



U.S. DEPARTMENT OF
ENERGY

Nuclear Energy

Fuel Cycle Research and Development

**Effects of Extended In-Process Aging of
Silver-Exchanged Mordenite on Iodine
Capture Performance**

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Neutron Activation Analysis
– David Glasgow (ORNL)

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- **Oak Ridge National Laboratory is conducting research on the off-gas capture systems required in advanced nuclear fuel reprocessing facilities. These facilities would be required to have integrated off-gas treatment systems designed to capture and sequester volatile and semi-volatile radionuclides (H-3, C-14, Kr-85, and I-129).**
- **Treatment pathways**
 - Head end (shear and dissolver)
 - Vessel off-gas (in-leakage to process equipment)
 - Ventilation off-gas (confinement to the process cell)

Iodine-129 Regulatory Requirements

■ EPA 40 CFR 190

- “The total quantity of radioactive materials entering the general environment from the entire uranium fuel cycle, per gigawatt-year of electrical energy produced by the fuel cycle, contains less than 50,000 curies of krypton-85, **5 millicuries of iodine-129**, and 0.5 millicuries combined of plutonium-239 and other alpha-emitting transuranic radionuclides with half-lives greater than one year.”

■ EPA 40 CFR 61.92

- Effective dose equivalent to any member of the public less than 10 millirem per year

■ EPA 10 CFR 20

- Limits specific activity of I¹²⁹ to
 - 4.0×10^{-11} curies per cubic meter in air at the site boundary
 - 2.0×10^{-7} curies per cubic meter in water

Motivation for Study

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- **Knowledge gap - what is the long term behavior of the sorbent materials, silver-exchanged mordenite (AgZ) and reduced silver-exchanged mordenite (Ag⁰Z), under extended exposure to process conditions?**
 - Temperature of Air
 - Composition of Air Stream
 - *Moisture*
 - *Chemistry*



- **Capacity Studies**
- **Retention Studies**

Capacity Studies - Experimental Setup



Thermogravimetric Analyzer (TGA)



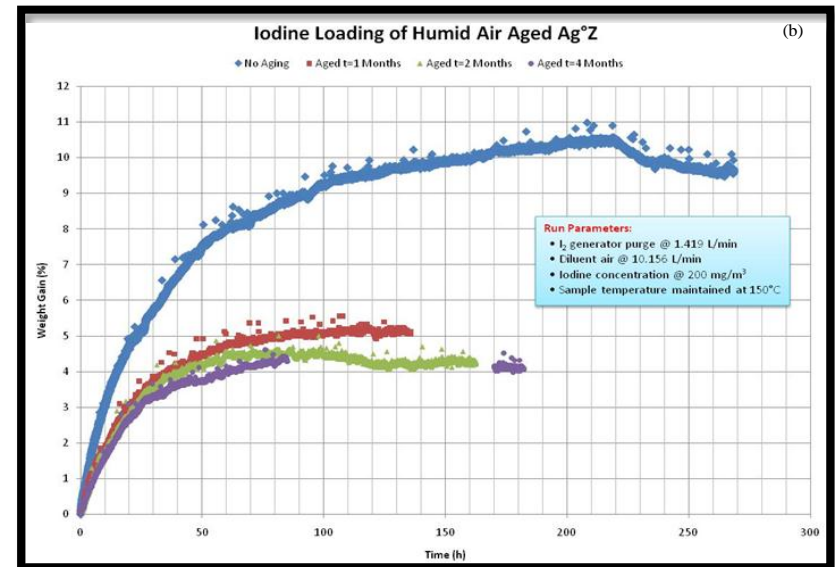
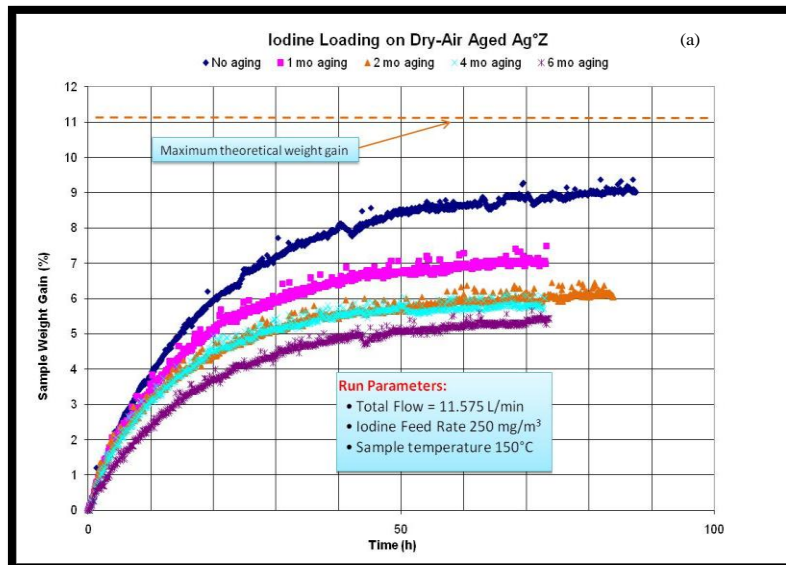
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■ Dry air aging studies

