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Investigating the lifetime of bromine-quenched G.M. Counters with temperature

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ABSTRACT

The amount of halogen quench gas as a percentage of the total fill gas contained within a gas-filled Geiger–Müller detector is directly linked to its operational characteristics. Preserving this halogen gas will help maintain the operating lifetime of the detectors. Such halogen gases are highly corrosive and the small quantities within a gas-filled detector can deplete rapidly via interactions with surrounding materials. The rate of interactions is thought to be proportional to not only temperature, but also to the current initiated by ionisation events associated with the formation of each signal pulse. As all pulses are of similar magnitudes, GM detector operational lifetimes are quantified in accumulated counts rather than a given operating time. We have studied three different types of corrosion-resistant mechanisms to protect the bromine halogen gas from any interactions with 446 stainless steel detector components of ZP1200 Geiger–Müller tubes at temperatures up to 125 °C. Three types of surface treatments for these detectors used were labelled as "raw" using only an oxygen-plasma-bombardment process, "passivated" using a combination of nitric acid passivation followed by an oxygen plasma-bombardment process, and thirdly plating with a few micron thickness of chromium followed by an oxygen plasma-bombardment process. 32 detector samples were irradiated at room temperature with a Caesium-137 source at dose rates of approximately 1.3 mSv/hr up to 5.7×10^{10} accumulated counts; another 32 detector samples were aged to 3.3×10^{10} counts at 125 °C. At room temperature, the chromium-plated samples exhibited an initial rise in their starting voltage readings. All other detector performance characteristics, for all detector types, did not change significantly during the ageing process, and the surface morphology of the detector cathodes was unaffected. At 125 °C, the chromium-based plating produced the most stable long-term response. These chromium-plated samples showed no evidence of bromine deposits on the cathode samples investigated via Energy-Dispersive X-ray Spectroscopy. The raw and passivated samples, however, did show traces of bromine independent of the age of the detector. Preliminary investigations we have carried out at 175 °C highlight the importance of the chromium plating layer in preserving long-term detector performances at elevated temperatures.

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1. Introduction

The gas-filled Geiger–Müller (GM) detector has long been used for radiation monitoring applications. One industry of particular interest in the development of GM detectors is the oil and gas industry. Carrying out gamma ray logging in deep-well drilling operations can subject monitoring devices to extreme conditions. The majority of oil wells have temperatures below 125 °C, as mentioned in [1]. It is for this purpose that the detectors must operate at temperatures up to 125 °C. While a GM detector with an infinite operational lifetime would be ideal, due to their design, such timespans cannot be achieved without compromise. One main reason for the limitations in a GM detector's functional lifetime is due to the necessary implementation of an effective quenching mechanism; this helps to re-establish the electric field necessary for further discharges and counts to occur.

After the fill gas of a detector is ionised by incident radiation, any positive ions formed will drift towards the negative cathode wall. Upon reaching the cathode, the ions will then interact with the cathode wall and return to their neutral state by acquiring an electron. Such a process often leaves the atom in an excited state. In this state, the atom can emit a photon having sufficient energy to cause further gas ionisations; this will result in constant spurious counts being detected. The quenching mechanism, however, interrupts the emission of any secondary photons to reduce the likelihood of spurious

counting occurring to a negligible level. This concept is explored further in [2].

The introduction of an additional gas component (passive quenching) or the use of an external circuit (active quenching) can be implemented to produce the required effect. For more information on the operating mechanism governing each method, along with their benefits and disadvantages, refer to [2]. Due to design practicality and efficiency in higher radiation environments, a halogen gas-based quencher is more commonly used in the manufacture of GM radiation detectors.

In gas-filled GM detectors, the depletion of the quenching gas governs the lifetime of the counter. Organic gases have a limited long-term quenching ability due to each molecule dissociating irreversibly after quenching. Halogen molecules have the ability to recombine after each quench event; for this reason, they are favoured over their organic counterparts. The drawback in using halogen quench gases is that, because of their very high activity when dissociated, they might deplete through interactions with surrounding materials or contamination, as mentioned in [2]. The selection of detector materials thus becomes essential in determining the most halogen-resistant construction. The compositions of GM detector components exposed to the quench gas are carefully selected to contain high quantities of chromium [approximately 28% by weight]; the trade name of such steels is Type 446. Chromium is a common alloying element in steels due to the corresponding mixture producing a highly corrosion-resistant oxide layer, as discussed in [3]. The loss of quench gas is thought to be a function of the number of energetic species produced per detection event, which relates to the measured pulse height of each count, as mentioned in [4]. It is for this reason that detector lifetimes are expressed in terms of accumulated counts.

