Stability of Fukushima Diiachi Fuel Debris and Corium Bearing Micro-particles in Exclusion Zone Soils

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Project Background and Overview:

The Fukushima Diiachi Nuclear Power Plant (FDNPP) accident released an estimated 5.2×10^{17} Bq of radioactivity into the environment. Wet and dry deposition of Cs and other volatile radionuclides resulted in contamination of large areas of land around the FDNPP site. Approximately 100,000 people were subsequently displaced from their homes due to the establishment of a nuclear exclusion zone, and the estimated cost of compensation, environmental cleanup, and decommissioning now stands at over 20 trillion Yen (~ £ 150 billion GBP).

Recently, work from Kyushu University and Manchester has shown that reactor fuel debris and molten reactor core materials (corium) formed during the FDNPP accident and fragments of these materials were ejected from the damaged reactors **[1, 2]**. These radioactive hot particles have been found in exclusion zone soils and they typically are 1-10 µm in size, metal rich (Zr, Fe, Zn), contain U at wt % levels (1 - 90 wt %; often as UO_2 or Zr-U-Fe phases), and also include other reactor fission products (e.g. ^{99}Tc).

Our initial work **[1, 2]** has shown that detailed forensic characterisation of these particles can yield otherwise unobtainable diagnostic information about the extent of meltdown in the damaged FDNPP reactors. This information can help inform future management and the decommissioning of the damaged reactors. However, given the complex nature of these newly discovered materials, and the long half-lives and radiotoxicity of their components, we now need to examine their reactivity and fate in the environment to better understand their long-term dose implications and environmental impacts **[3]**. This fundamental information can then be used to inform clean-up efforts in the exclusion zone and guide management plans for contaminated soils collected from within the exclusion zone. This information is also useful in the UK as it can help inform the management of UK nuclear sites impacted by hot particles (e.g the Dounreay and Sellafield nuclear facilities) and provide knowledge and know-how in nuclear accident / incident response.

PhD Aims and Objectives:

This PhD will build on an existing collaboration between The University of Manchester and Kyushu University to deliver new insights into the environmental behaviour of nuclear accident derived radioactive hot particles in the environment. It also builds on experimental and analytical capabilities developed in NERC funded research on radioactive hot particles between The University of Manchester and Diamond

Light Source, which has focussed on synchrotron enabled analytical approaches to examine hot particle alteration mechanisms in soil systems (Figure 1). The ***aims*** of the PhD will be to: **(1)** Isolate representative fuel debris and corium-bearing hot particles from FDNPP exclusion zone soils and prepare them for characterisation in the UK and with project partners in Japan (Figure 2). **(2)** Use the combined UK and Japanese analytical capability to characterise the isolated particles. **(3)** Use the generated data to help assess (the current environmental impact of the particles. **(4)** Synthesize model fuel debris and corium-bearing hot particles at Manchester and conduct alteration experiments in soils / sediments under representative saturated (terrestrial) and vadose zone conditions, to provide a mechanistic understanding of how these materials alter after deposition, and to gauge their environmental stability.

The ***specific objectives*** of the project are to: **(1)** Determine the radionuclide, stable element, and isotopic composition of FDNPP fuel debris and corium-bearing hot particles using mass spectrometric (particularly TOF-SIMS and nano-SIMS), electron microscopy, and radiometric techniques. **(2)** Measure stable and radio-element speciation, identify phases and characterise the structure of single fuel debris and corium-bearing hot particles using synchrotron-based micro- and nano-focus techniques and mass spectrometry, ESEM and TEM approaches. **(3)** Perform experiments with synthesised fuel debris and corium-bearing hot particles in column and lysimeter systems to explore rates and mechanisms of environmental alteration (e.g. radionuclide release, secondary uptake or phase formation) to develop a detailed understanding of hot particle evolution in soils.

Training, Analysis, and the Research Team:

The successful researcher will have access to state-of-the-art experimental and analytical facilities at The University of Manchester (reaction systems, SIMS, electron microscopy, and radiometric techniques) and Diamond Light Source (micro and nano-focus spectroscopies). They will also work with academics at Kyushu University who specialise in transmission electron microscopy. Training will be provided. At Manchester they will work across two vibrant research groups: the Centre for Radiochemistry Research and Research Centre for Radwaste Disposal, which combined has 25+ researchers already focused on environmental radioactivity research.

References. **[1]** Imoto, J et al., (2017). Isotopic signature and nano-texture of cesium-rich micro-particles: Release of uranium and fission products from the FDNPP. Sci. Rep., 7, 5409; **[2]** Ochiai, A et al., (in review, Sci. Adv.). Nuclear fuel fragments released with caesium-rich micro-particles from the FDNPP **[3]** Salbu, B and Lind, OC (2016). Integrated Env. Assess. Man., 12(4), 687-689.

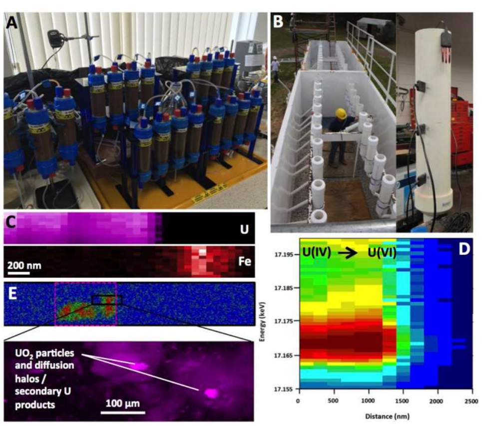

Figure 1 Caption – (A) University of Manchester hot particle column reaction system; (B) Lysimeter rig used for vadose zone alteration experiments; (C) DLS I14 nano-probe XRF maps of U and Fe collected from a FIB'd section of a UO_2 particle (left particle centre, centre particle edge) that had been altered in oxic groundwater; (D) Top down view of XANES stack plot of the same FIB'd particle showing U speciation changing from the particle interior to the particle edge; (E) autoradiography (top) of a resin embedded hot particle thin section, allowing targeted micro-focus spectroscopy at DLS I18 (bottom).

Figure 2 Caption – University of Manchester Researcher searching for reactor fuel debris particles in the FDNPP Nuclear Exclusion “Red” Zone, about 2 km from the FDNPP reactors.