Nuclear Energy Research Initiative (NERI) Program
DE-FG03-99SF21923/A00
Technical Progress Report

Narrative:

General:

A project coordination meeting of the NDRL and PNNL co-PIs is planned over the period 6/25-30/1999 at the Radiation Chemistry Gordon Conference at Plymouth State College. Poster presentations of some of the work performed under the auspices of this grant will be presented at this meeting.

Task 1. (Investigator: Simon M. Pimblott, NDRL)

1. Task Status

A literature search of the temperature dependence of radical and molecular product chemistry in the radiolysis of water and aqueous systems continues. Compilation and analysis of the literature on the rate coefficients for the fast reactions of the water radicals is underway. Development and testing of track structure simulation and nonhomogeneous diffusion-kinetic modeling codes is advancing. Simulations of the temperature dependence of the yields of the radicals and molecular products formed by gamma- and fast electron radiolysis are beginning.

2. Issues / Concerns

None

Task 2. (Investigator: Jay A. LaVerne, NDRL)

1. Task Status

The effect of hydrogen on the production of hydrogen peroxide in the radiolysis of water has been examined using deterministic models. It is found that radical chain reactions responsible for the decrease in hydrogen peroxide are relatively slow. They are homogeneous in nature and essentially occur following the dissipation of the initial track structure. However, the track structure controls the escape yields of radicals that take part in the chain reactions.

2. Issues / Concerns

None

Task 3. (Investigator: Dani Meisel, NDRL)

1. Task Status

Pulse radiolysis experiments were conducted on ZrO_2 suspensions at high particle concentrations. The yields of hydrated electron radicals have been measured following a pulse of high-energy electrons in the presence of acceptor molecules. The yield of reduction equivalents increases with the zirconia loading. Under heavy loading of ZrO_2, a significant percentage of the energy is absorbed by the zirconia but that fraction contributes to the radiolysis of water (e.g., gas generation unless prevented). It was discovered that the probability that an electron will escape from the particle into the water is higher than the probability that it will escape recombination if it would originally be produced in the water. These observations imply that energy originally deposited in zirconia crosses the solid/liquid interface and appears in the aqueous phase at higher yields than if it was absorbed in water alone. Experiments are underway to determine the fate of the holes that must be generated in the zirconia in parallel with the generation of electrons.

2. Issues / Concerns

None

DISCLAIMER

Task 4: (Investigator: Thom Orlando, PNNL)

1. Task Status

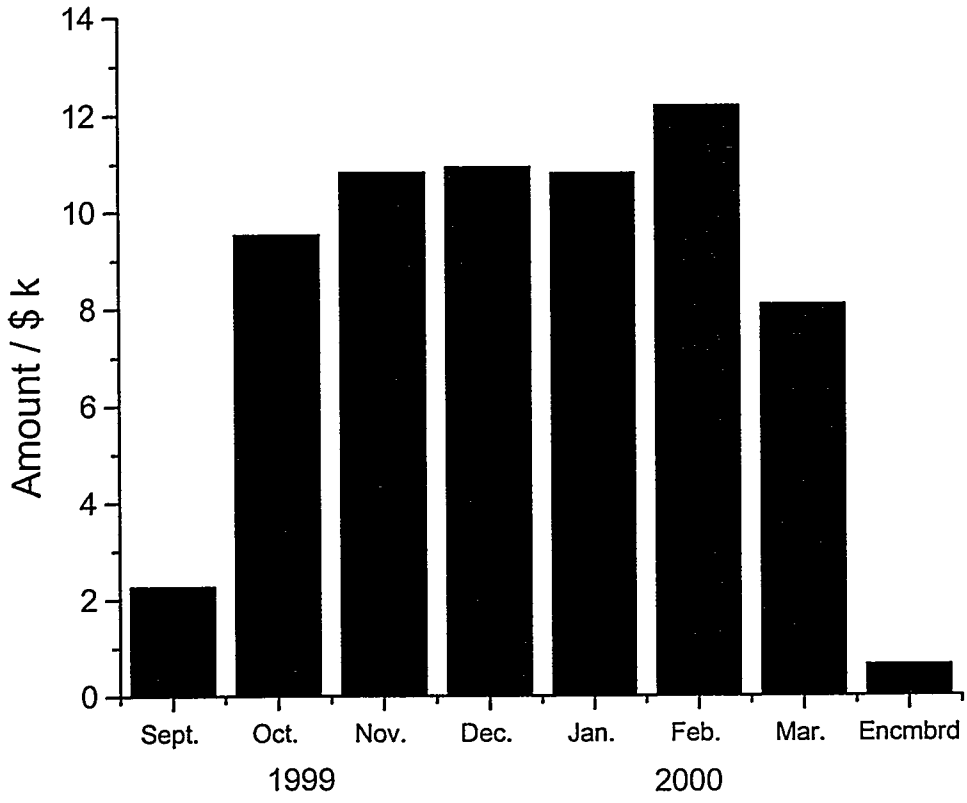
Techniques for growing thin films of ZrO2 which range from a few to several hundred layers thick have been established. Auger spectroscopy of these films demonstrates that they do not contain any contaminants such as hydrogen or carbon. Controlled water adsorption/desorption studies are underway. Single crystals of Fe2O3 have been obtained and studies of the structure and cleanliness of these substrates has begun.

