STUDY OF THE CHEMICAL CHANGES OF MICROMETER-SIZED PARTICLES OF URANIUM TETRAFLUORIDE IN ENVIRONMENTAL CONDITIONS BY MEANS OF MICRO-RAMAN SPECTROMETRY

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Analyses of particulate material sampled in nuclear facilities provide a lot of relevant information useful for nuclear safeguards about the processes implemented in the facilities, as a small fraction of the collected particles – with sizes typically in the hundreds of nm to the tens of µm range – may come from the nuclear materials handled or stored in the facility. Among the information of interest are the chemical phases of the nuclear materials. Indeed, many different uranium compounds may be encountered in the nuclear industry: several pure uranium oxides and fluorides, uranium-ores and ore concentrates. So the identification of the composition of a uranium compound at the micro-particle's level is of great interest as an indicator of the industrial steps and processes implemented in the facility. Yet micro-Raman spectrometry (MRS) has proven to be a well-suited analytical tool for determination of the chemical composition of µm-sized objects and notably of uranium micro-particles.

Amongst the uranium compounds used or produced in the nuclear industry, uranium tetrafluoride (UF_4) is of great importance as it is an intermediate product in the conversion of U-ore concentrates to UF_6, a key compound in the enrichment U process, and in the production of UO_2 (nuclear fuel). So detecting UF_4 or UF_4 oxidation/degradation products in the vicinity or inside a nuclear facility certainly indicates that a conversion activity has taken place or is currently taking place inside this facility or a related one.

UF_4 is an emerald green compound, non-volatile, insoluble in water and organic solvents, and, as a bulk material, is regarded as a chemically stable compound in air at ambient temperature. However, chemical reactivity of UF_4 micro-particles in air (oxidation kinetics and parameters influencing the oxidation in environmental conditions) is not documented.

The purpose of this work is to determine how long UF_4 in micro-particles can be detected after release in the environment, what the degradation products are and what are the parameters which mostly lead to degradation of UF_4. In this presentation, we describe and give results of an ageing experiment of UF_4 particulate material in various controlled environmental conditions (three different temperatures, air or inert atmosphere, humid or dry atmosphere, with/without exposure to UV light). Particles of pure UF_4 with typical sizes of a few µm were sampled and deposited on graphite disks, located by means of a scanning electron microscope (XL-30, FEI, The Netherlands) and regularly analyzed for three months by MRS (In Via, Renishaw, UK). We studied the influence of the main environmental parameters on the alteration of UF_4, we identified the compounds produced and estimated the degradation kinetics, and so the typical time span during which UF_4 can always be identified in micro-particles by MRS after its release in the environment.