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# Tritium Speciation in Nuclear Reactor Bioshield Concrete and its Impact on Accurate Analysis

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Tritium ( $^3\text{H}$ ) is produced in nuclear reactors via several neutron-induced reactions [ $^2\text{H}(\text{n},\gamma)^3\text{H}$ ,  $^6\text{Li}(\text{n},\alpha)^3\text{H}$ ,  $^{10}\text{B}(\text{n}, 2\alpha)^3\text{H}$ ,  $^{14}\text{N}(\text{n}, ^3\text{H})^{12}\text{C}$ , and ternary fission (fission yield  $<0.01\%$ )]. Typically,  $^3\text{H}$  is present as tritiated water (HTO) and can become adsorbed into structural concrete from the surface inward where it will be held in a weakly bound form. However, a systematic analysis of a sequence of subsamples taken from a reactor bioshield using combustion and liquid scintillation analysis has identified two forms of  $^3\text{H}$ , one weakly bound and one strongly bound. The strongly bound tritium, which originates from neutron capture on trace lithium ( $^6\text{Li}$ ) within mineral phases, requires temperatures in excess of  $350\text{ }^\circ\text{C}$  to achieve quantitative recovery. The weakly bound form of tritium can be liberated at significantly lower temperatures ( $100\text{ }^\circ\text{C}$ ) as HTO and is associated with dehydration of hydrous mineral components. Without an appreciation that two forms of tritium can exist in reactor bioshields, the  $^3\text{H}$  content of samples may be severely underestimated using conventional analytical approaches. These findings exemplify the need to develop robust radioactive waste characterization procedures in support of nuclear decommissioning programs.

A wide range of early generation nuclear facilities (military and civil) are undergoing decommissioning worldwide. These include research reactors, civil nuclear power plants, isotope production plants, particle accelerators, experimental fusion facilities, and isotopic enrichment facilities. In the UK in particular, most first- and second-generation nuclear research sites and nuclear power plants are being decommissioned or are scheduled to begin decommissioning within the next decade.

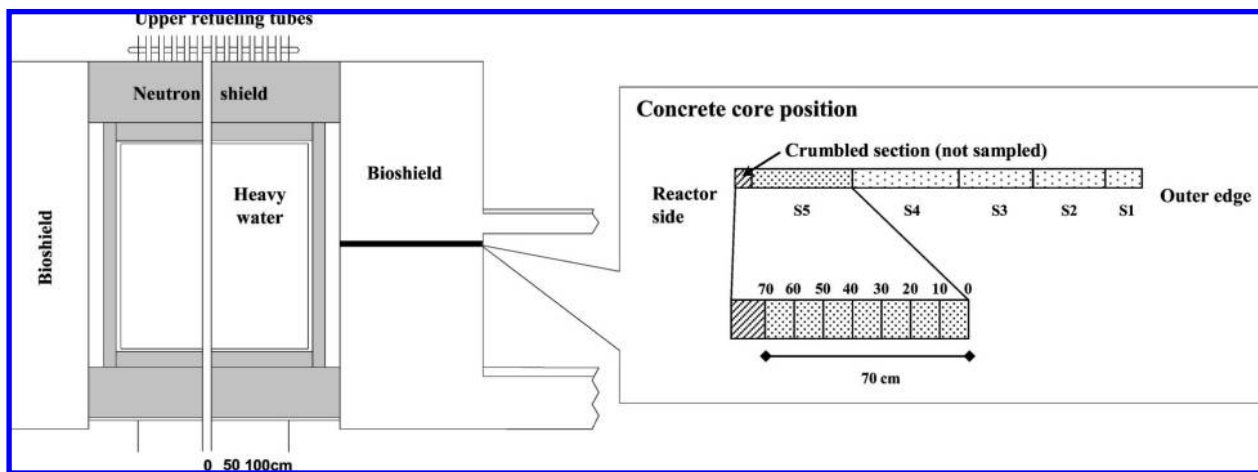
The nature of decommissioning works varies from site to site but typically involves the extensive clean out, refurbishment, or demolition of buildings and other facilities and remediation of the land. As a result, large volumes of potentially radioactive waste materials will be generated that require accurate characterization prior to waste sentencing. In many instances, novel radioanalytical approaches must be developed and validated to permit quantification of radionuclides in diverse and complex matrixes. Concrete, cements, and other cementitious materials account for 22% of the total weight of low-level radioactive waste and 12% of the total

