RADIOLYTIC DECOMPOSITION OF ZINC STEARATE IN PRESENCE OF PuO_2 POWDERS

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Zinc stearate is a metal salt of fatty acid used as lubricant in nuclear industry. The lubricant is added for the manufacturing of the oxide powder as fuel pellet. In 4th generation nuclear reactors, fuels will probably contain increasing amount of plutonium with a higher percentage of ^{238}Pu isotope. This latter is an alpha emitter with a quite high specific activity (17.12 $Ci.g^{-1}$). Then, this will enhance the radiolysis phenomena induced by plutonium.

The radiolytic decomposition of zinc stearate was mostly studied through radiolysis gas analysis. To carry these experiments, zinc stearate powder was put in close contact with plutonium oxide according to two configurations :

- Zinc stearate and plutonium oxide powders are pressed separately as pellets, which are piled alternately.
- Zinc stearate and plutonium oxide powders are directly mixed with a weight content of zinc stearate between 1 and 2%.

The first kind of experiments was carried out placing on top of each other several zinc stearate pellets and plutonium dioxide discs to increase the contact surface between the materials and to control it. The second configuration aims to reproduce representative conditions of manufacturing process of nuclear fuel. These two configurations are compared in order to determine as much as possible the impact of radiolysis during pelletizing of nuclear fuel and green pellet storage.

Powders are placed in an airtight cell connected to a micro gas chromatograph in order to determine the nature and the quantity of the gases produced in the cell. Many experiments are made using plutonium oxides with different isotopic vectors, i.e. by varying quantity of ^{238}Pu.

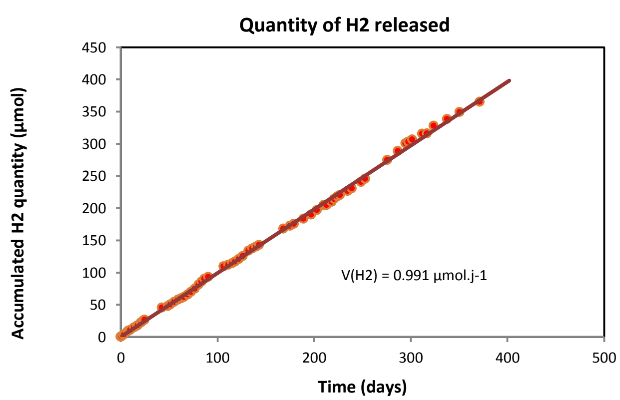

Fig.1: H_2 release kinetic during zinc stearate radiolysis

Five gases (H_2, CO, CO_2, CH_4 and C_2H_6) have been analyzed; the major one being H_2 (Fig 1). Based on the kinetics of gases evolution, the radiolytic yields have been estimated and it appears that they do not depend on the isotopy of plutonium. By contrast, changing configuration shows that the release of carboned species is more important when powders are mixed.

Zinc stearate pellets have also been irradiated externally thanks to Cyclotron of CEMHTI laboratory. Alpha radiations are simulated using helium particles. Different energies of incident particles are employed to vary dose rate. Other particles were used – protons, deutons – to compare impact of radiations with very different linear energy transfer (LET). Gas production was studied in the same way that during first experiments with plutonium oxide. In this way, external and internal (with PuO_2) irradiations can be compared based on radiolytic yields. The major advantage of using external irradiations is that they allow a solid study more accurate than in glove box, and a better understanding of irradiation of zinc stearate, thanks to the non-contamination of the samples.

Based on the results acquired through these experiments, radiolytic decomposition mechanisms of zinc stearate are proposed.