NUCLEAR ENERGY RESEARCH INITIATIVE

# Neutron and Beta/Gamma Radiolysis of Supercritical Water

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Commercial nuclear reactors provide a source of heat, used to drive a "heat engine" (turbine) to create electricity. A fundamental principle of thermodynamics is that the higher the temperature at which any heat engine is operated, the greater its efficiency. Consequently, an obvious way to increase the operating efficiency and profitability of future nuclear power plants is to heat the water of the primary cooling loop to higher temperatures. Current pressurized water reactors (PWRs) run at roughly 300°C and 100 atmospheres pressure. Designs under consideration would operate at 450°C and 250 atmospheres, i.e., well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30 percent. A major unanswered question is, *what changes occur in the radiation-induced chemistry in water as the temperature and pressure are raised beyond the critical point, and what does this imply for the limiting corrosion processes in the materials of the primary cooling loop?*

The cooling water of any water-cooled reactor undergoes radiolytic decomposition, induced by gamma, fast-electron, and neutron radiation in the reactor cores. Unless mitigating steps are taken, oxidizing species produced by the coolant radiolysis can promote intergranular stress-corrosion cracking and irradiation-assisted, stress-corrosion cracking of iron- and nickel-based alloys. These will alter corrosion rates of iron- and nickel-based alloys, and of zirconium alloys, in reactors. One commonly used remedial measure to limit corrosion by oxidizing species is to add hydrogen in a sufficient quantity to chemically reduce transient radiolytic primary oxidizing species (OH, $H_2O_2$, $HO_2/O_2^-$), thereby stopping the formation of oxidizing products ($H_2O_2$ and $O_2$). It is still unclear whether this will be effective at the higher temperatures proposed for future reactors. While an earlier NERI project has investigated some of the most important radiation chemistry in supercritical water, there is no information at all on the effect of neutron radiolysis, which is the main source of the troublesome oxidizing species.

The collaboration proposed here is ideally suited to discover most of the fundamental information necessary for a predictive model of radiation-induced chemistry in a supercritical water reactor core. Electron pulse radiolysis coupled with transient absorption spectroscopy is the method of choice for measuring the kinetics of radiation-induced species, as well as product yields for fast electron and gamma radiation. The Argonne Chemistry Division's linac is capable of producing 20 MeV electron pulses of 30 picoseconds duration, and the principal investigators at Argonne have extensive experience in measuring transients on a nanosecond and sub-nanosecond timescale. The University of Wisconsin's Nuclear Reactor Facility is a very convenient source of neutron radiation that can be exploited for radiolysis experiments from room temperature to 500°C. The combined capabilities of these facilities will make it possible to create a quantitative model for water radiolysis in both current PWR systems and supercritical water-cooled plants in the future.

The subject of this proposal touches on several areas of research mentioned in the NERI call for proposals. At its heart, the work is fundamental chemical science, which can be applied to both current and future reactor problems, and other areas of endeavor such as supercritical water oxidation technology. The direct application to nuclear engineering research is the design of reactors with higher performance and efficiency. The work proposed here is a follow-up and extension of research in a previous NERI project (#M9SF99-0276) on the same subject.