Assessment of the long-term performance of potential wasteforms for plutonium under conditions relevant for geological disposal in the UK

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The required long-term isolation of radioactive wastes from the biosphere can be achieved by deep geological disposal through a multiple barrier system, consisting of a combination of a man-made engineered barrier system (EBS) with a suitable geological barrier, the repository host rock. The first barrier against the release of safety-relevant radionuclides from disposed high-level radioactive wastes (HLW) is the waste matrix (wasteform), which should exhibit sufficiently low radionuclide release rates over time. The understanding of processes that govern the corrosion behaviour of and the consequent radionuclide release from the disposed waste matrices constitutes an integral part of a long-term safety assessment for a geological disposal facility. Plutonium generated in uranium-based fuels during the operation of nuclear reactors due to capture of neutrons is recovered during reprocessing of spent nuclear fuels. The preliminary preferred policy on the long-term management of separated stocks of UK civil plutonium (about 90 t_{HM} as PuO₂), which are currently held in storage as zero value asset, is reuse as MOX fuel, but consideration of disposal options will continue [1]. Notwithstanding future UK government strategies for plutonium disposition, at least a portion of the UK plutonium inventory (i.e. some tonnes) will probably be designated for geological disposal. However, experimental data on the performance of plutonium wasteforms under disposal conditions is rather limited to date and a detailed understanding of relevant processes that govern long-term radionuclide releases on a molecular level is still missing [2].

This paper will describe outcome and conclusions of a review on the long-term durability and performance of potential plutonium wasteforms under conditions relevant for geological disposal in the UK, performed on behalf of the NDA RWMD. Key issues addressed were the durability and chemical reactivity of the wasteforms in aqueous environments and the long-term radionuclide release under disposal conditions. An essential part of this work formed the elicitation of corrosion rate data for the potential wasteforms, based on available experimental data and analogue evidence from other nuclear wasteforms, e.g. HLW-glasses and spent nuclear fuels. Generic candidate wasteform types for plutonium considered in this study comprised nuclear waste glasses (i.e. borosilicate and phosphate glasses), single- and polyphase ceramic wasteforms, cementitious wasteforms, as well as lowspecification (storage) MOX. The current UK disposal programme is in a generic stage, where no preferred disposal concept or type of host rock has yet been selected. Thus various disposal scenarios and a range of possible environmental conditions in the repository near-field were considered, including alkaline conditions potentially arising due to the usage of cementitious materials in the EBS or from a co-located cementitious LILW-repository module. The elicited corrosion rates for the various wasteforms can be used in Post-Closure Safety Assessments to calculate performance and safety indicators such as time-dependent radionuclide fluxes between repository compartments or mean annual individual risks that are related to plutonium disposal.

^[1] Department of Energy & Climate Change (2011) Management of the UK's Plutonium Stocks, London.

^[2] Deissmann et al. (2012) Min. Mag. 76, 2911-2918.