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## Alpha radiolysis induced by plutonium dioxide

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Up to now, plutonium dioxide is the major end product of nuclear fuel reprocessing. Due to the radioactive properties of plutonium, handling and management both of plutonium dioxide and of contaminated wastes involve to take into account the radiolysis phenomena plutonium can induce. For instance, plutonium is known to be a highly hygroscopic compound. Kept under a humid atmosphere, up to 3% wt. water can be sorbed on the surface of plutonium dioxide. The sorbed water then is going to undergo a radiolytic decomposition and this latter will lead to hydrogen evolution. It has been shown through our studies and other reported results that hydrogen formation kinetic depends on the specific surface area of plutonium dioxide, the isotopic composition of plutonium and the relative humidity in the atmosphere. Unfortunately no clear dependences between the rate of hydrogen formation and these parameters can be established. However, it has been shown that in many cases hydrogen amount in the atmosphere reaches a steady-state. Such behaviour is probably linked to surface evolution of plutonium dioxide in presence of moist air and under radiolytic decomposition of water. Studies are still in progress to identify surface changes due to these phenomena. Hydrogen can be produced under radiolysis of some plastic waste contaminated by plutonium dioxide too. Whereas  $\gamma$ -radiolysis of plastic materials is quite well described,  $\alpha$ -radiolysis of such compounds, especially the ones in close contact with actinides, is very poorly known. It can be demonstrated that only a small part of the whole energy emitted by plutonium dioxide is going to be deposited in the material and leads to a radiolytic decomposition with hydrogen and others hazardous gaseous products (CH<sub>4</sub> for instance) formation. Taking into account the amount of energy transferred to the plastic material,  $\alpha$ -radiolytic yield of hydrogen can be calculated and then used for waste repository scale-up. It should be noticed that plutonium dioxide can exhibit specific behaviour towards some radiolytic products. For instance, it has been shown that chlorine arising from PVC radiolytic decomposition is going to be retained mainly on plutonium dioxide surface, whereas without plutonium dioxide chlorine is evolved as gaseous HCl. In the same way,  $\alpha$ -radiolysis induced by plutonium dioxide on several other materials is now studied, such as hydrogen, carbon oxides and short-length alkane production from some lubricating compounds used in fuel pellet manufacturing or from specific waste embedding materials (bitumen, mineral polymers...). As the present trend is still to increase fuel burn-up in one hand, and, in the other hand to manufacture new fuel with increasing amount of plutonium, it appears necessary to look closer at the radiolytic phenomena induced by actinide oxide and especially plutonium dioxide to actualize the safety analysis for handling and management of future nuclear fuels and wastes.