Radiation hardness testing of an organic liquid scintillation detector for use in high dose rate accident response scenarios

Ashley R. Jones, Malcolm J. Joyce, *Member IEEE*, Arron Griffiths, Barry Lennox, *Senior Member IEEE*, Dale Potts, Simon A. Watson, *Member IEEE*, Jun-ichi Katakura, Keisuke Okumura, Kangsoo Kim, Michio Katoh, Kazuya Nishimura and Ken-ichi Sawada

Abstract – **Organic liquid scintillation detectors offer the advantage relative to many alternatives that they are sensitive to both fast neutrons and γ rays, whilst radiation type can be discerned on the basis of pulse-shape discrimination. Mixed radiation fields of this type can arise in the context of reactor accidents via, for example, ^{137}Cs (γ) and ^{244}Cm (neutrons). However, performance degradation of such scintillators, such as EJ-301, is a significant possibility that might limit the use of this technology in accident response applications. The premise behind the high dose rate testing of such a liquid scintillator described in this paper is for fuel debris characterisation at Fukushima Daiichi, which has expected dose rates of up to 1000 Gy/hr in close proximity to fuel debris. The tests carried out for this investigation involved using the ^{60}Co gamma irradiation facility at the Dalton Cumbria Facility, Cumbria, United Kingdom to expose the detector to a similar dose rate to that which is estimated within the primary containment vessel for survivability tests. Radiation hardness tests have rarely been reported for such devices and it is expected that the performance will be dependent on the survival of the window of the photomultiplier tube rather than the liquid scintillant itself. A major advantage of the use of this detector is its physical size, due to the limitations on access into Fukushima reactors physical space is a premium. The research described in this paper presents the results of the dose rate exposure of the detector before signal was lost with the total dose observed providing information on any degradation affecting the performance of the device post-irradiation.**

I. INTRODUCTION

The research carried out within this paper is the radiation hardness testing of an organic liquid scintillator detector for use in high dose rate scenarios. The scenario specifically is for fuel debris characterisation of the reactors at Fukushima Daiichi Nuclear Power Station. The design goal for this device for the dose rates within the primary containment vessel (PCV) are to withstand as high as 1000 Gy/hr in close proximity to the fuel debris, with a more general rate of 10 Gy/hr elsewhere. Detectors are currently being investigated for use within a submersible ROV to be utilised within the reactors at Fukushima to detect the location and state of any fuel debris. Investigative probes for inside the PCV are been designed to withstand a dose rate of 100 Gy/hr for a maximum of 10 hours giving a total absorbed dose of 1 kGy as a guideline to the average dose that may be experience.

The ROV to be utilized is a modified version of the Aqua Vehicle Explorer for In-situ Sensing (AVEXIS) MiniROV [1, 2] developed by The University of Manchester. The AVEXIS MiniROV, Figure 1, measures 145 mm in diameter and 250 mm in length, and is designed for the inspection of legacy storage ponds. The modified version of this design developed specifically to inspect the PCVs at Fukushima Daiichi is the AVEXIS MicroROV, Figure 1, which measures 110 mm in diameter and 450 mm in length and has 5 degrees of freedom (DOF). This ROV has been designed specifically to carry a payload of detectors for the fuel debris identification and classification within the reactors at Fukushima Daiichi deployable through 115 & 150 mm access ports.
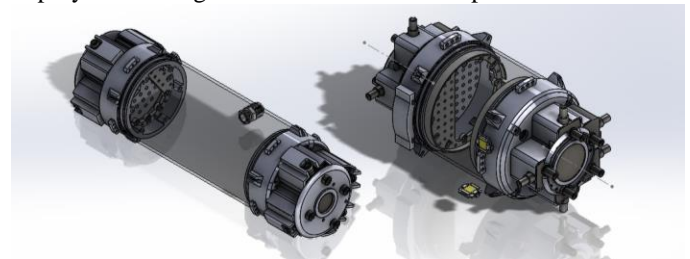

Figure 1. AVEXIS MicroROV (left) and AVEXIS MiniROV (right).