- Iodine capture capacity down after 1 month
- 6 months aging results in 40% loss in capacity

■ Humid air aging studies

- 40% loss in capacity after 1 month
- Additional minor losses after first month



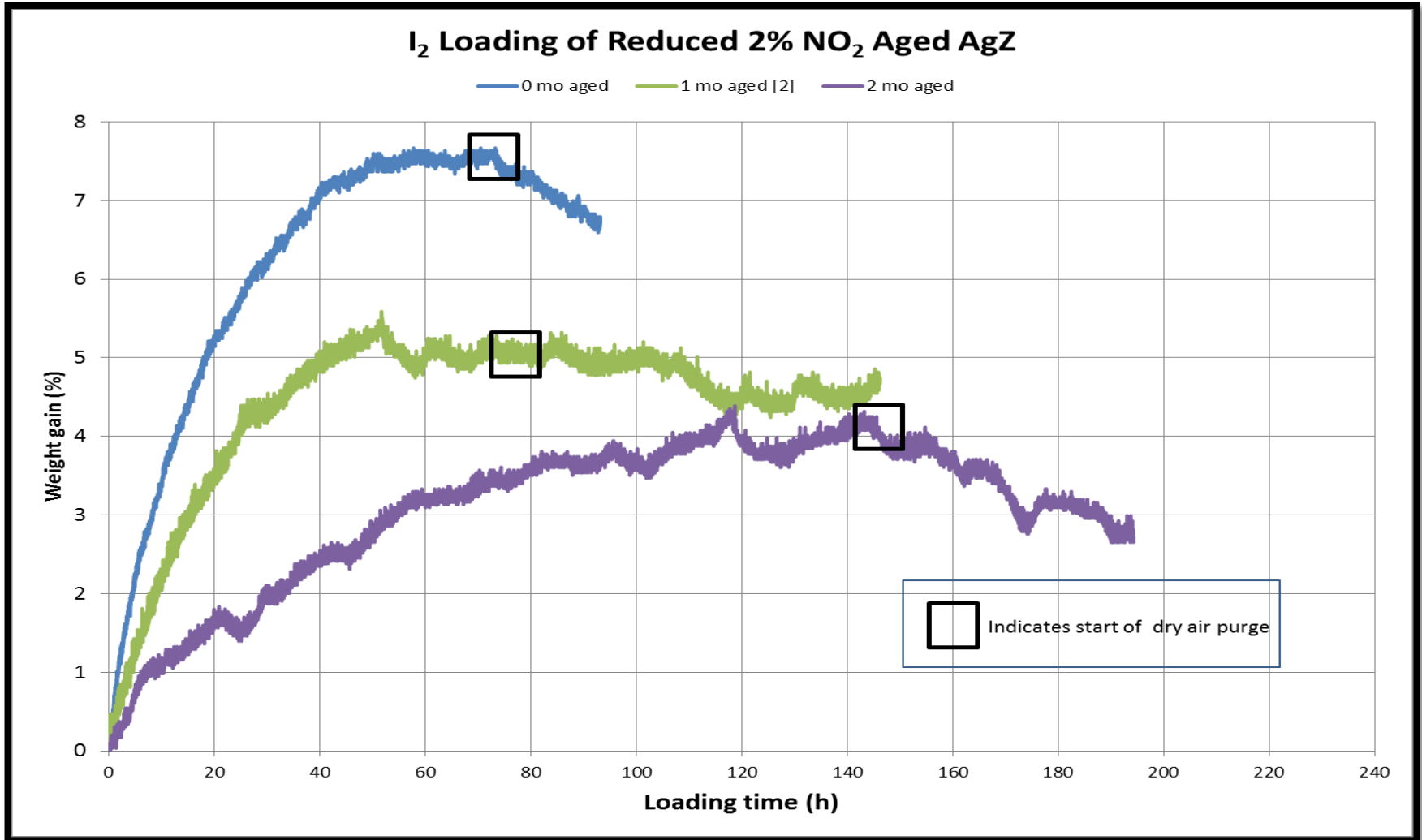


- **Reduced and unreduced material evaluated**