weight of intermediate-level waste in the UK.<sup>1</sup> Concrete has been widely used in the nuclear industry due to its strength, durability, low cost, and good radiation shielding characteristics. Concretes, along with many other structural materials, become contaminated with  $^3\text{H}$  during routine operations as a result of the widespread presence of tritiated water (vapor) in reactor buildings coupled with its rapid diffusion into relatively porous materials. The high diffusivity of  $^3\text{H}$  makes prediction of activity concentrations difficult and there is therefore a need to directly determine  $^3\text{H}$  in concretes. In certain types of reactor design using  $\text{D}_2\text{O}$ , such as CANDU reactors and the now decommissioned SGHW reactor (Winfrith, UK), significant quantities of tritium were produced via neutron activation of deuterium ( $\text{D}_2\text{O}$  moderator) or lithium and ternary fission (0.01% fission yield). The quantities of tritium produced in heavy water reactors by neutron activation of deuterium exceeds that in light water reactors by almost 100 times.<sup>2,3</sup> Reactor bioshield concrete will also be directly exposed to neutrons resulting in the in situ formation of a range of radionuclides including  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{63}\text{Ni}$ ,  $^{60}\text{Co}$ ,  $^{36}\text{Cl}$ ,  $^{55}\text{Fe}$ ,  $^{41}\text{Ca}$ ,  $^{152}\text{Eu}$ , and  $^{133}\text{Ba}$ .<sup>4</sup> Of these, tritium is considered to be one of the critical radionuclides.<sup>5–7</sup> Previous studies of tritium in concrete,<sup>4,8–11</sup> have mainly focused on structural concretes exposed to tritiated water (HTO) and have not specifically considered bioshield concrete. Neutron activation of trace Li impurities in bioshield concrete provides an alternative mechanism for  $^3\text{H}$  production, and such  $^3\text{H}$  produced in situ may be expected to be present in a different, potentially less-available, form since it may be locked/trapped inside mineral lattices.

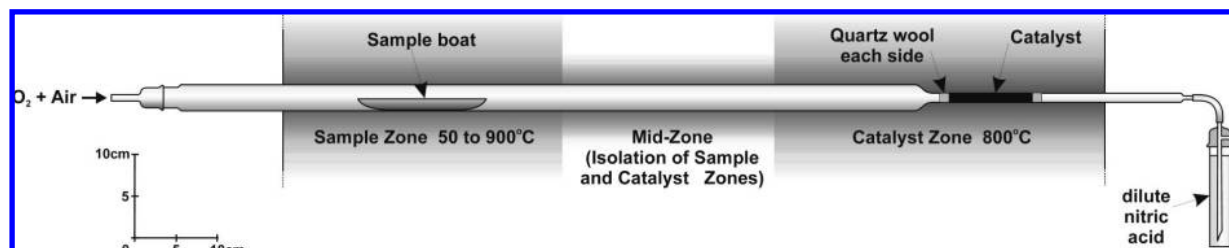
Direct nondestructive detection of  $^3\text{H}$  at low activity concentrations is difficult given the low-energy pure  $\beta$  emission of the

- (1) DEFRA(2002). Radioactive Wastes in the UK: A summary of the 2001 inventory. Report DEFRA/RAS/02.003. Department for Environment, F. a. R. A., London, UK, p 14.
- (2) Johnson, J. R.; Brown, R. M.; Myers, D. K. *Radiat. Res.* **1992**, *50*, 197–211.
- (3) Harolf, T. P. J.; Baker, D. A. *Fusion Technol.* **1985**, *8*, 2544–2550.
- (4) Hou, X. *Applied Radiation and Isotopes* **2005**, *62*, 871–882.
- (5) Kinno, M.; Kimura, K.; Ishikawa, T.; Miura, T.; Ishihama, S.; Hayasaka, N.; Nakamura, T. J. *Nucl. Sci. Technol.* **2002**, *39*, 215–225.
- (6) Bisplinghoff, B.; Lochny, M.; Fachinger, J.; Brucher, H. *Nuclear Energy—Journal of the British Nuclear Energy Society* **2000**, *39*, 311–315.
- (7) Bushuev, A. V.; Verzilov, Y. M.; Zubarev, V. N.; Kachanovskii, A. E.; Matveev, O. V.; Proshin, I. M.; Bidulya, L. V.; Ivanov, A. A.; Kalugin, A. K. *Atomic Energy* **1992**, *73*, 959–962.
- (8) Furuichi, K.; Takata, H.; Katayama, K.; Takeishi, T.; Nishikawa, M.; Hayashi, T.; Kobayashi, K.; Namba, H. *J. Nucl. Mater.* **2007**, *367–370*, 1243–1247.
- (9) Furuichi, K.; Takata, H.; Motoshima, T.; Satake, S.; Nishikawa, M. *J. Nucl. Mater.* **2006**, *350*, 246–253.
- (10) Numata, S.; Amano, H.; Okamoto, M. *J. Nucl. Mater.* **1990**, *171*, 350–359.
- (11) Numata, S.; Amano, H.; Minami, K. *J. Nucl. Mater.* **1990**, *171*, 373–380.