Due to the limitations on space within the ROV it would be desirable to either use a singular detector with mixed field properties or to utilize multiple detectors of very small dimensions. Another desirable property for the detector is

M. J. Joyce and A. Jones are with the Engineering Department, Lancaster University, UK (telephone: +44 1524 593812, e-mail: m.joyce@lancaster.ac.uk).

A. Griffiths, D. Potts, S. Watson and B. Lennox are with University of Manchester, UK

J. Katakura is with Nagoaka University of Technology, Japan

K. Okumura is with the Japan Atomic Energy Agency, Japan.

K. Kim, M. Katoh, K. Nishimura and K. Sawada are with the National Maritime Research Institute, Japan.

short pulse times to alleviate any saturation of the detector when exposed in the currently unknown but presumed high flux environment within the PCV area. The use of any organic plastic scintillators has been ruled out due to the discolouration that occurs under very low dose rate. This discolouration limits the radiation transport until it is totally redundant giving it very limited use for an application as described. Alternatively many inorganic scintillators lack the ability to exhibit mixed field radiation sensitivity meaning multiple detectors would be required using up premium space inside the ROV.

This has led to the investigation of radiation damage upon liquid organic scintillators. It was hypothesised that the liquid scintillant cocktail itself would be unaffected and for the damage to occur at least in the first instance to the borosilicate glass window of the PMT. To carry out the investigation the ^{60}Co gamma irradiator facility at the Dalton Cumbria Facility, Cumbria, United Kingdom, was utilized to set up multiple experiments of varying dose rates for robustly testing the detector.

II. EQUIPMENT

The detector used for this investigation consisted of a cell containing the EJ-301 liquid scintillant [3], coupled to an R-5611 PMT by Hamamatsu Photonics K.K. [4], the device in total measures 35 mm in diameter by 80 mm in height shown in Figure 2. This ability for mixed field radiation information and the small size makes it very desirable for use within the ROV system.
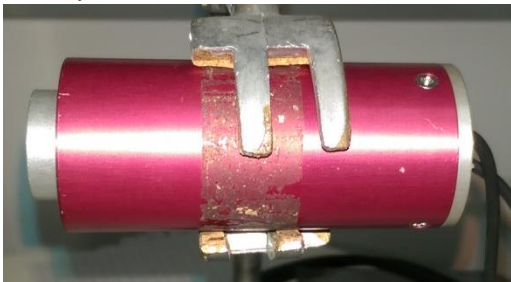

Figure 2. EJ-301 liquid scintillator detector, model type 19A15/0.75-E1-LS-Z-N supplied by Scionix, Netherlands [5]

The signal is fed back 4-channel mixed-field PSD analyzer (MFA) Hybrid Instruments Ltd., UK [6] that makes use of the Pulse Gradient Analysis (PGA) technique [7] for separating out gamma and neutron signals. PGA allows the discrimination of these radiation transients in real time making it the optimum technique to be coupled with the use of the submersible ROV. The information being produced by the MFA can be fed through to a computer that can give real-time visual feedback through the population of a scatter plot of peak amplitude vs discrimination amplitude, additionally the information of the pulses can be saved to a log file for post processing applications. The radiation hardness tests using the irradiator described below will only produce gamma rays, producing a singular plume of pulses on the PGA scatter plot.

The irradiator facility houses a self-shielded model 812 ^{60}Co gamma irradiator that was developed and installed by Foss Therapy Services, Inc. [8] shown in

Figure 3. The irradiator is capable of supplying absorbed dose rates from 240 Gy/hr up to 27,000 Gy/hr. These dose rates can be attenuated and shielded to match desired experimental setups. The irradiator consists of three source rods of ^{60}Co that can be used simultaneously or individually depending on the desired dose. The irradiator chamber has two service ports of 19mm diameter to accommodate cables for power supplies and signals.

The dose rate within the irradiator was measured using an ion chamber dose sensor model 10x6-0.18 supplied by Radcal [9]. The ion chamber has a detection limit between 500 nGy/s to 6.31 Gy/s. This dose rate data is fed through a Radcal Accu-Dose+ digitizer module model ADDM+ [10] to a desktop computer. The ion chamber was placed at the position in which the sensitive area of the detector would be positioned and the source exposed for a minute to gather a dose rate reading.