 - Reductions at 270 °C, 4% Hydrogen/Balance Nitrogen
- **Sample into stainless steel tubing holders**
 - 2% NO₂, balance zero air
- **TGA study parameters**
 - 10 m/min
 - 150 °C
 - 50 ppm I₂





Capacity Study Cases – Reduced Silver Zeolite, Ag⁰Z





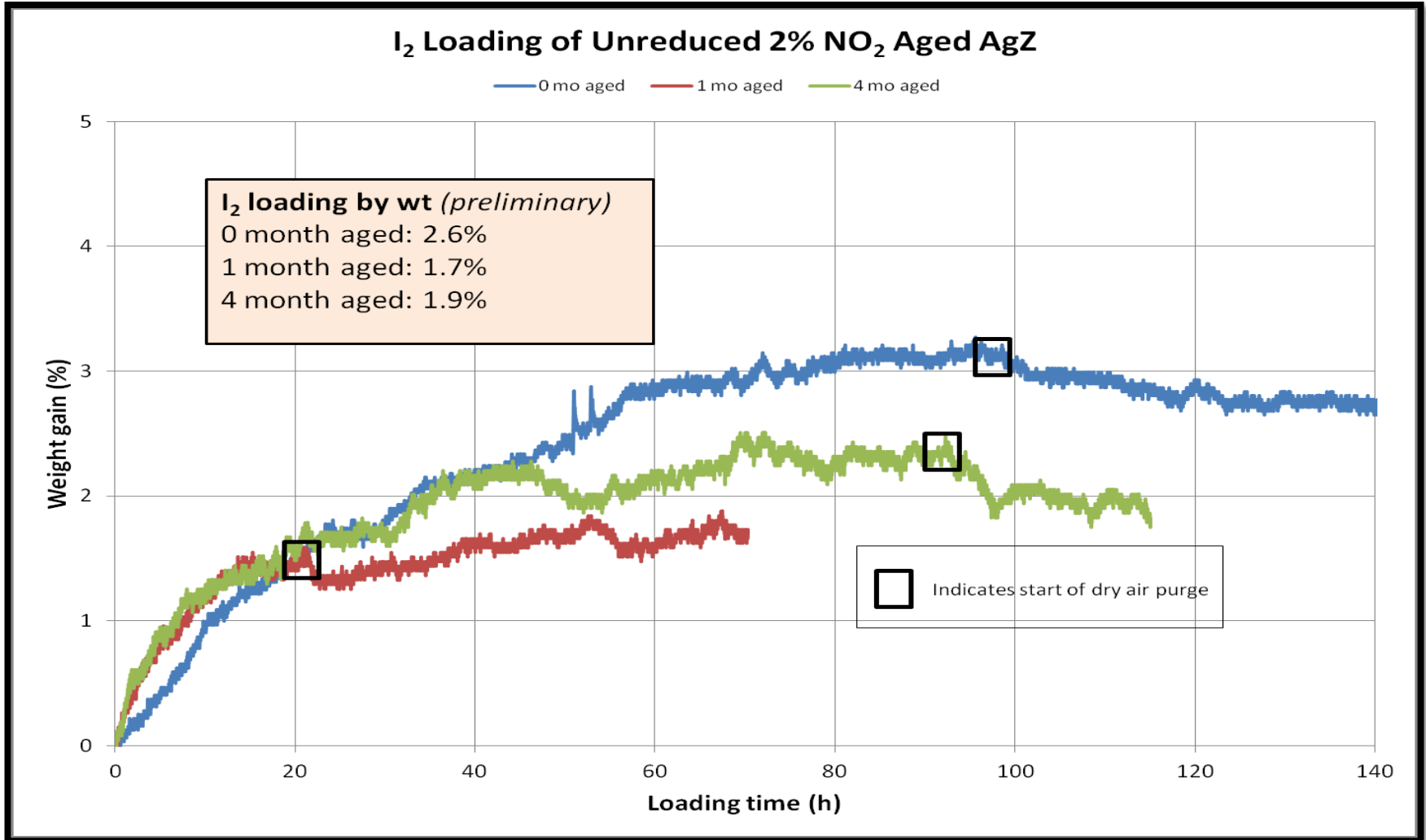
Capacity Study Cases – Reduced Silver Zeolite, Ag⁰Z

