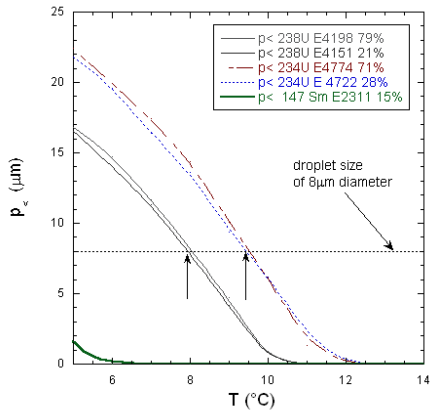

Figure 2. Minimum $p_<$ in C_2ClF_5 required for bubble creation by uranium and samarium with temperature. The ^{234}U α emitters start their contribution at 2 °C higher temperature than the ^{238}U α- emitters for the same droplet size.

For surface emission, the situation is described by the geometric intersection of two spheres with centers separated by a distance R, one of radius $p_<$ and the second with droplet radius R, as shown in Fig. 3.

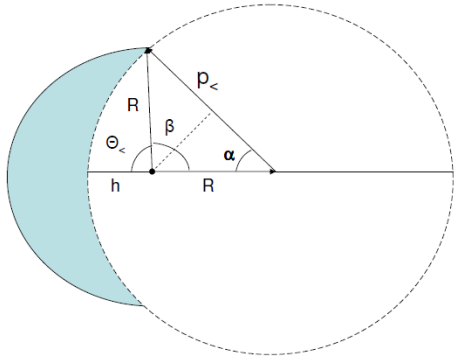

Figure 3. schematic view of the surface-emitted α - droplet interaction; the α's must enter the shaded region of the droplet to provoke a bubble nucleation.

A bubble nucleation efficiency is written as the ratio of the shaded-to-droplet volume:

$$\varepsilon_{nuc}(p_<;R) = 1-(\frac{p_<}{2R})^2 - 2(\frac{p_<}{2R})^3(1-\frac{p_<}{2R})^2(2+\frac{p_<}{2R}) \qquad (1)$$

The SDD counting rate at each temperature (corresponding to a specific value of $p_<$) is given by:

$$\tau_\alpha(T \leftrightarrow p_<) = \frac{1}{2}A_0\varepsilon_{att}\sum_{k_\alpha} f_{k_\alpha}\varepsilon(p_<)F(p_<) \qquad (2)$$

with A_0 the injected activity, ε_{at} is an attenuation efficiency as determined from the measurements, f_{ka} the number of alphas per unit activity inside the SED, and $\varepsilon(p_<)F(p_<)$:

$$\varepsilon(p_<)F(p_<) = \left(\frac{\int_{p_<\!/2}^{\infty}\varepsilon(p_<;R)\left(\frac{1}{1+(\frac{R-4}{\Gamma\!/2})^2}\right)dR}{\int_0^{\infty}\frac{1}{1+(\frac{R-4}{\Gamma\!/2})^2}dR}\right)\left(\frac{\int_{p_<\!/2}^{\infty}\frac{1}{1+(\frac{R-4}{\Gamma\!/2})^2}dR}{\int_0^{\infty}\frac{1}{1+(\frac{R-4}{\Gamma\!/2})^2}dR}\right) \qquad (3)$$

describes the "droplet number" efficiency, or the number of droplets that were involved in nucleation at each temperature.

RESULTS AND DISCUSSIONS

The results are shown in Fig. 4, in events per unit time and liquid mass. In the samarium case, there is no increase in the counting rate, only a flat response because only one α is involved. The theoretical expression is in good agreement with the experimental result, giving a rate of 1.35 evt/g/min vs. the measured 1.28 evt/g/min at 5 °C. For uranium-doping, the event rate in both cases increase with temperature, consistent with the reduction in nucleation threshold with increasing superheat.

Different event rates are however measured for the same nominal activity of different emitters. This is partly explained by the different α emission rates for the same activity of the parent isotope in the ^{238}U. In the case of uranium, the event rate begins increasing rapidly above 8 °C. In Fig. 4, one sees that the contribution from ^{234}U begins above this temperature if we assume a DSD centred on 8 µm in diameter. The measurements also show that, above 8 °C, the event rate increased by a factor 2, demonstrating that twice more α were involved in the measurement: the ^{234}U and ^{238}U were indeed in equilibrium. The disagreement between the measurements and model above 10 °C is unexplained. The model is simplistic, and requires further development. The Bragg curves, from which the $p_<$ are determined, are track-averages over calculated particle trajectories, and do not allow for statistical variations of the SSD response. It does not account for α-emission near the droplet surface, which although estimated to contribute at < 1.5% would alter the response via decreased $p_<$. Nevertheless, the model is seen to provide a basic description of the SED

response which reproduces to a large extent the experimental results.

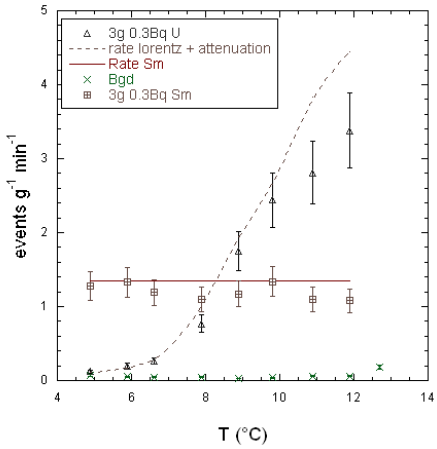

Figure 4. Comparison of model analysis with experimental uranium and samarium results at 0.3 Bq.

CONCLUSIONS

The effect of the droplet size on the α-response suggests the idea of an α-spectrometer construction which replaces the DSD with mono-size droplets. Larger mono-size droplets would be sensitive to higher α energy**,** but depending on $p_>$ also to other lower energy α's. The dependence of $p_<$ on temperature correlates the "kinks" in the SDD response function with the α energy; the two-fold increase in the signal at 8 ^{o}C corresponds to the emergence of the ^{234}U contribution. With mono-sized droplets, these features should become more evident. By using a droplet size of 2r = 15 μm, for example, only 4.15 MeV α particles would trigger the droplet at 5 °C; by increasing the droplet size to 20 μm at the same temperature, the detection would include both 4.72 MeV and 4.15 MeV α particles since the $p_>$ (5 °C) = 26 μm is the maximum penetration length of an α to trigger the droplet. By increasing the droplet size, the detection of higher α-energy (case for natural thorium) is increased. By choosing a droplet size and temperature ramping, different kinks arising from the emergence of other α emitters would be evidenced. In the case of C_2ClF_5, only α from a few to 5 MeV would be detectable. For higher α energy, a larger droplet would be necessary -- but smaller α energies would also trigger nucleation depending on the $p_>$.

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