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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 expected detector response. Experiments with alpha-emitting uranium- and samarium-doped SDDs at temperatures of 5–12°C confirm that the event rate is related to the size of the droplets, and are in model agreement for temperatures below 8°C; 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 the viability of 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 centered 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 (α) 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 SEs is alpha-particles emitted from radioactive impurities in materials[(2)]. Industry roadmaps call for instruments with detection limits of 1×10^{-3} alpha $h^{-1}\,cm^{-2}$ in the energy range 1–10 MeV for measurement times of <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 counts per hour) 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 alpha-emission rates, based on SDDs developed in the context of the SIMPLE dark matter search[(4)] which have demonstrated intrinsic neutron- and alpha-background to be smaller than 1×10^{-4} and $3 \times 10^{-3}\,h^{-1}$ respectively, a factor 100 less than ionization chambers[(5)]. The study focused on their response to low energy alpha'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 alpha 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 alpha-emitting elements uranium (to mimic natural radioactive impurities) and samarium (to evaluate the effect of low energy alpha), 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 SDDs were scaled-down 150 mL versions of the standard SIMPLE SDD, prepared following standard protocols[(4)] using 3–5 g of C_2ClF_5. The hot gel was outgassed to remove all air trapped during the 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 alpha's; for samarium, only ^{147}Sm with its 2.25 MeV alpha. The contribution from alpha-decays of other natural isotopes can

be neglected due to low natural abundance (^{235}U) or significantly longer half-lives (^{148}Sm, ^{149}Sm)[9].

The droplet size distributions (DSD) from randomly-selected sites in the SDD volumes were measured using an Olympus Model Bx 60 M optical microscope; the results in each slice were similar, following a Lorentzian distribution (expected value = 4 μm, width (Γ) = 4 μm). For the alpha-response measurements each SDD was placed inside a temperature-controlled (±0.1°C), 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 (IKA-Werke, Pt 100, ± 0.1°C), which also provided background measurements. Signal was recorded by a top-mounted MCE-200 Panasonic microphone with a 0.020–16 kHz (3 dB) range; the acoustic instrumentation employed the same as the SIMPLE experiment[4], with the fixed frequency filter retuned to a window of 100–300 Hz to account for the smaller DSD of the measurements.

Data was obtained at atmospheric pressure at temperatures between 5 and 12°C in 1°C steps (above 13°C, alpha-ray nucleation sensitivity begins). The time required for thermalization between adjacent steps was of order 1–2 h. Signal acquisition began after thermalization, lasting for 20–60 min depending on the event rate.

The recorded event rates were observed to decrease above 8°C. Previous studies[10] 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, 11 and 12°C; after each temperature run, the SDD was recompressed. The exponentially decreasing signal attenuation became evident after 10^3 bubbles, corresponding to a volume fraction of gas of ~0.4%[10].

SURFACE MODEL

The general physics of SDD operation is based on the 'thermal spike' model of Seitz[11]. 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).

In the case of the alpha-emitter doping, both U_3O_8 and Sm_2O_3 have an electrochemical affinity for C_2ClF_5[12]. 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 density-dependent alpha Bragg curves computed for C_2ClF_5 at the experimental temperatures, such as shown in Figure 1. The LET_c was obtained from $E_c/\Lambda r_c$, with $\Lambda(T) = 4.3(\rho_l/\rho_v)^{1/3}$ where ρ_l is the liquid density, using tabulated thermophysical properties of C_2ClF_5[4].

As seen, alpha'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. Droplets with diameters corresponding to LET ≤ LET_c ($p_<$) cannot contribute since the apha 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.

From Figure 2, for a fixed droplet size, the ^{234}U alpha emitters should begin triggering the droplets at 2°C higher temperature than the ^{238}U alpha emitters, yielding an increase in the event rate with temperature ramping. The ^{147}Sm alpha 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 to 12°C at ambient pressure could be expected.

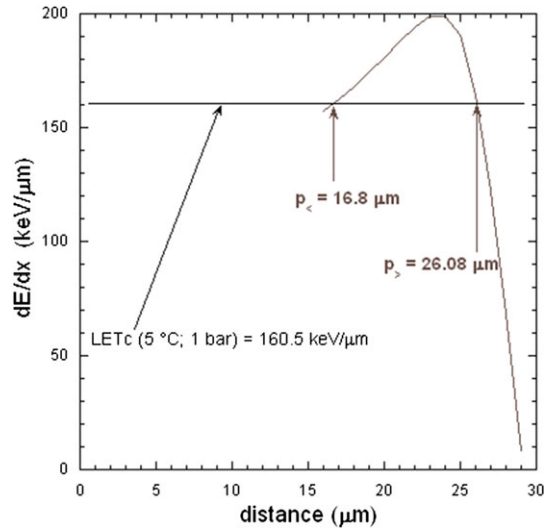

Figure 1. SRIM-computed 4.20 MeV alpha Bragg curve in C_2ClF_5 at 5°C and ambient pressure. The intersection with the LET_c (161 keV/μm) gives $p_<$ (16.8 μm) and $p_>$ (26.08 μm), respectively.

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 Figure 3.

