



U.S. Department of Energy  
Energy Efficiency and Renewable Energy

# Radiolysis Process for the Regeneration of Sodium Borate to Sodium Borohydride

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## Objectives:

Develop a viable method for regenerating sodium borohydride from sodium borate, to meet DOE's fuel cost target of \$1.50/kg H<sub>2</sub>.

- Demonstrate radiological methods of converting borate to borohydride
- Validate earlier observations, outcomes, and results
- Initiate processes for identifying, qualifying and quantifying conversion mechanisms
- Estimate production capability of process



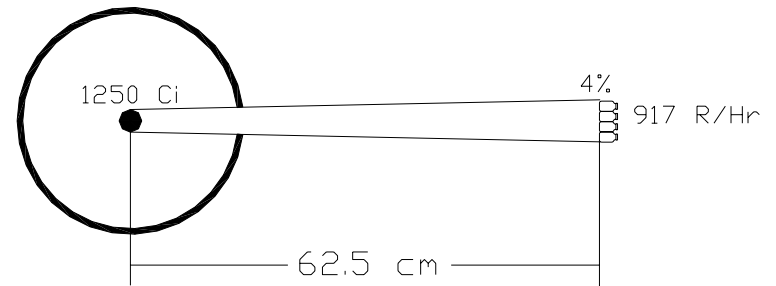
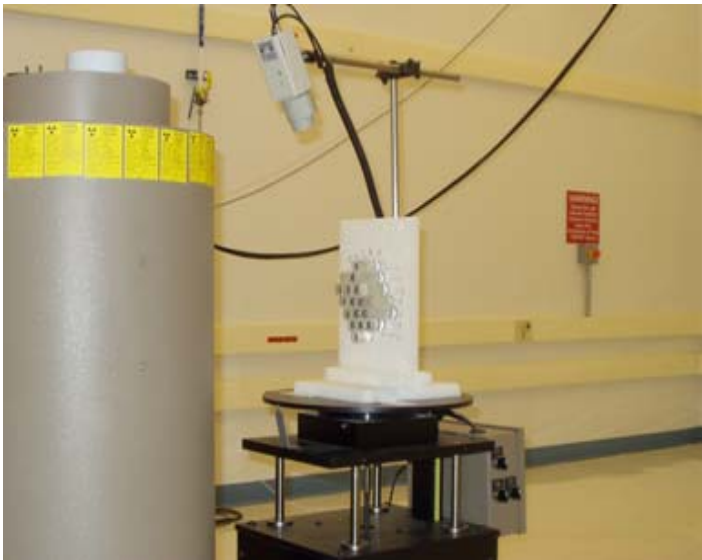
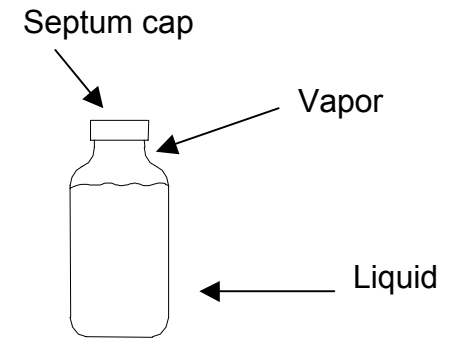
## Targets and Barriers

- DOE Technical Barriers for Chemical Hydride Storage Systems
  - A Cost
  - C Efficiency
  - G Life Cycle and Efficiency Analysis
  - Q Regeneration Processes for Irreversible Systems
  - R By-Product Removal
- DOE Technical Targets for Chemical Hydride Storage Systems for 2010
  - Fuel Cost \$1.50 per gasoline gallon equivalent



## Procedure

- **Radioactive sources used (Cs<sup>137</sup>, Co<sup>60</sup>, Sr<sup>90</sup>, X-ray)**
- **Both Tetraborate and Metaborate tested at controlled concentrations**
  - Tetraborate  $\text{Na}_2\text{B}_4\text{O}_7$
  - Metaborate  $\text{NaBO}_2$
- **DI water sample used for baseline comparison**
- **Vapor space sampling of converted hydrogen**
- **NMR and XRD analysis**
- **1,3,4,7,14, and 20 day samples tested thus far**





## Challenges

- Very little information is known about borate radiochemistry mechanisms and reactions
- Limiting borate solubility's in water
- Limiting detection methods for discriminating converted borohydride (analytical issues)
- Aqueous stabilization issues of borohydride (prevent back reaction to borate)