Figure 3. ^{60}Co gamma irradiator, Foss Therapy Service, Inc. [8]

III. RADIATION TESTING

The testing was to try and characterise two different variables on the detector. The first of these characteristics being the hardness of the detector, this test will require exposing the detector to a high dose rate for an envisaged period of time for the ROV to be investigating within the reactor. This was decided to be a test of any damage on the detector after one hour of exposure at the highest estimated dose rate of 1000 Gy/hr. The second characteristic is the saturation point of the detector, this will be decisive on how effective the detector will be for this application.

The initial setup at DCF was to expose the liquid scintillator to roughly a 1000 Gy/hr dose rate and observe the pulses

produced as well as the throughput of these. This dose rate was selected to simulate rough estimate of the highest possible dose rate within the PCV's at the reactors. The actual measured dose rate was 942 Gy/hr ±2%, which was provided by using 2 of the source rods. It was expected to be able to observe degradation in either the pulse shape or throughput when radiation damage was starting to take place, at which point the dose would be terminated. The liquid scintillator was placed inside the irradiator as shown in

Figure 4, with the power supply control box shielded further still behind a lead block as shown. The high voltage was supplied via a coax cable through the side of the irradiator chamber similarly the signal was being fed back the same way.

The polystyrene in-front of the detector was to prevent the detector from moving whilst being investigated, polystyrene was used due to its lack of interaction. Once the setup was in place the irradiator was closed and the ^{60}Co rods were raised into place for exposure. It was observed that the instant the sources were raised the detector went dead, producing no pulses or signal. Upon the detector going dead, the sources were retracted and the detector became instantly operational again. This instant recovery of the detector suggests that the problem the detector exhibited was not due to damage, as this would have been expected to exhibit a gradual recovery if any at all. With this knowledge it was concluded that the detector must have become saturated due to the dose levels being used. To try and determine if there would be any radiation damage of the detector at this level of dose rate a long exposure was run, 50 minutes, giving a total dose of approximately 8 kGy, after this dose the detector was observed to still be operating as normal the instant after the sources were removed.
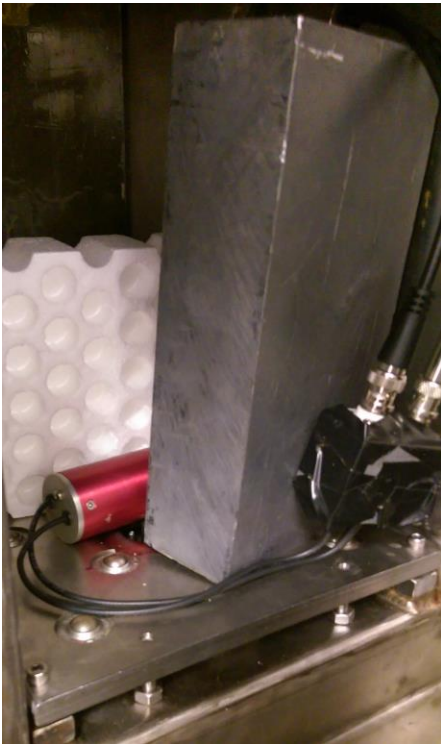

Figure 4. EJ-301 organic liquid scintillator inside the ^{60}Co irradiator.

To try and corroborate this idea the dose rate was progressively reduced to try and find the point at which it saturated. Initially the dose rate was halved to 420 Gy/hr ±2% by only using one of the source rods, this exhibited the same result as previously with the detector instantaneously producing no pulses or signal output and hitting its saturation point. To find a dose rate that the detector could withstand more attenuation was added inside the irradiator. The final setup in which the detector was operational is shown below in Figure 5.