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

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

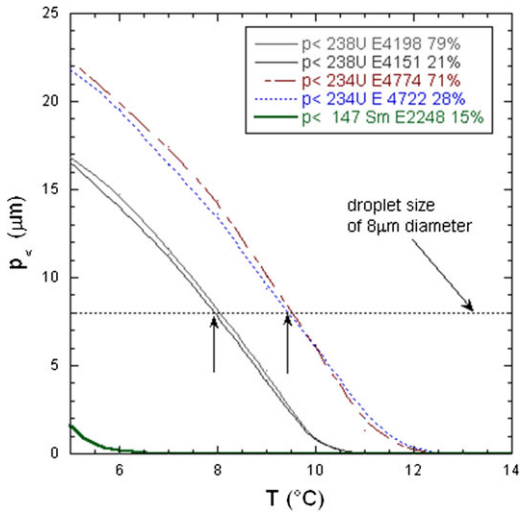

Figure 2. Variation of $p <$ with temperature in C_2ClF_5 required for bubble creation by isotopes of U and Sm with alpha's of energy E and % isotopic abundance.

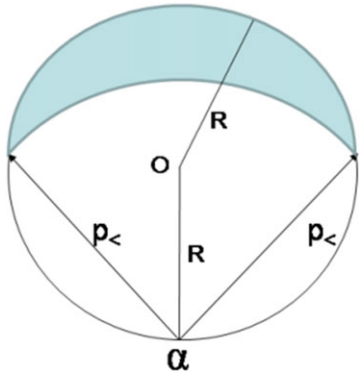

Figure 3. Schematic view of the surface-emitted alpha-droplet interaction of center O; the alpha's must enter the shaded region of the droplet of radius R to provoke a bubble nucleation.

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

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

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

$$\varepsilon(p_<)F(p_<) = \left(\frac{\int_{p_</2}^{\infty}\varepsilon(p_<;R)\left[\dfrac{1}{1+\left(\dfrac{R-4}{\Gamma/2}\right)^2}\right]\mathrm{d}R}{\int_0^{\infty}\dfrac{1}{1+\left(\dfrac{R-4}{\Gamma/2}\right)^2}dR}\right)\left(\frac{\int_{p_</2}^{\infty}\dfrac{1}{1+\left(\dfrac{R-4}{\Gamma/2}\right)^2}dR}{\int_0^{\infty}\dfrac{1}{1+\left(\dfrac{R-4}{\Gamma/2}\right)^2}dR}\right) \quad (3)$$

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

Oxides outside the droplet surfaces may also contribute. A similar estimate, using alpha Bragg curves in the SDD gel, gives ~1.5% contribution and negligible theoretical counting rate compared to experimental results.

RESULTS AND DISCUSSIONS

The results are shown in Figure 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 alpha is involved. The theoretical expression is in good agreement with the experimental result, giving a rate of $1.35\,g^{-1}\,min^{-1}$ vs. the measured $1.28 \pm 0.20\,g^{-1}min^{-1}$ 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 alpha 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 Figure 4, one sees

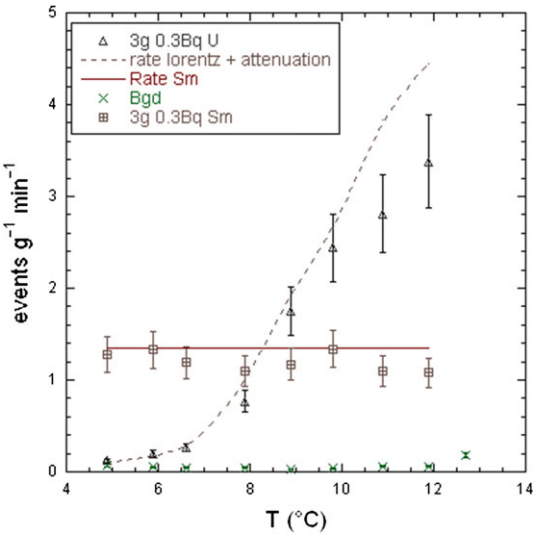

Figure 4. Comparison of model analysis with experimental uranium and samarium results at 0.3 Bq.

that the contribution from ^{234}U begins above this temperature if we assume a DSD centered 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 alpha particles 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 SDD response. It does not account for alpha-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 SDD response which reproduces to a large extent the experimental results.

CONCLUSIONS

The effect of the droplet size on the alpha-response suggests the idea of an alpha-spectrometer construction which replaces the DSD with mono-size droplets. The droplet size is directly linked to $p_<$ which is temperature dependent. By fixing the droplet size, $p_<$ is revealed by varying the temperature: a sudden increase in counting rate is seen, giving the energy of the alpha emitters ($E_\alpha \leftrightarrow p_<$) and yielding the natural radioactive abundance of the contaminant for SE assessment. The dependence of $p_<$ on temperature correlates the 'kinks' in the SDD response function with the alpha energy; the 2-fold increase in the signal at 8°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 alpha 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 and 4.15 MeV alpha particles since the $p_>$ (5°C) = 26 μm is the maximum penetration length of an alpha to trigger the droplet. By increasing the droplet size, the detection of higher alpha-energy (case for natural thorium) is increased. By choosing a droplet size and temperature ramping, different kinks arising from the emergence of other alpha emitters would be evidenced. In the case of C_2ClF_5, only alpha from a few to 5 MeV would be detectable. For higher alpha energy, a larger droplet would be necessary—but smaller alpha energies would also trigger nucleations depending on the $p_>$.

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