SIMULATION AND ASSESSMENT OF THE PERFORMANCE CHARACTERISTICS OF A HIGH CAPACITY ION-EXCHANGE SYSTEM FOR THE DECONTAMINATION OF SPENT FUEL COOLING-POOL WATER

1.0 TOPIC

BNFL Magnox Electric has recently pioneered a novel submersible caesium removal system for installation in the spent-fuel cooling pools of its Magnox fleet of reactors. This cartridge-based plant is capable of the decontamination of up to 37 m^3/h of pool-water with high decontamination efficiency for a period of months. It is intended that the use of this technology will reduce operator doses, minimise the environmental impact of authorised discharges and enable waste volume minimisation. It is expected that the volume of operational intermediate level waste arisings associated with radiocaesium removal will be reduced to approximately 2% of the previous level. The high performance of this inorganic system is a consequence of the exceptional selectivity and kinetics of a synthetic zeolite material known as a crystalline silicotitanate (CST). The genre of crystalline silicotitanates was first reported in 1993[1] following research at Sandia National Laboratory. These materials were designed to provide extensive waste minimisation in the processing of defense wastes containing high sodium content and high pH. The optimised interplanar spacing of 7.8 Å provides size selectivity for hydrated caesium species to enter the crystalline structure, ion-exchanging for sodium atoms without subsequently allowing significant back-exchange.[2] Strontium is similarly absorbed, however it is held less tightly than caesium and may be desorbed in the presence of high salt, high pH solutions. One such CST is marketed by UOP as IONSIV IE-911; in batch contact trials and simple kinetic studies it was identified as the optimum material for decontamination of Magnox pool-water.

Previous studies report the use of CSTs for the decontamination of aqueous wastes at a number of locations in the US,[2] however those applications differed from ours in that the waste requiring decontamination contained high levels of dissolved salts. In contrast, Magnox pool-water contains *ca.* 340 parts-per-million (ppm) of sodium and only part-per-billion (ppb) levels of other species. The concentration of caesium ions, even at the operational limit for ^{137}Cs, is only 0.31 ppb and the concentration of strontium is an order of magnitude lower. Under these conditions the lifetime of a cartridge containing 30 litres of Ionsiv IE-911 adsorbent under high throughput conditions (*ca.* 100,000 m^3 per cartridge) is not dependent upon the ion-exchange capacity of the material. Under these circumstances the operational lifetime of a bed of Ionsiv IE-911 adsorbent is not governed by the ion-exchange capacity. Rather, the lifetime is a function of the selectivity for caesium (or strontium) over sodium, *ie.* the distribution coefficient (K_d) for that system. Hence, the maximum volume of pond water that can be treated before the system's removal efficiency drops to below the required value determines the operational lifetime of such a bed. It is said to be 'throughput-limited' rather than 'capacity-limited'. This paper presents novel methodology for the simulation of the lifetime operation of a throughput-limited system.

2.0 BASIS FOR ORIGINALITY

2.1 Plant

The novel submersible system comprises a pre-filter, two adsorbent cartridges in parallel and a post-filter. The cartridges operate in an in-to-out mode and contain an annulus of adsorbent, the volume of which is a compromise between operational requirements and dose-rate limitation of the spent cartridge. The high volume setting of the operational system (37 $m^3.h^{-1}$) equates to a face-velocity at the inner annulus of the adsorbent bed of 2 $cm.s^{-1}$ and a standard setting of 18.5 $m^3.h^{-1}$ equates to a face-velocity of 1 $cm.s^{-1}$. It is believed that this is the most compact and versatile plant available, capable of treating extremely high volumes of wastewater under continuous, protracted operation. The plant is operated in a re-circulating mode, submerged in a cooling pool. As such, absolute efficiency is less important than treatment rate and a decontamination factor of >75% will be fit for purpose.

2.2 Experimental Design

Our laboratory was required to determine the operational efficiency and to determine the radionuclide composition of the spent Ionsiv IE-911 adsorbent. As will be seen below, it is not possible to measure the radionuclide inventory outside of a hot-cell, so we developed some novel methodology to enable this work to be undertaken in a standard active laboratory environment.

The main problem in mimicking the operation of an ion-exchange bed over a period of several months in the laboratory is that it is not a simple matter to continuously input activity to a bed of large enough cross-sectional area required to successfully replicate cartridge operation. The volume of simulated pond water required to achieve a face-velocity of 1 $cm.s^{-1}$ for a column of 20 mm diameter is 271 litres per day. Securing such a supply would be impractical on a logistical and dose (ALARP) basis. Experimental simplicity can however be achieved by exploiting the throughput-governed character of the caesium-CST relationship. Since essentially none of the Ionsiv IE-911 adsorbent will be spent at the end of its operational lifetime (*ie.* have reached ion-exchange capacity) it is possible to simulate continuous activity input by transferring a single spike of activity to the bed and passing *inactive* pond water simulant through the bed in a cycling system. By monitoring the change in the activity distribution of this single spike over time it is possible to mathematically model the build-up of activity throughout the cartridge. Moreover, by monitoring the relative activity of segments of adsorbent which are beyond the path length of the cartridge it is possible to model the percentage breakthrough and hence the decontamination efficiency versus bed lifetime.