**Table I. Iodine Capacity of Ag⁰Z over extended exposure to NO₂
Neutron Activation Analysis Results**

Aging Time (months)	I₂ Loading (wt%)	Uncertainty (wt%)
0	7.19	0.26
1	4.89	0.16
2	2.43	0.12
3	4.78	0.16
4	5.12	0.15



Capacity Study Cases – Unreduced Silver Zeolite, AgZ



Capacity Study Conclusions

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- **NO₂ reduces Ag^oZ iodine loading capacity**
- **Majority of capacity loss in AgZ occurs within first month exposure; iodine capacity loss not due to oxidation of Ag**



- **Materials loaded in a deep bed with iodine first**
- **Sample into stainless steel tubing holders**
 - 2% NO₂, balance zero air
- **Aged in oven for 1, 2, 4 months**
- **Sample tubes purged with 1 l/min air for 1 hour to sweep NO₂, I₂ removed from sample**
- **Re-evaluated for iodine content at end of aging periods**





**Table II. Retention Studies
Neutron Activation Analysis**

Aging Time (months)	I₂ (wt.%)	Uncertainty (wt.%)
0	7.15	0.100
1	8.90	0.014
2	7.71	0.017
4a	7.78	0.014
4b	7.44	0.012

■ Retention

- No significant loss of adsorbed iodine over four month period
 - *Outlier sample resubmitted for neutron activation at HFIR*



- **These studies were designed to fill a gap in our understanding of the long term behavior of silver mordenite under extended exposure to process conditions**
- **Previously reported studies have examined dry air and moist air exposure**
- **This investigation explored the effects of NO₂ on iodine sorbent materials for reprocessing**
 - Effects on Capacity on AgZ, Ag°Z
 - Effects on Retention
- **Results**
 - Capacity – Main loss in capacity after one month
 - Retention – No significant loss of adsorbed iodine
- **Future studies to include combined effects of other gases**

Thank you for your attention