Study of tritium (^{3}H) speciation in a reactor bioshield concrete

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1. Introduction

A significant number of first and second generation nuclear research sites and nuclear power plants in the UK are being decommissioned and many others are due to begin decommissioning within the next decade. The nature of decommissioning works will vary from site to site but, in most cases, will involve either the extensive clean out, refurbishment or demolition of buildings and other facilities, and remediation of the land. With this process, large volumes of waste materials will be generated. Some of these materials will be contaminated with radioactivity therefore they must be treated as radioactive waste and carefully disposed of in accordance of the requirements of national radioactivity regulations. Concrete, cements and rubble consist of 22% of total weight of low level waste (LLW) and 12% of total weight of intermediate level waste (ILW) in the UK [1]. The radioactivity in the reactor bioshield concrete is derived from the integrated effects of neutron irradiation of many nuclides and includes ^{3}H, ^{14}C, ^{63}Ni, ^{60}Co, ^{36}Cl, ^{55}Fe, ^{41}Ca, ^{152}Eu, ^{90}Sr, ^{133}Ba, ^{137}Cs and some transuranics [2]. However, tritium is considered to be the critical radionuclide [3-5]. Detecting tritium is potentially difficult in many materials because of its weak beta energy and absence of gamma rays and is consequently rather expensive. There are several reported methods in use for tritium determination, which are partly dependent on sample type and activity but combustion is particularly successful as it can physically extract and separate ^{3}H and ^{14}C from other radionuclides. In this study, the tritium distribution across a concrete bioshield core was analysed and compared with tritium from structural concrete.

2. Methodology

A concrete core, traversing the bioshield, was taken from of a UK reactor being decommissioned. Sub-samples of the core were crushed and measured. Tritium thermal evolution profiles for a sequential series of these sub-samples were determined along with the total tritium concentration profile. Tritium extraction was achieved using a Raddec Pyrolyser Trio™ System (Figure 1) which provides simultaneous extraction/oxidation of six samples at a time. The methods used were developed and validated by GAU-Radioanalytical at the University of Southampton (www.gau.org.uk) a group specialising in radionuclide R&D and analysis. All final tritium measurements were performed using a 1220 liquid scintillation counter (Wallac Quantulus™). Samples were run in a manner where sub-samples of the tritium vapour were taken at selected times so the evolution profile with temperature and time could be evaluated.

3. Results and discussion

There were two distinct styles of tritium loss from the concrete sub-sample as they were progressively heated. In concrete sampled further away from the reactor (outer bioshield) tritium was lost more rapidly and at lower temperature. The evolution profile was similar to that seen for tritium loss from structural concrete (non-bioshield concrete). However, for the concrete sub-samples closer to the reactor (inner bioshield concrete) the evolution of tritium was quite different (Figure 2). For these samples the following were noted.

- Loss of tritium in sub-samples occurred episodically and over a broad range of temperatures.
- The overall ^{3}H concentration in bioshield concrete near the reactor decreases exponentially with distance from the reactor.

These different rates of loss were interpreted as being caused by the speciation of tritium in the concrete caused by the origin of the tritium.