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**Figure 1.** Sampling of bioshield concrete.



**Figure 2.** Schematic diagram of the Pyrolyser triple zone furnace.

radionuclide ( $E_{\max} = 18.6$  keV). Therefore, standard laboratory procedures for the determination of  $^3\text{H}$  in solid matrixes will liberate  $^3\text{H}$  by aqueous leaching, distillation, freeze-drying, azeotropic distillation, or chemical/thermal oxidative decomposition prior to quantification of the liberated  $^3\text{H}$  usually by liquid scintillation counting. With the exception of oxidative decomposition, the techniques depend on the  $^3\text{H}$  being present as tritiated water, which is readily extractable. The efficacy of these methods could be significantly affected by the form of  $^3\text{H}$  present and in particular may not quantitatively recover  $^3\text{H}$  produced in situ via neutron activation of Li. There has been no previous attempts to compare the extractability of  $^3\text{H}$  in bioshield and structural concretes and to assess the impact that this may have for accurate analytical measurements, although measurement of total  $^3\text{H}$  in bioshield concrete has been previously reported using oxidative combustion.<sup>4</sup> This investigation aimed to determine whether different analytical approaches are required for these two concrete types, if a single method would be effective for all types, and to consider the implications for improper waste characterization prior to waste sentencing.

## EXPERIMENTAL SECTION

A cylindrical concrete core, traversing the reactor bioshield, was taken from the 100 MW steam generating heavy water reactor (SGHWR), which was operated by the United Kingdom Atomic Energy Authority at Winfrith, Dorset, from 1967 to 1990 (Figure 1). The core was sectioned at 10 cm intervals and its outer surface, which may have been compromised during coring, was removed and discarded. Core subsections were crushed to ensure a homogeneous sample suitable for analysis and 30 g of sample were transferred to a 20 mL polythene vial.  $\gamma$ -Emitting radionuclides were determined

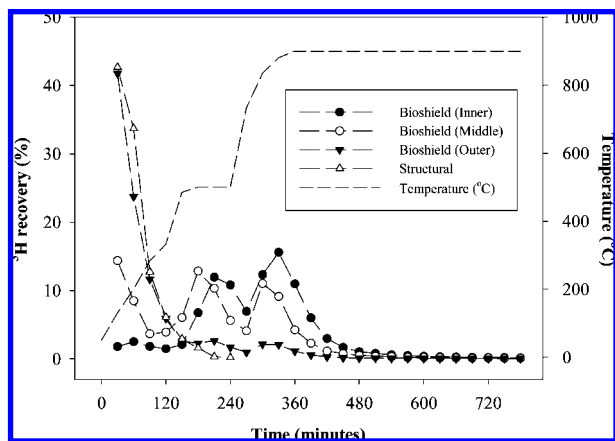
using a Canberra 40% well-type HPGe  $\gamma$ -spectrometer previously calibrated against a matrix-matched mixed radionuclide standard of identical geometry (QCYK8563). Spectral deconvolving was performed using Fitzpeaks software (JF Computing). Additional corrections for cascade-summing effects were applied for  $^{152}\text{Eu}$ . Total tritium activity concentrations and  $^3\text{H}$  thermal evolution profiles were determined on each of the subsections.

Structural concrete samples and other construction materials used in the leaching trials were taken from the nuclear reactor site (Winfrith) and from other non-nuclear sites for comparative purposes. These materials had not been exposed to a neutron flux and the  $^3\text{H}$  present originated from exposure to tritiated water vapor only.