## Project Safety

- Safety Analysis Processes
  - Independent Hazard Review Process
  - Environmental Checklist
- Hazards Identified
  - Chemical hazards (mitigate by using proper PPE)
  - Hydrogen gas and air mixtures
  - Pressure building in bottles due to the release of Hydrogen and Oxygen (mitigate by controlling sample size)
  - Radiation exposure (technical, facility, and security processes to minimize radiation exposure)



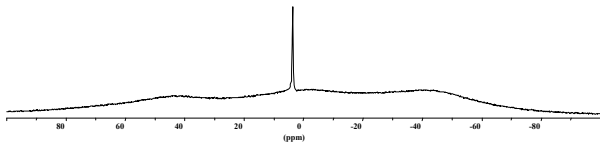
## Technical Accomplishments

- Duplicated FY02 tests in Jan and Mar/Apr 2004 (5 times equivalent hydrogen production quantified)
- Duplicated the ability to generate hydrogen from borate solution (3 times in CY04 obtained the same yield efficiency)
- Developing analytical method/process
- Quantified absorbed radioactive energy
- Have some indication analytically of the possibility of borate to borohydride generation
- Vary parameters to increase ability to detect borohydride (e.g., nmr, x-ray diffraction)
  - Use solid or paste borate material
  - Use more radioactive energy per time
  - Work to provide stabilized aqueous solution for borohydride
  - Dehydrate borate/borohydride solution
- Radiolysis of borate showed 53% conversion efficiency

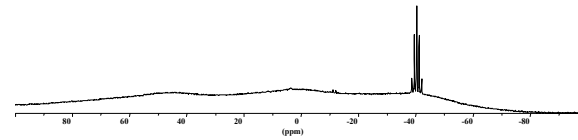




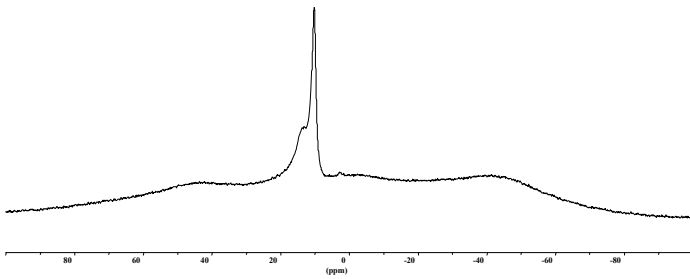
## NMR Spectra of Control and Actual Samples



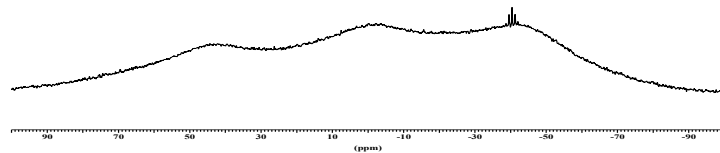
BNMR (sodium borate - control sample)



BNMR (sodium borohydride – control sample)



$^{11}\text{B}$  NMR (sodium borate peak)