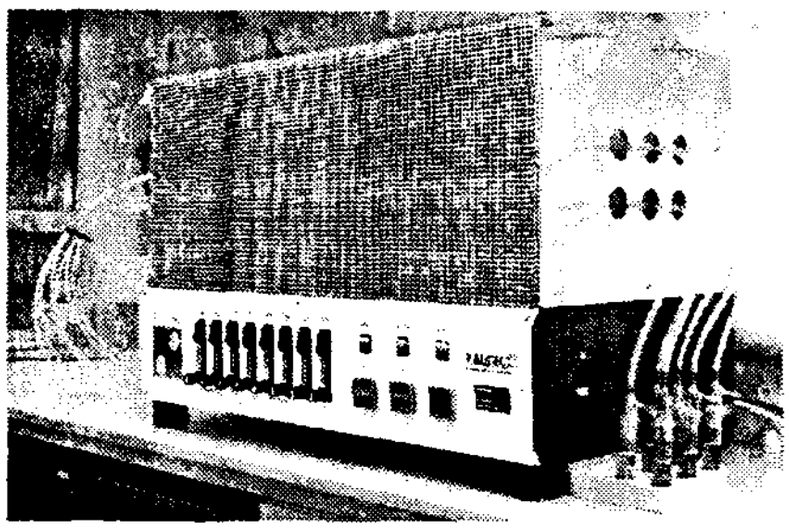

Figure 1: Raddec Pyrolyser™ System

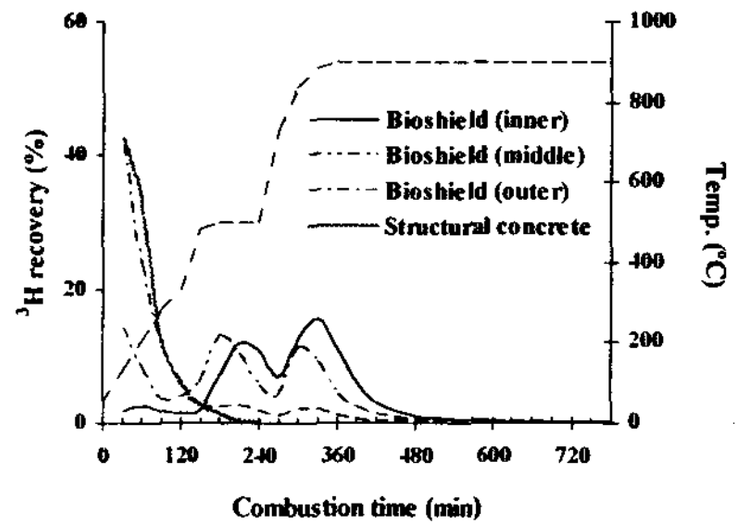

Figure 2: Evolution profile of structural and bioshield concrete

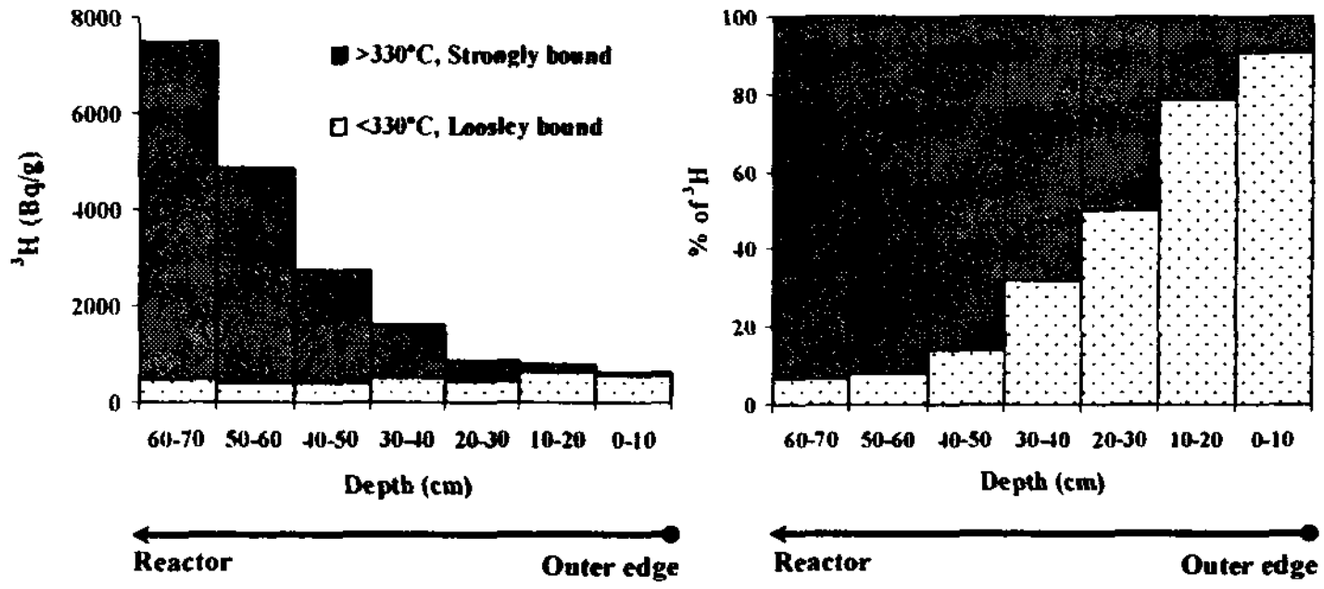

Figure 3: Tritium form in bioshield concrete

4. Conclusions

Bioshield concrete can contain tritium in several forms with possible production routes being by one or more processes. These are neutron capture by deuterium, an (n,α) reaction on lithium-6 and ternary fission (fission yield <0.01%). Systematic analysis of a sequence of sub-samples taken from the examined core showed different tritium-loss profiles that depended on their position in the core. Tritium lost at low temperatures (100℃) was as HTO and associated with water loss from hydrated mineral components. Tritium lost at higher temperatures (350℃) was held more strongly in mineral structures and required a temperature of >860℃ was required to lead to total liberation. The more-strongly retained tritium originates from tritium generated by neutron capture of trace lithium (lithium-6) distributed throughout minerals in the concrete. The higher concentrations seen in concrete sub-samples near the reactor are due to the greater neutron flux seen by those samples. On the other hand tritium from the outer edge samples of the bioshield concrete (and non-bioshield structural concrete) are related to the absorption of tritiated water (HTO) from the reactor hall atmosphere. The results of this study have important implications for the disposal options for bioshield concrete.

5. References

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