Tritium extraction was achieved using a Raddec Pyrolyser Trio System (Figure 2), which permits simultaneous extraction/oxidation of up to six samples. This system was designed specifically for quantitative extraction of tritium and  $^{14}\text{C}$  from nuclear and environmental samples.

Each sample was heated using a preset heating cycle (Figure 3) in its own silica glass work tube that carried a stream of air/oxygen. The liberated gases pass through a zone containing 10 g of platinumized alumina catalyst that oxidizes any organic combustion products to  $\text{CO}_2$  and  $\text{H}_2\text{O}$  (and HTO). Any water vapor, including HTO, is trapped in a bubbler containing 20 mL of 0.1 M  $\text{HNO}_3$ . Bubbler changes are made every 30 min to determine the  $^3\text{H}$  evolution profile, and the gas supply is isolated during the bubbler changeover. Reported temperatures refer to that recorded by the furnace thermocouple, and the sample temperature is typically 10–20% lower than the furnace temperature during the ramping stages but <5% lower during the hold stages.

The amount of tritium emanated at various temperatures was also investigated by heating subsamples of concrete to set



**Figure 3.** Tritium evolution profiles for structural and bioshield concrete.

temperatures for 12 h and then allowing the samples to cool to room temperature. The  $^3\text{H}$  content and evolution profile of the  $^3\text{H}$  remaining in the sample after this heating stage was then determined as described previously.

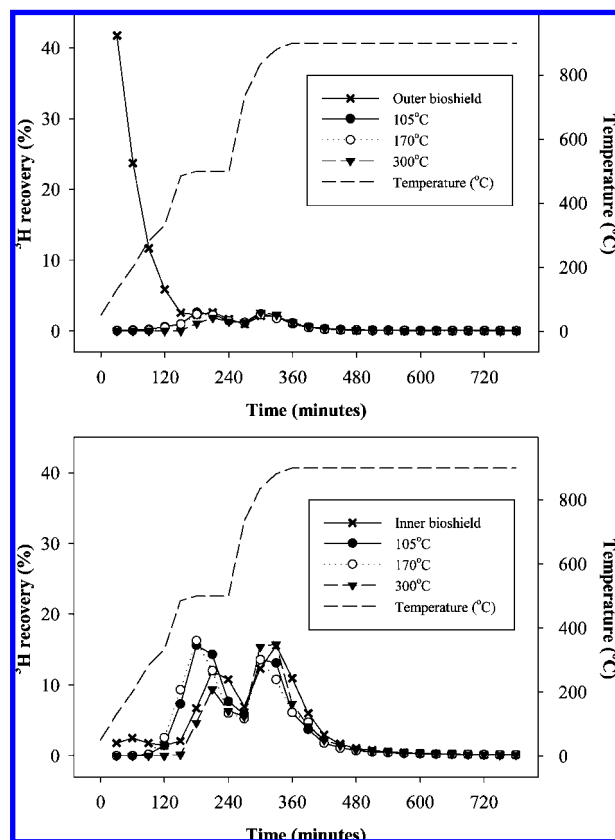
The aqueous leachability of  $^3\text{H}$  from the bioshield concrete was compared with that from a range of other building materials sampled from the SGWH reactor building and also from a non-nuclear HTO handling facility that had been exposed to HTO vapor in situ (but which were not exposed to a reactor neutron flux). Approximately 1 g of solid material was mixed with 10 mL of water for a defined time with constant agitation. Bioshield concretes were leached for 12 h while all other materials were leached for 2 h. The samples were then filtered, and the  $^3\text{H}$  content of the filtrate was determined by liquid scintillation counting.

All tritium measurements were performed using a 1220 liquid scintillation counter (Wallac Quantulus). Eight milliliters of aqueous sample was mixed with 12 mL of Gold Star (Meridian) scintillation cocktail in a 22 mL Polythene vial. The counter was calibrated for  $^3\text{H}$  using a traceable tritiated water standard (TRY-44, Nycomed Amersham PLC, Bucks, UK). For all samples, the measured quench levels (SQPE) ranged from 700 to 850, which correspond to  $^3\text{H}$  counting efficiencies of 17–31%. All uncertainties are quoted at the 95% confidence level and refer to propagated method uncertainties.