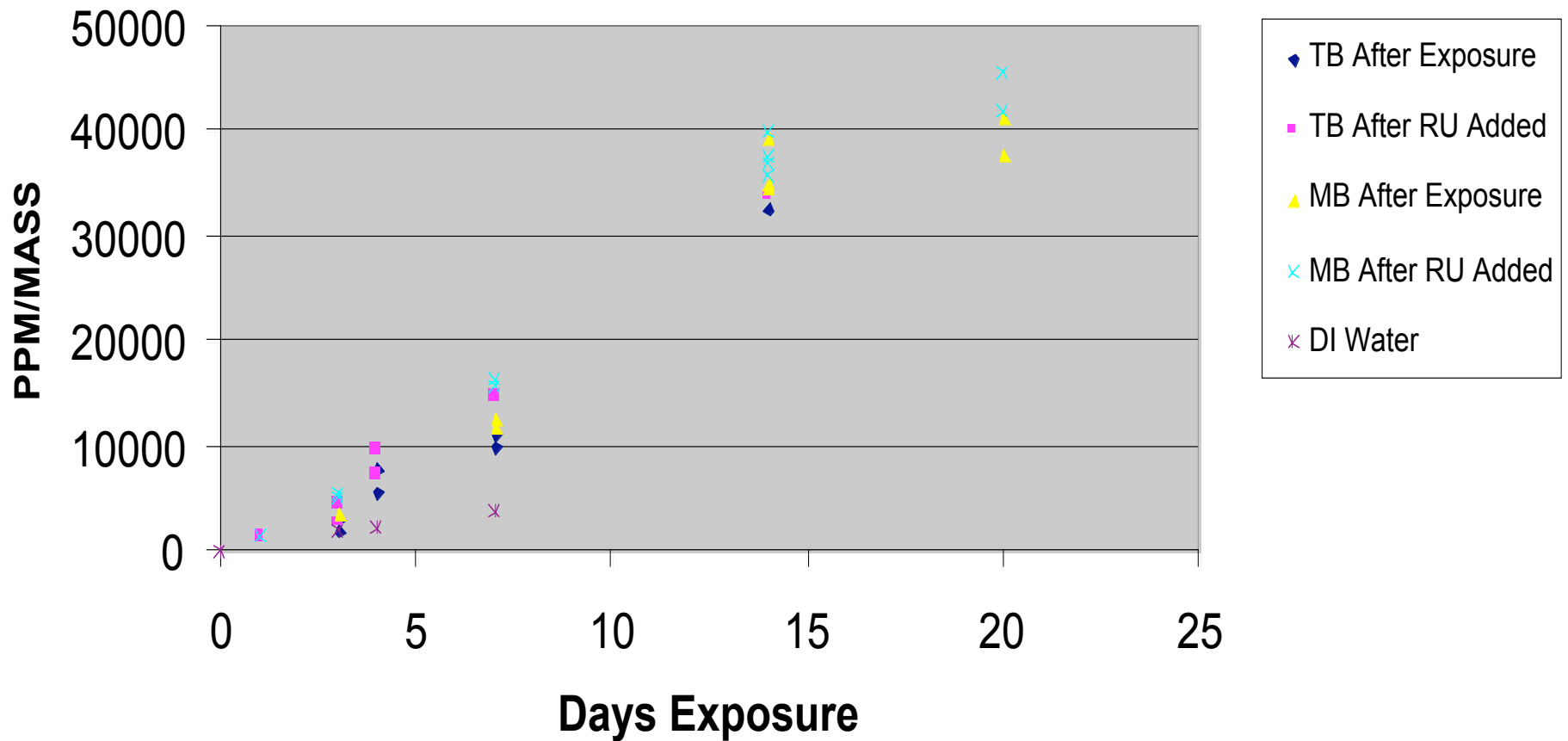


$^{11}\text{B}$  NMR (sodium borohydride peak)





# Hydrogen Production from Borate Conversion





## Radioactive Waste Energy Rough Estimates

### Commercial Reactor Waste

- Commercial power plants produce  $7.52E9$  Ci/yr-reactor waste Approximately 103 reactors in the U.S. that have been running for 25 years.
- Spent waste is being removed from the reactors every 18 months
- Assuming new waste replaces waste with low decay rates and using an average  $2.28E8$  Ci/yr-reactor.
  - Borate to borohydride conversion requires 1440 kJ/mole
  - 325 M kg/yr Hydrogen produced per year
  - At \$1.50/kg Hydrogen production creates \$486 M per year
  - Reclassifies waste into a usable product

### Commercial Reactor Radiation

- Utilizing 10% of the available radiation in a commercial reactor
- 1.0 B kg/yr of Hydrogen produced per reactor



## Interactions and Collaborations

- Millennium Cell
  - Discussed radiation concepts and conversion rates
  - Collaboration on the use of ruthenium
- Idaho State University
  - Radiation measurement to verify amount absorbed
  - Analysis of chemical reactions



## Technical Team's Comments and Resolutions

- Verify identification of borohydride (e.g., nmr, x-ray diffraction)
- Verify 53% conversion
- Determine mass and energy balances
- Investigate methods to eliminate or greatly reduce aqueous back reaction of borohydride
- Continue to provide more data



## Future Work

- Continue investigating methods of improving efficiency and yield
- Identify methods to control back reactions of borohydride
- Quantify the impact of different radiation sources
- Investigate the impact of catalysts
- Qualify conversion mechanism
- Applicability of this process to other chemistries



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