

## STUDY OF RUTHENIUM VOLATILIZATION AND TRAPPING DURING A LOSS OF COOLING ACCIDENT ON HIGH LEVEL LIQUID WASTE (HLLW) STORAGE TANKS IN REPROCESSING PLANTS

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### Abstract

The reprocessing of spent nuclear fuel produces high level liquid waste (HLLW). Due to the decay heat, these concentrated nitric solutions containing fission products are stored in cooled tanks to prevent the solution from boiling, evaporating and drying out, which could lead, in case of loss of cooling, to large releases of radioactive materials into the environment, especially ruthenium volatile species. In the post-Fukushima complementary safety assessments, the loss-of-cooling accident on HLLW storage tanks is one of the accident scenarios identified as a dreaded situation. It is also taken into account in defining the on-site emergency plan in Orano La Hague (France) reprocessing plant. Besides, an extensive literature review performed at IRSN confirms the lack of reliable data on the behaviour of ruthenium in nitric acid solutions. It highlights that research works on this topic can be classified in several categories: ruthenium chemistry in a nitric medium characterized by the formation of nitrosyl ruthenium ion  $\text{RuNO}^{3+}$ ; behaviour of volatile forms of ruthenium in presence of steam, nitric acid and nitrogen oxides (recombination, decomposition, etc.); transfer phenomena of the different gaseous species containing ruthenium. Subsequently, the efficiency and the performance of various systems that can be used to trap and filter ruthenium (gas/liquid absorbers, steel filters, zeolites, etc.), or even prevent its volatilization (recombination, addition of reducing agents *in situ*, etc.) have been investigated by different authors. Previous experimental work performed at IRSN on severe accident scenarios in nuclear facilities allowed to characterize usual filtration devices such as active charcoals or metallic filters, with respect to gaseous  $\text{RuO}_4$ . It showed that these latter do not trap efficiently  $\text{RuO}_4(\text{g})$ .

From these findings, IRSN started a research program aiming at improving the knowledge on this topic. A specific test bench has been developed in order to study the volatilization of a nitric acid solution containing Ru nitrosyl, simulating a real HLLW in terms of acidity and ruthenium concentration, and to investigate the possible inhibition of Ru volatilization by addition of specific reducing compounds. The experimental device is also used to study  $\text{RuO}_4$  trapping by different materials: zeolites, rare earth oxides, etc. First experimental results are presented.

**Keywords:** ruthenium, volatilization, nitric acid, high level liquid waste (HLLW)