Lithium concentrations in the bioshield concrete subsections were measured to allow calculation of the theoretical quantity of  $^3\text{H}$  produced via the  $^6\text{Li}(n,\alpha)^3\text{H}$  reaction. Stable Eu was also determined to permit the calculation of theoretical  $^{152}\text{Eu}$  production and, hence, the expected  $^3\text{H}/^{152}\text{Eu}$  ratio. Approximately 1 g of concrete was accurately weighed and completely digested using a combination of  $\text{HNO}_3$  and  $\text{HF}$ . The residue was dissolved in 2%  $\text{HNO}_3$  and diluted to reduce the total dissolved solid content. Li concentrations in the digest were determined using ICP-AES while Eu concentrations were determined by ICPMS. Measurement uncertainties were typically  $\pm 3\%$  relative standard deviation ( $2\sigma$ ).

## RESULTS AND DISCUSSION

Two distinct stages of  $^3\text{H}$  loss were identified for the bioshield concrete. In concrete located further away from the reactor core (outer bioshield), where neutron fluxes would have been relatively low, tritium was liberated in the early stages of the combustion cycle at  $<350^\circ\text{C}$ . The tritium evolution profile was similar to that



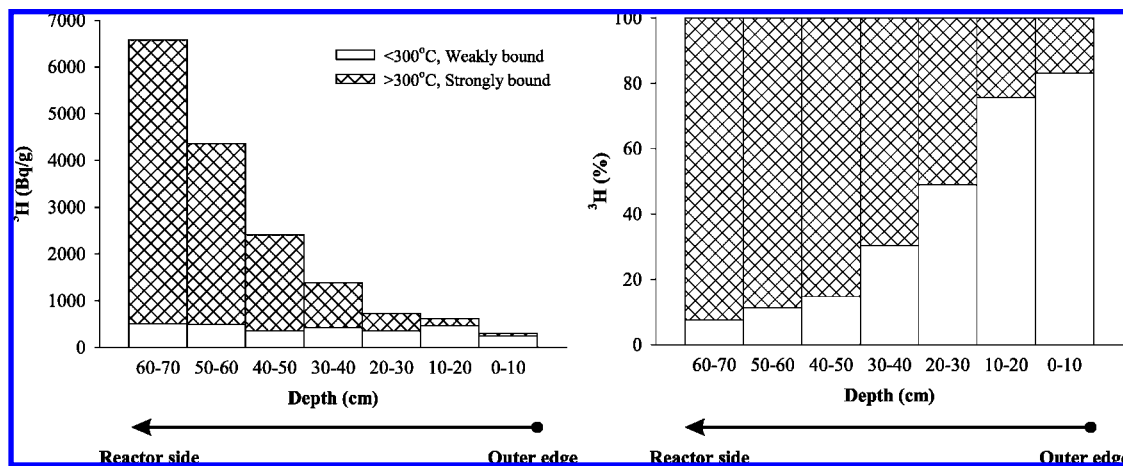
**Figure 4.** Thermal profiles showing the proportion of weakly bound tritium in inner and outer bioshield concrete.

seen for structural concrete (nonirradiated concrete) where  $^3\text{H}$  contamination arose from exposure of the concrete to HTO vapor. For the concrete subsamples located nearer to the reactor core (inner bioshield) and which had been exposed to higher neutron fluxes, the tritium evolution mostly occurred over a  $500\text{--}900^\circ\text{C}$  temperature range (Figure 3). The two distinct forms of tritium are subsequently referred to as “weakly bound  $^3\text{H}$ ” and “strongly bound  $^3\text{H}$ ”, respectively.

