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Interfacial Radiation Chemistry in Nuclear Technology

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Radiation effects on materials constitute a unique and often crucial feature in the nuclear industry. The effects can be either direct, due to radiation energy deposition in the material, or indirect, due to radiation energy deposition in the surrounding media subsequently transferred to the material via chemical reactions. In the majority of nuclear technological systems such as nuclear reactors, fuel reprocessing plants and geological repositories for used nuclear fuel, the surrounding medium exposed to ionizing radiation is water. This is one of the reasons why the radiation chemistry of aqueous solutions has been studied so extensively. However, even though processes occurring in aqueous solutions are extremely important, the most crucial processes from a practical point of view are those occurring at the interface between the aqueous phase and solid materials as these can strongly influence the performance and the integrity of the facility. Of particular concern are corrosion processes induced by oxidative radiolysis products. In most cases the solid material of relevance is metallic or ceramic (primarily UO_2 in nuclear fuel). Hence, the surface of the material is an oxide. It has been reported that the yields (G-values) of the radiolysis products can change significantly in the vicinity of oxide surfaces. The rationale for this phenomenon is still debated. In addition, oxide surfaces have been shown to catalyze several reactions involving the molecular aqueous radiolysis products (H_2 , H_2O_2 and O_2). Consequently, it is of utmost importance to gain fundamental understanding of the radiation driven processes of interfacial systems. In this paper, interfacial radiation chemistry will be discussed in general and the reactions of molecular radiolysis products catalyzed by oxide surfaces in more detail.

The oxide materials that have been studied are UO_2 , Cu_2O and CuO of relevance in geological repositories for spent nuclear fuel according to the KBS-3 concept (copper coated canister), ZrO_2 of relevance in nuclear reactors (fuel cladding) and numerous other oxides that were mainly studied in order to understand the influence of fundamental oxide properties on the catalytic properties of the surface.

The experiments show that the catalytic decomposition of H_2O_2 yielding O_2 and H_2O as final products proceed via the intermediate formation of surface bound hydroxyl radicals. This process is catalyzed by most oxide surfaces. For irradiated systems it is also obvious that radiolytically formed hydroxyl radicals adsorb to oxide surfaces present in the aqueous phase. Specific studies of metallic Copper in water have shown that Copper irradiated in anoxic aqueous solution undergoes significant corrosion and takes up more hydrogen than Copper exposed only to water during the same period. Other experiments reveal that H_2 and O_2 produce H_2O_2 at room temperature in aqueous solution containing oxide surfaces.

In conclusion, oxide surfaces catalyze a number of reactions that are usually not accounted for in homogeneous aqueous radiation chemistry. The molecular radiolysis products yield surface bound radicals of unknown stability. To fully understand present and future systems of relevance in nuclear technology, we must strengthen our knowledge in interfacial radiation chemistry.

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