Another factor to consider in GM design is the surface conditioning of detector components exposed to the halogen. The main detector performance characteristics to consider, discussed in more detail in [5], are the detector starting voltage (Vs), the length of the Geiger plateau, the plateau gradient, the detector's sensitivity, and the background count rate. Background count rates are measured in the absence of a source and high values can signify the presence of undesirable spurious counts.

Liebson and Friedman, [6], have shown that a GM detector's Vs will drop with less quench gas and can be used to estimate the extent of losses in the detector quench gas. It has also been shown in [4] that Energy-Dispersive X-ray (EDX) analysis is suitable for investigating any halogen quench gas attack on the internal Geiger components with increasing age at temperatures up to 200 °C. In [4], it has also been suggested that quench gas interactions can occur with other components, such as the anode pins or the glass seal of the detectors in addition to the large cathode surface of the detector. However, this paper will focus on three cathode surface protection processes, which are compared in terms of their effectiveness in maintaining the operational Geiger–Müller parameters by reducing the depletion of halogen quench gases at different temperatures.

2. Materials and methods

To carry out these investigations, Centronic Ltd.'s ZP1200 Geiger–Müller counter was selected; the halogen quench gas used is high-purity bromine. The minimum protection process considered, referred to as an oxygen bombardment, was the forced formation of a chromium oxide layer by exposing the internal surfaces of the 446 stainless steel to pure oxygen plasma. Detectors produced using this technique are labelled as 'raw samples'.

The second protective technique considered is a nitric passivation process. Such a procedure is commonly used to cleanse stainless steel surfaces of impurities and eliminate any potential corrosion sites, as shown in [7]. This is possible due to the highly oxidising nature of

nitric acid. After the nitric passivation, the oxygen bombardment technique was carried out. By using both techniques, it is expected that the internal surfaces will be free from any traces of contamination and also have the protective chromium oxide layer. This dual-protective process is used to produce detectors labelled as 'passivated detectors'.

The third protective process investigated was chromium plating the internal surface of the detectors. Chromium plating is a common industrial process used to produce corrosion-resistant materials, as shown in [8]. The deposition of a pure chromium layer that is several micrometres thick along the inner detector surfaces will yield a purer chromium oxide. In addition to the deposition of chromium on the internal cathode surfaces, these detector samples also undergo the oxygen bombardment process to form a chromium oxide layer before filling. Such detectors are labelled as 'plated detectors'. All samples for all detector types have been filled with identical gas mixtures and to the same fill pressure.

Fifty-four detectors were processed for each of the three protective processes. Two separate ageing jigs with 32 positions were manufactured, one for each temperature, using the recommended circuit for this type of GM detector described in [5] and illustrated in Fig. 2.1. Each jig was loaded with 10 plated detectors, 11 passivated detectors and 11 raw detectors. The room temperature jig was positioned 60 cm from a 6 GBq Caesium-137 gamma source. The high temperature jig was placed within an environmental chamber set to a temperature of 125 °C also situated 60 cm from the same Caesium-137 gamma source.

The majority of the test equipment has been developed in-house at Centronic Ltd. A variable power supply produced a variable voltage of $(0–800 \pm 2)$ volts across each jig. A multi-channel PicoLog 1216 data logger was then connected to the setup to monitor the output voltage pulse for each tube. The output was also connected to a scaler-timer to measure the count rate of each detector; this equipment was designed to count all input pulses above a set threshold of 100 mV. A switch-box was manufactured to isolate the signal from each GM counter.

The recommended 500 V ZP1200 operating voltage was applied during the ageing process. The initial average count rate for the detectors at room temperature was found to be (2572 ± 50) counts per second (cps) with a standard deviation of 110 counts and (1954 ± 45) cps with a standard deviation of 144 counts for the high temperature jig. Since the radiation is emitted uniformly in all directions, this difference is due to the environmental chamber attenuating some of the incident radiation.

All detector samples were exposed to the source for 24 h a day. The Vs value, which is defined as the lowest applied voltage that produces a 1 V pulse across the anode resistor, for each detector was measured using the PicoLog every few weeks to capture any potential changes.

Count rate measurements for 1 s at fixed dose rate as a function of applied voltage, are typically acquired between 0 and 750 V resulting in the characteristic Geiger Plateau plots. For this purpose, the detector samples were removed from the set up and placed at a distance of 1 cm from a significantly weaker 0.6 GBq Caesium-137 gamma source at room temperature; this produced a count rate of approximately 450 cps.

At different ageing intervals, an internal cathode surface of each detector type was analysed. The samples were carefully cut open and studied under a Hitachi S3200N Scanning Electron Microscope (SEM) with the accelerating voltage set to 20 kV. Several regions of each cathode sample were studied. An energy dispersive X-ray (EDX) attachment was then used to obtain the elemental constituents at each region of interest. The aim of this EDX analysis was to determine whether the halogen quench gas had reacted with the cathode. For a set of regions of interest per sample, the elemental compositions were recorded and averaged.