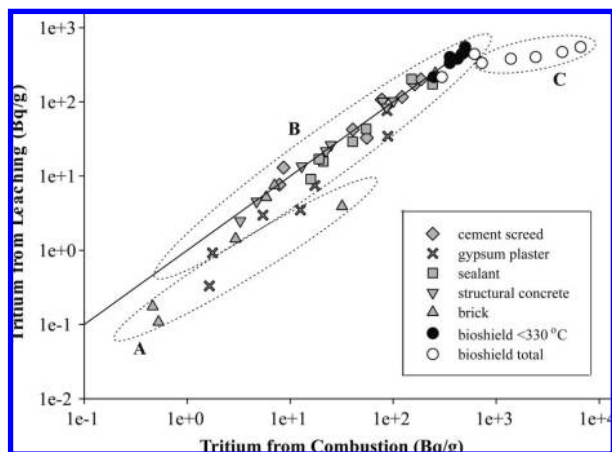
The temperature of  $^3\text{H}$  evolution will depend on whether the  $^3\text{H}$  is present in the concrete as free water, water of crystallization, structural OH groups, or otherwise lattice-bound  $^3\text{H}$ . Some earlier studies<sup>12</sup> have identified that water loss from concrete occurs in three key transition stages at approximately 105, 170, and  $300^\circ\text{C}$ . In the present study, the  $^3\text{H}$  evolution profiles of the concrete subsamples previously heated to these key temperatures for 12 h were compared with that for nonthermally treated bioshield concrete (Figure 4). Heating the concrete to  $105^\circ\text{C}$  resulted in the complete loss of the weakly bound tritium component, which is dominantly present in outer bioshield concrete. However, the strongly bound tritium, which is the main component of the inner bioshield concrete, was not significantly affected by any preliminary heating to 105 or  $170^\circ\text{C}$ . The quantitative removal of weakly bound  $^3\text{H}$ , following drying of the sample at  $105^\circ\text{C}$ , suggests that the more prolonged release of  $^3\text{H}$  observed up to  $300^\circ\text{C}$  in the evolution profile arises from a slow rate of release as opposed to a strongly bound form of  $^3\text{H}$  requiring higher temperatures to

(12) Alarcon-Ruiz, L.; Platret, G.; Massieu, E.; Ehrlicher, A. *Cem. Concr. Res.* 2005, 35, 609–613.





**Figure 5.** Proportions of different types of tritium along a section of bioshield concrete core.



**Figure 6.** Relationship between  $^3\text{H}$  extracted from various construction materials by aqueous leaching and combustion. Zone A, partial  $^3\text{H}$  leaching correlating with total  $^3\text{H}$ ; zone B, quantitative leaching of  $^3\text{H}$  correlating with total  $^3\text{H}$ ; zone C, partial leach not correlating with total  $^3\text{H}$  (bioshield samples).

effect decomposition and indicates that the weakly bound  $^3\text{H}$  is present as adsorbed water.

The total  $^3\text{H}$  concentration in bioshield concrete is highest near the reactor core and decreases exponentially with distance from the reactor. However, when the tritium species in the bioshield concrete are considered, the relative proportion of strongly bound tritium decreases exponentially while the weakly bound tritium increases with increasing distance from the core (Figure 5). The activity concentration of weakly bound  $^3\text{H}$ , however, remains approximately constant throughout the core. The different proportions of the two forms of tritium identified suggest different origins.

A simple aqueous leach test is often employed to assess the level of  $^3\text{H}$  contamination in a solid sample. A comparison of leached  $^3\text{H}$  versus total  $^3\text{H}$  ( $^3\text{H}_{\text{total}}$ ) highlights three different relationships. For the majority of sample types exposed to HTO (including structural concrete), the leached  $^3\text{H}$  compares very well with the  $^3\text{H}_{\text{total}}$  as determined by combustion (Figure 6 zone B). Similar results are also observed between leaching and the weakly bound  $^3\text{H}$  component of the bioshield concrete samples (Figure 6 •). For certain materials (notably brick, plaster and sealant), the  $^3\text{H}$  leach results were lower than the  $^3\text{H}_{\text{total}}$  although a correlation still existed between the two measurements (Figure

6 zone A). The low bias may indicate a slow leach rate and may be improved by extending the leach time. However, no correlation was observed between the leached  $^3\text{H}$  and the total  $^3\text{H}$  as measured by combustion (Figure 6 zone C) suggesting that the leaching technique was not capable of extracting the strongly bound  $^3\text{H}$  fraction in the bioshield concrete.

The systematic analysis of a sequence of subsamples taken from the examined core showed tritium evolution profiles that varied with distance from the reactor. In concrete sampled further away from the reactor (outer bioshield), rapid tritium loss at lower temperature occurs that is similar to that seen for structural concrete (nonbioshield concrete). Conversely, for the concrete closer to the reactor (inner bioshield concrete), the rate of HTO lost at low temperature is considerably reduced and temperatures in excess of 860 °C were required to completely liberate the  $^3\text{H}$ .

