U.S. DEPARTMENT OF ENERGY
NUCLEAR ENERGY RESEARCH INITIATIVE
ABSTRACT

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Collaborators: **Pacific Northwest National Laboratory, Atomic Energy Canada Ltd.**

Title: **Effects of Water Radiolysis in Water Cooled Nuclear Reactors**

The goal of this project is to develop an experiment-and-calculation based model to describe the effects of radiation on non-standard aqueous systems like those encountered in the Advanced Light Water Reactor (ALWR).

The water that acts as the heat transport medium in the reactor core and primary circuit of nuclear power plants experiences high doses of mixed field radiation. This radiolysis produces radicals, ions and molecular species that are highly reactive at the elevated temperatures corresponding to normal operating conditions. For instance, the oxidizing species have the potential to cause extensive damage to the infrastructure (pipework, etc.) of the primary circuit.

Only very limited information is available concerning the effects of radiation on aqueous systems at elevated temperatures, and on aqueous solution – metal oxide interfaces. This data is almost exclusively for the radiolytic effects of fast electrons, and it does not address the radiation effects of heavy ion, high LET particles. The research program proposed here will use a combination of experiment and modeling to address this need. It will by develop a model for the radiation chemistry of water and aqueous solutions at elevated temperatures under high doses similar to those found in nuclear power plant. The model will consider two aspects of the radiation chemistry,

- the initial diffusion and reaction of the radiation-induced reactants within the primary radiation track and with additives, and
- the bulk, homogeneous and heterogeneous reaction of the radicals and molecular products escaping the track.

The former is important as it determines the initial yields and formation rates of the radiation induced species. This information is central input in the description of the bulk chemistry, which is important for benchmarking experimental data and describing radiation effects in the water of water cooled reactors. The nonhomogeneous component of the model will also phenomenologically describe recombination, trapping and escape to the aqueous phase from nanoscale particles. Processes in heterogeneous suspensions of iron and zirconium oxides will be modeled using stochastic Monte Carlo techniques with experimentally determined mean free path estimates.

A state-of-the-art stochastic simulation methodology will be developed to describe the nonhomogeneous

radiation chemistry of water and aqueous systems at elevated temperatures and pressures similar to those experienced in nuclear reactors. The simulation technique will incorporate the effects of radiation particle type and energy on the physical and chemical processes. Each calculation will involve simulation of the deposition of energy by the radiation particle, and modeling of the subsequent diffusion limited chemistry. The fast chemistry determines the rate of production and the yields of the highly reactive e_{aq}^-, OH, H_2O_2, and the potentially flammable H_2 (and perhaps O_2) in the bulk. Once nonhomogeneous reactions are complete and a homogeneous distribution of reactants is obtained, the code will provide input information for bulk kinetic calculations, based on the FACSIMILE program and the IBM Chemical Kinetic Simulator. Simulations will be performed to model the effects of various additives (H_2 and other OH scavenging solutes) on the initial radiolysis, and the consequences of radiation-induced chemistry at the water/oxide interfaces (with focus on iron oxides). It is expected that the addition of solutes to an aqueous system will alter the nonhomogeneous radiation chemistry changing the yields used in the bulk, homogeneous calculations. At room temperature, this effect is known to occur at lower concentrations for high Linear Energy Transfer (LET) heavy ion radiation particles than for fast electrons.

The model calculations will rely heavily on input information from the experimental components of the research program. There are to be two complementary thrusts to the experimental component of this proposal. These projects will operate in parallel deriving information

- on the yields of the oxidizing OH and H_2O_2 produced in the high-dose radiolysis of aqueous solutions, and
- on radiolytic processes at, and in the presence of, aqueous solution – metal oxide interfaces.

Many models currently utilized to predict the behavior of water in reactors assume given yields of the radiation-induced species. However, these yields are not independent of the irradiation conditions. The yields of hydroxyl (OH) radical and of hydrogen peroxide (H_2O_2) produced in the high dose irradiation of water will be measured for γ-rays/fast electrons at elevated temperatures, and for H^+, He^{2+} and O^{8+} (C^{6+}) ions at room temperature. The effects of additives (specifically H_2) and their concentration will be studied also. Because realistic scenarios involve temperatures above the boiling point of water, the high temperature experiments will necessitate the design and development of an apparatus for irradiation at high temperatures and pressures.

Interfaces are very common in nuclear power plants and in radioactive materials management. Current descriptions of the interaction of radiation with the surface and of the effect of the interface on radiation-induced processes are very limited. Because of the prevalence of iron oxides in the water-cooled reactors (e.g., in the pipes of heat transfer system), experimental (and modeling) studies to address the effects of radiolytic reactions at iron-oxide/water interfaces will be made. These experiments will be limited to fast electron and γ-radiolysis. A temperature range similar to that encountered in reactors will be used. The possibility of radiation-initiated redox dissolution of iron oxide particles will be studied under reductive conditions. The rate of the dissolution will be determined in the presence of various different additives. Mechanisms for the dissolution will be developed, and the database collected will be used in an attempt to simulate the kinetic processes. As direct absorption of the radiation by the oxide may contribute to the dissolution process, the distribution of energy absorption and the resultant distribution of charge carriers between the solid and aqueous sub-phases will be determined. Recombination, trapping and escape to the aqueous phase from nanoscale particles in heterogeneous suspensions of iron and zirconium oxides will be modeled using stochastic Monte Carlo techniques with experimentally determined mean free path estimates.

By combining the knowledge and expertise from the complementary experimental and computational efforts a predictive model for calculating the effects of radiation of aqueous systems under non-standard conditions like those in nuclear power plant will be developed.