Figure 1 shows the simulated caesium breakthrough curve resulting from treating the experimental data obtained at a face-velocity of 1 $cm.s^{-1}$ in the manner described above. It is apparent that after 100 days of simulated operation at this ‘optimum’ throughput the radiocaesium removal efficiency was in excess of 83%. This means that the treatment volume of 37 $m^3.h^{-1}$ of pond water will be decontaminated to 17% of the inlet activity after this length of operation. At the lower throughput rate, corresponding to 18.5 $m^3.h^{-1}$ in plant operation the decontamination efficiency profile was very similar, indicating a film-diffusion ion exchange mechanism. This experiment is ongoing and updated data will be presented.

This methodology has the additional benefit of simulating the activity distribution profile (Figure 2) through the Ionsiv IE-911 adsorbent annulus. Due to the high levels of activity (*ie*. up to 400 Ci of radiocaesium) accumulated throughout the lifetime of a cartridge the shielding requirements of the spent cartridge are paramount. Hence, it was important to have this knowledge of the anticipated activity distribution prior to undertaking radiological calculations to derive appropriate shielding.

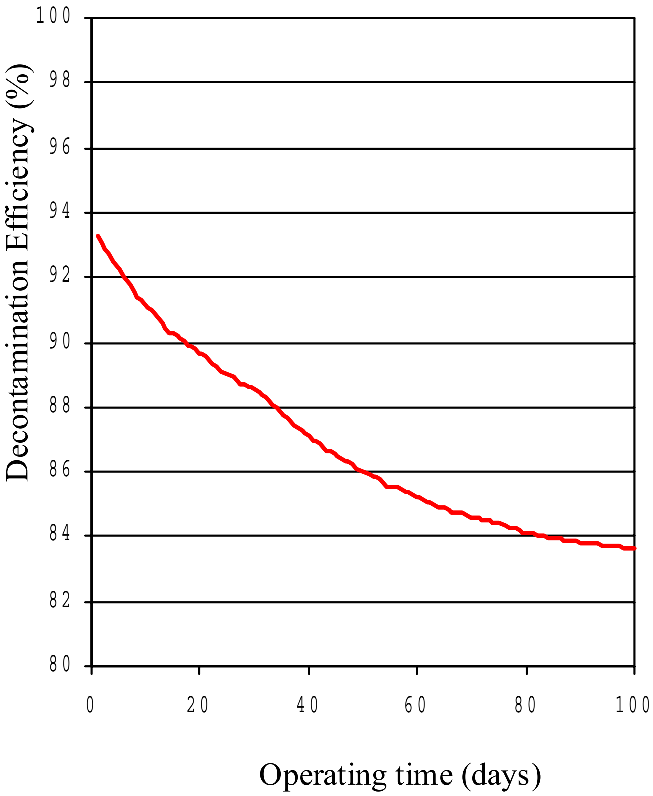

Figure 1. Simulated Efficiency Profile (Outlet : Inlet) Versus Time for the Submersible Caesium Removal System

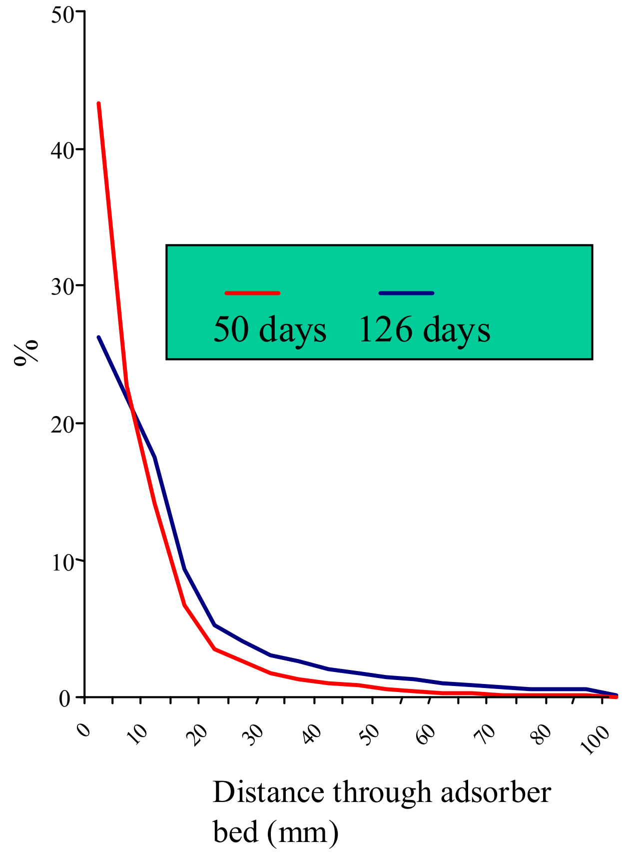

Figure 2. Graphical Representation of the Simulated Activity Profile for a High Specificity Ion Exchange System based on UOP's IONSIV IE-911.

3.0 CONCLUSIONS

BNFL Magnox Electric's novel submersible decontamination system is due to go operational during mid-2001. This study presents new methodology and data treatment for the simulation of the operational performance of such throughput-governed ion exchange systems. It is predicted from this work that the decontamination efficiency of a single pair of cartridges using Ionsiv IE-911 will be in excess of 80% for a period of months. The study is ongoing and the latest data will be presented.

4.0 REFERENCES

1. Anthony, R.G., C.V. Philip, and R.G. Dosch, *Selective Adsorption and Ion Exchange of Metal Cations and Anions with Silico-Titanates.* Waste Management (N.Y.). **13**: p. 503-512. (1993).

2. Klavetter, E.A., et al., Ion-Exchange Performance of Crystalline Silicotitanates for Cesium Removal from Hanford Tank Waste Simulants, in "Ion-Exchange Performance". p. 709-713 (1994).