Concrete is a permeable hydrophilic material typically formed from reacting Portland cement, sand, water, and rock aggregates. However, in terms of the tritium capacity of concrete, the cement is the most significant component owing to its abundant structural water capable of trapping tritium by isotopic exchange.<sup>8</sup> The hydrated cement is composed of four major compound classes: tricalcium silicates ( $\text{C}_3\text{S}$ ), dicalcium silicate ( $\text{C}_2\text{S}$ ), tricalcium aluminate ( $\text{C}_3\text{A}$ ), and tetracalcium aluminoferrite ( $\text{C}_4\text{AF}$ ). The most important products of the hydration reactions are the calcium silicate hydrate ( $\text{C-S-H}$ ) and portlandite (also called calcium hydroxide,  $\text{CH}$ ).<sup>13</sup> Thermogravimetric analysis and differential thermoanalysis curves for the Portland cement show major mass losses in the temperature range <200 °C (release of free water), 200–450 °C (release of water of crystallization), 450–550 °C (decomposition of  $\text{Ca}(\text{OH})_2$ ), and 600–850 °C (decomposition of hydrated calcium silicates).<sup>10,12</sup> The proportions of  $^3\text{H}$  lost at the key transition temperatures from inner and outer bioshield concrete were compared with other published data (Table 1).<sup>10</sup> The relative proportion of  $^3\text{H}$  in cement exposed to HTO vapor, and of  $^3\text{H}$  in outer bioshield lost at <200 °C and 200–450 °C, agrees well with previously published data indicating that the weakly bound  $^3\text{H}$  is distributed as expected between free water and water of crystallization. However, the proportion of  $^3\text{H}$  lost at key transition temperatures in inner bioshield concrete does not

(13) Sha, W.; O'Neill, E. A.; Guo, Z. *Cem. Concr. Res.* **1999**, *29*, 1487–1489.

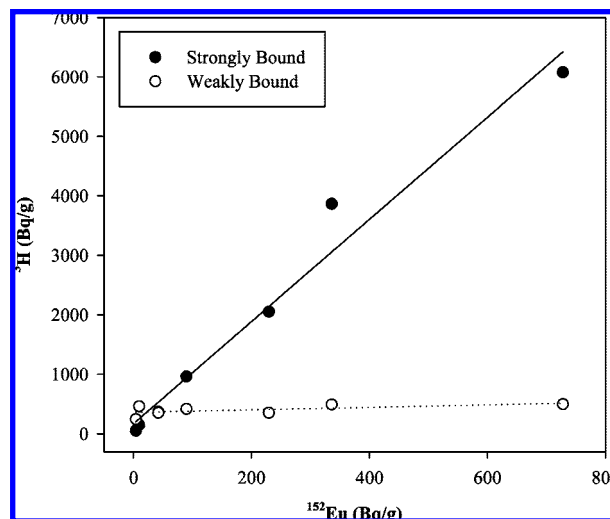
**Table 1. Comparison of  $^3\text{H}$  and Water in Concrete and Cements**

water group	temp ( $^{\circ}\text{C}$ )	Numata et al. <sup>a</sup>		present study (Bioshield)		
		% of $^3\text{H}$ in cement material	% of total water liberated	% of $^3\text{H}$ of outer (0–10 cm)	% of $^3\text{H}$ of inner (60–70 cm)	temp ( $^{\circ}\text{C}$ )
liquid water (free and capillary water)	<200	77	65	66	4	<200
water of crystallization	200–450	15	20	20	5	200–485
water constituent (calcium hydroxide)	450–550	7	12	7	29	485–500
water constituent (calcium silicate hydrate)	600–850	1	3	8	61	500–900

<sup>a</sup>  $^3\text{H}$  data obtained from cement contaminated by tritiated water vapor.<sup>10</sup>

correlate with that observed for concretes exposed to HTO with  $\sim 90\%$  of the  $^3\text{H}$  being liberated at  $>485^{\circ}\text{C}$ .