The dose rate was reduced to the minimum obtainable within the irradiator facility by adding the maximum number of attenuation plates with additional lead shielding within the chamber. This was measured using the ion chamber to be 9.9 Gy/hr ±2%. at this dose rate the detector remained operational for the first few seconds of exposure, with intermittent count readings through exposure. It is presumed that this dose rate is just above the upper saturation limit of the detector. Looking at the DAQ readout from the first few seconds of irradiation after the experiment had been completed gave a scatter plot as shown in Figure 6. This shows a lot more noise than would normally be expected, this can be associated to both radiation effects imposed upon the cables that are feed through the service ports and the detector being on the verge of saturation.

Figure 5. EJ-301 organic liquid scintillator inside the ^{60}Co irradiator.

IV. RESULTS

During the low level irradiation tests at 9.9 Gy/hr the information being seen by the detector was logged for the

purpose of post processing. The post processing allows a picture of the pulses to be built up into a scatter plot of two different integrals, one of these being the peak of the pulses that are being seen by the detector, the second being a set user defined time after this peak amplitude. The scatter plot can be seen in Figure 6.
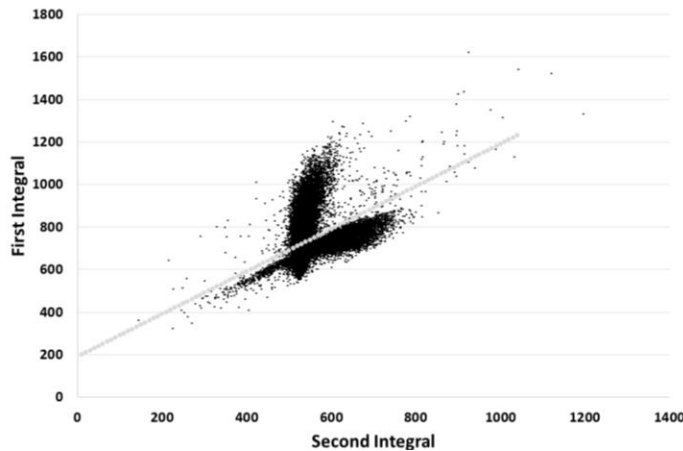

Figure 6. Scatter plot showing gamma radiation and noise threshold line (grey) for the 30 minute exposure run, (arbitrary units).

With the radiation field associated with this experiment it would be expected to view one clear plume of data, as can be seen above two plumes are visible. This second plume has been isolated by the addition of a threshold y=x+c line. The second plume that has manifested is associated with saturated or clipped pulses and noise. When a pulse is processed if the pulse is of greater amplitude than is observable through the system it will produce a flat top at the maximum observable amplitude. The effect this flat top has on the processing is that the peak amplitude sample and the discrimination amplitude sample are either identical if the pulse is very large, or very similar depending on the time this flat top is maintained for, producing this second plume around the y=x line as shown above.

Each of the three tests showed a prominent plume of gamma counts as well as noise. This is suggestive that the detector is on the verge of saturation at 9.9 Gy/hr for these first few seconds, before becoming fully saturated.

V. CONCLUSIONS

The advantages of the liquid scintillator over other detectors is the ability to gather both gamma and neutron information from just one detector, however with the intrinsic safety concerns of using the EJ-301 cocktail in such an environment coupled the more recently developed low-hazard version EJ-309 has been selected for use.

With the detector being unable to operate in dose rates of such high intensity its usefulness as a solo detector in the ROV is diminished. This characteristic means that the detector would survive passing through areas of high dose rates without noticeable radiation damage, but would not be operational until the dose rates drops to a level below 10 Gy/hr. As the liquid organic scintillator has the ability to survive a high total dose before showing signs of damage this will allow it to be incorporated still as a back-up detector for

the lower dose rate areas and upon saturation it will indicate that the dose rate where the ROV and detector is located has increased above this 10 Gy/hr limit. With the limited information that is currently available about the environment this will allow information to be gathered on localised hotspots within the PCV at a minimum as well as indicating when to start analyzing the other results being produced by its accompanying detectors within the ROV. Although space is at a premium the limited size of the organic liquid scintillator does allow for multiple detectors to be used alongside under the criteria that they also are small in size.

The localised hotspot map will be a significant step forward in terms of information currently held with regard to the dose rates within the primary containment vessel and more importantly in the lower submerged parts of the reactor.

VI.   REFERENCES

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