2. Issues / Concerns

None

Cost Performance:

NDRL:



PNNL: Funding provided directly as separate grant to PNNL.

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Status Summary of NERI Tasks - Phases 1-3:

Phase 1:

Milestone/Task Description	Planned Completion Data	Actual Completion Date
Task 1.		
1. Compilation of information on radiation chemistry of water and aqueous solutions at elevated temperatures.	8/14/2000	In progress
2. Algorithms for TRACKKIN code that include the effects of temperature on energy loss and the results of calculations on hydrogen yields at elevated temperatures.	8/14/2000	In progress
Task 2.		
1. Results of hydrogen peroxide yields from gamma and high LET irradiation in the presence of H_2 scavengers at high dose.	8/14/2000	In progress
2. Schematics for the high temperature cell for gamma irradiation.	8/14/2000	In progress
Task 3.		
1. Compilation of information on radiation chemistry of water at interfaces of interest.	8/14/2000	In progress
2. Tested procedures to synthesize (or concentrate dilute suspensions of) iron and zirconium oxide.	8/14/2000	In progress
3. Results from the irradiation of these oxides.	8/14/2000	In progress
Task 4.		
1. Results from the irradiation of doped zirconia films with water overlayers.	8/14/2000	In progress
2. Results from adsorption/desorption following pre-irradiation.	8/14/2000	In progress
3. Results from radiolytic defect production in clean iron oxide.	8/14/2000	In progress

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Status Summary of NERI Tasks - Phases 1-3: cont

Phase 2:

Milestone/Task Description	Planned Completion Data	Actual Completion Date
Task 1. 1. Algorithm and testing of code to simulate high-LET heavy-ion track structure in water. 2. Simulate results of H_2 saturated solutions at ambient temperature.	8/14/2001 8/14/2001	
Task 2. 1. Tested protocol for O_2 measurement from gamma irradiation. 2. Results from the effect of H2 on O2 yields in gamma irradiated solutions at high doses.	8/14/2001 8/14/2001	
Task 3. 1. Schematics of cell for high temperature pulse radiolysis at elevated temperatures. 2. Results from irradiation of heavy loaded suspensions at ambient temperature. 3. Effect of surface potential on escape depth from narrow bandgap oxide materials. 4. Results from feasibility tests of EPR and conductivity techniques to measure the charge escape of electrons and holes from these oxides.	8/14/2001 8/14/2001 8/14/2001 8/14/2001	
Task 4. 1) Electronic band structures of doped zirconia. 2) Results from controlled irradiation of water covered with iron oxide. 3) Results from integrity measurements on the zirconia and iron-oxide/water overlayers.	8/14/2001 8/14/2001 8/14/2001	

Status Summary of NERI Tasks - Phases 1-3: cont

Phase 3:

Milestone/Task Description	Planned Completion Data	Actual Completion Date
Task 1. 1. Results from simulations of irradiation at various temperatures. 2. Comparison between simulations and experimental results of Task 2 and 3. 3. Incorporation of Task 4 into the model.	8/14/2002 8/14/2002 8/14/2002	
Task 2. 1. Results from the effect of H_2 on O_2 yields in high LET irradiated solutions at high dose. 2. Results from the effect of H_2 on O_2 yields in gamma irradiated solutions at elevated temperatures. 3. Measurements of the effect of H_2 on H_2O_2 yields from gamma irradiation.	8/14/2002 8/14/2002 8/14/2002	
Task 3. 1. Results from the irradiation of suspensions at elevated temperatures. 2. Flat band potentials of the relevant oxides at various temperatures. 3. Results from the effects of core-shell structures on yields of water radiolysis.	8/14/2002 8/14/2002 8/14/2002	
Task 4. 1. Hydrogen yield profiles as a function of depth within doped zirconia. 2. Quantitative comparison of low-energy with high energy radiolysis.	8/14/2002 8/14/2002	

NERI Progress Chart

ID	Task Name	Duration	Start date	Finish date	1999		2000				2001				2001		
					Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3
1	Radiation chemistry model development	3 years	8/15/99	8/14/02													
2	High temperature and high LET effects	3 years	8/15/99	8/14/02													
3	Interfacial effects of radiation	3 years	8/15/99	8/14/02													
4	Low energy electrons at surfaces and interfaces	3 years	8/15/99	8/14/02													
	Progress	3 years															

Key	Task	Summary	Rolled up progress
	Progress	Rolled up task	
	Milestone	Rolled up milestone	