

THEORETICAL INVESTIGATION OF ASPECTS OF RADIOACTIVE CONTAMINATION



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Abstract

The BNFL programme of work has investigated theoretical aspects of the mechanisms responsible for the deposition and adherence of contamination to metallic surfaces and the energetics of physical decontamination processes. The work has been conducted in two phases:

- The theoretical and laboratory study of deposition of species from aqueous media on to stainless steel
- Theoretical assessment of the forces causing the attraction of PuO_2 and UO_2 particles to stainless steel in an air environment and comparison of these forces with the energies delivered by physical jetting processes.

The first phase produced a model which was found to give good agreement with plant operational experience of the deposition of simple aqueous ions such as Cobalt. Due to the complexities, however, of surface / colloid and surface / particle interactions the model was found not to be successful at predicting deposition for more complex compounds, such as Ruthenium Nitrosyls. At this stage the model had fulfilled its original requirement of underpinning design work on pipework shielding systems and it was decided not to pursue the library of chemical speciation data that would be necessary to model the behaviour of a full spectrum of possible contaminants.

The second phase predicts by theoretical analysis that the relation of the energy delivered by jetting techniques to the physical forces causing the adherence of PuO_2 and UO_2 particles will vary considerably with particle size. This is particularly notably for larger PuO_2 particles which are firmly held as a result of high levels of electrostatic charge due to their intense alpha activity. Small particles tend to be difficult to remove due to the low profile that they present to the jetting medium. Large and small PuO_2 particles and small UO_2 particle are thus predicted to be difficult to remove and will present an energy threshold which may not be crossed by all decontamination techniques.

Introduction

This study addresses the theoretical assessment of the mechanisms of the contamination of stainless steel surfaces by radioactive species. This is intended to complement the more practical projects that form the basis of the present Research Group, with the aim of facilitating the selection and optimisation of decontamination processes for specific contamination scenarios. Two different types of contamination are considered; adsorption from aqueous solution and particle deposition in atmospheric air. Worked examples have concentrated on Cobalt for the aqueous solution work and Uranium and Plutonium Oxide for the particle deposition work.

Much of the work was performed by BNFL Research and Development teams, and draws upon practical plant operational experience at BNFL's Sellafield works. Experimental work has also been performed at Kings College, University of London.

Background

The initial phase of the study grew from the necessity to have a model to understand the accumulation, or plate out, of active species on pipework at the Sellafield plant to support the design of appropriate shielding. The information gathered as part of that investigation was developed into a model which assisted the understanding of the mechanisms by which radioactive cations became attached to the surfaces of process plants. The aim of the model was to identify parameters which affected the adsorption and desorption mechanisms, and hence to selectively target these to improve decontamination performance; or better, to prevent contamination in the first place.

The second phase of the study addressed the problem of accumulation of dry particles of Uranium and Plutonium to stainless steel surfaces and compares the theoretically calculated energies necessary to remove these particles with those delivered by commonly used jetting techniques.

Technical Approach

The two phases of the study were largely theoretically based. The first phase started with the derivation of a model to describe deposition of Cobalt from aqueous solution on to stainless steel. The model considers the interactions between hydrated Co^{2+} ions at a pH range of 2-14 and the Cr rich Oxide layer covering the surface of the steel. Deposition of Cobalt was also measured experimentally under laboratory conditions and these results compared to plant data relating to the accumulation of plate out of active species on pipework at the Sellafield plant.

The results of the modelling work were found to give reasonable agreement with the observed behaviour of Cobalt, but it was less successful for more complex species. It was felt at this stage that the modelling had fulfilled its original requirement of underpinning design work on pipework shielding systems. Considerable effort would be necessary to compile a library of chemical speciation data to enable the model to address a full range of likely contaminants and this was not considered worthwhile.

For this reason the second phase of the study involved a change of direction and has investigated a problem facing all decommissioners of nuclear facilities; that of deposition of Uranium and Plutonium Oxides on stainless steel surfaces. Information from the scientific literature and BNFL's own data was used to enable first principle calculation of the forces binding these particles to steel surfaces and to allow comparison of the energetics of particle removal with the energy delivered by simple jetting techniques.

Results

The following are the principal findings from the study:

- The plate out of material on to a stainless steel surface is controlled by the pH of the liquor modifying the surface potential of the surface oxide layer. This potential interacts with the charged ion or surface potential of colloids or solids.
- Plate out for ions and colloids is a non-linear function of isotopes in solution. For suspended solids this is less obvious since saturation of surfaces at specific levels for certain solids has been demonstrated.

- Once deposited it is suggested that reversible, dynamic equilibrium, chemical reactions exist at the oxide surface. Equilibrium conditions have been observed to occur (experimentally) within a matter of days.
- Stainless Steel surface damage is believed to 'capture' ions and micron size particles and influence the equilibrium by delaying the release of captured material.
- Transient conditions in pH or concentration, that occur for short periods (eg: 24 hours) should not lead to a major plate out problem. (An element of judgement is necessary here, due to the complexity of surfaces interactions to short term changes).
- The influence of liquid turbulence can be ignored.
- Currently there is no data from experiments to predict or estimate deposition of fuel fines or floc. Upper bound magnetite surface saturation data is available.
- Multi layer deposition of ions, colloids and solids has not been identified as excessive. All data presented is for equilibrium conditions which takes due account of multi-layer deposition.
- UO_2 and PuO_2 particles are held on to stainless steel surfaces by a range of physical and chemical forces.
- The removal of particles held by chemisorption require energy several orders of magnitude higher than for particles held by physical forces alone.
- For physically bound PuO_2 , the image-charge force is the most significant physical force for particles above 10 microns. This is several orders of magnitude greater than that on UO_2 particles in this size range, and is due to the α -activity of the PuO_2 .
- Total physical forces acting on a particle do not vary significantly with the orientation of the substrate. This is because the Van der Waals and image-charge forces, which always favour adhesion, dominate the gravitational force for the particle size range considered (up to 20 microns).
- UO_2 particles larger than 20 μm will not be deposited on a horizontal surface from below as the gravitational force on larger particles is stronger than the Van der Waals and image-charge forces. For PuO_2 particles of 20 microns, the image-charge force is significantly greater than gravity and thus these particles will be strongly adhered.
- In comparison with the energy delivered by jetting techniques, physically adsorbed PuO_2 particles show a critical intermediate size range which is easier to remove than larger or smaller particles. In the case of PuO_2 , particles between approximately 0.3 and 3 microns can be removed by 15 bar water jetting at 0.5 m stand off distance, whilst larger and smaller particles will not be removed.
- Air jetting can remove only the particles adsorbed by physisorption. Particle sizes in the range of 0.1 to 2.5 μm can be removed with velocities of the order of few cm/s. Higher velocities are required for particles larger than 10 μm and also for particles smaller than 0.1 μm .
- UO_2 particles larger than 10 μm require air velocities ten to sixty times higher than those for UO_2 , due to the strong image-charge forces experienced by PuO_2 .

REFERENCES

1. Information on Cobalt deposition taken from J B Taylor; 1990 Thesis: A Theoretical Analysis of the Deposition of Radioactive Ions, Particles and Colloids from Aqueous Liquids on to Stainless Steel
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