The strongly bound  $^3\text{H}$  is inferred to originate from the  $^6\text{Li}(n,\alpha)^3\text{H}$  reaction ( $\sigma_{\text{th}} = 940\text{ b}$ ), which has been previously suggested as the dominant mechanism for  $^3\text{H}$  production in bioshield concrete.<sup>5,14</sup> Though this reaction has been previously identified, this is the only study that has examined the relative behavior (through thermal loss patterns) of different tritium types in concrete and considered the impact. The concretes analyzed in this study typically contain  $12 \pm 1\text{ mg/kg Li}$ , which is comparable with previously reported values.<sup>5</sup> Conversion of  $^2\text{H}$  to  $^3\text{H}$  is not considered significant given the low thermal neutron capture cross section of  $0.508 \pm 0.015\text{ mb}$  for this reaction.<sup>15</sup> Predicted maximum  $^3\text{H}$  activity concentrations arising from  $^2\text{H}(n,\gamma)^3\text{H}$  (based on MCNP model neutron flux data supplied by UKAEA) for the 60–70 cm fraction are  $<0.01\text{ Bq/g}$  compared with  $15\,000\text{ Bq/g}$  via Li activation. Previous studies<sup>5</sup> have investigated the theoretical correlation between  $^3\text{H}$  and  $^{152}\text{Eu}$  in neutron-irradiated concretes. Europium-152 is produced via neutron activation of stable  $^{151}\text{Eu}$  (47.7% of natural Eu) with a high thermal neutron capture cross section of 5900 barns and provides a good indicator of neutron flux. The comparable half-life of  $^{152}\text{Eu}$  (13.54 years) and  $^3\text{H}$  (12.33 years) means that the  $^3\text{H}/^{152}\text{Eu}$  ratio should be predictable for a given  $[\text{Li}]/[\text{Eu}]$  ratio irrespective of the irradiation time. Strongly bound  $^3\text{H}$  activity concentrations measured in the bioshield concrete correlate with  $^{152}\text{Eu}$  and hence neutron flux (Figure 7), which further supports the proposition that the  $^3\text{H}_{\text{strong}}$  originated from neutron activation. The  $^3\text{H}_{\text{strong}}/^{152}\text{Eu}$  ratio of 8.6 is low compared with the value of  $\sim 27$  reported for materials with a similar stable Li/Eu concentration ratio<sup>5</sup> (mean  $[\text{Li}]/[\text{Eu}] = 44$ ). This may indicate that not all  $^3\text{H}$  formed in this manner is strongly bound or that the strongly bound  $^3\text{H}$  slowly converts to the more loosely bound form (e.g., via migration out of mineral lattices) with time and which subsequently diffuses through the bioshield. The weakly bound  $^3\text{H}$  probably arises from a combination of diffusion of HTO from the reactor core into the concrete and a slow conversion of strongly bound  $^3\text{H}$  to a more weakly bound form. The presence of significant quantities of weakly bound  $^3\text{H}$  at distance from the neutron flux confirms that diffusion controls the distribution of this form of  $^3\text{H}$ . Furuichi et al.<sup>9</sup> observed a pronounced  $^3\text{H}$  diffusion profile in cement pastes exposed to tritiated water vapor after 5 days, which flattened to a more evenly dispersed profile to depths of 8 cm after only 318 days. Given the operational period of the reactor and the time that has elapsed since shutdown, it is feasible that  $^3\text{H}$  originating



**Figure 7.** Relation between measured  $^3\text{H}$  and  $^{152}\text{Eu}$  activity concentrations.

from the reactor has had sufficient time to diffuse to relatively uniform concentration throughout the bioshield.

The presence of a hitherto uncharacterized, strongly bound component of  $^3\text{H}$  in bioshield concretes has significant implications for accurate analytical characterization of  $^3\text{H}$  in such materials. For quantitative analysis of  $^3\text{H}$  in bioshield concretes, a thermally aggressive treatment is required to quantitatively liberate the  $^3\text{H}$ . Typically, this should involve heating samples up to  $900^{\circ}\text{C}$ . For other sample types, the temperature that  $^3\text{H}$  is completely liberated from the sample will depend on the chemical form of the  $^3\text{H}$  and the physical properties of the sample material. The necessity for determining thermal evolution profiles for specific materials prior to commencing routine analysis is clearly highlighted from these studies. The study also demonstrates that although a simple leaching technique can be used to provide rapid screening of  $^3\text{H}$  activity concentrations in a range of matrixes, it is not appropriate in all instances and should be used with caution.

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(14) Kinno, M.; Kimura, K.; Nakamura, T. *J. Nucl. Sci. Technol.* **2002**, *39*, 1275–1280.

(15) Jurney, E. T.; Bendt, P. J.; Browne, J. C. *Phys Rev. C* **1982**, *25*, 2810–2811.