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SAND2014-2959P

The Fuel Cycle Research & Development

Iodine Capture and Storage

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Sigma Off-Gas Team April 30, 2014

Funding Provided by DOE/NE-FCRD- SWG

CH_3-I Deep Bed Sorption Studies (H_2O, NO, NO_2, CH_3-I): Iodine adsorbs as AgI after *Catalytic Cleaving* of CH_3-I on MOR – Milestone M4FT-14SN0312041

(T.M.Nenoff (SNL), N.Soelberg (INL), K.W.Chapman (ANL))

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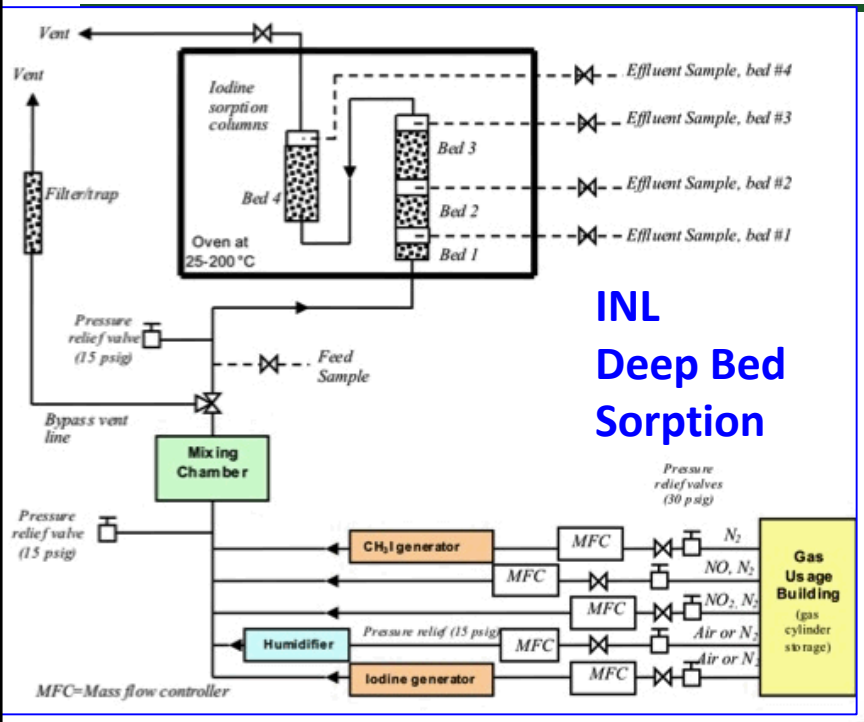
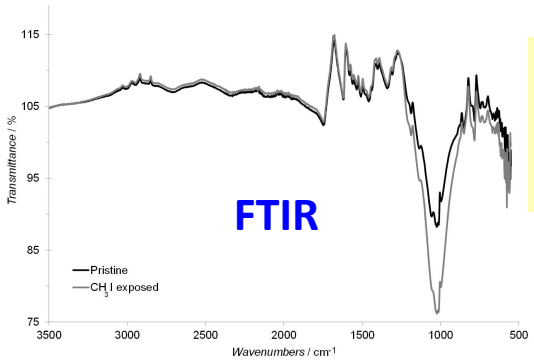
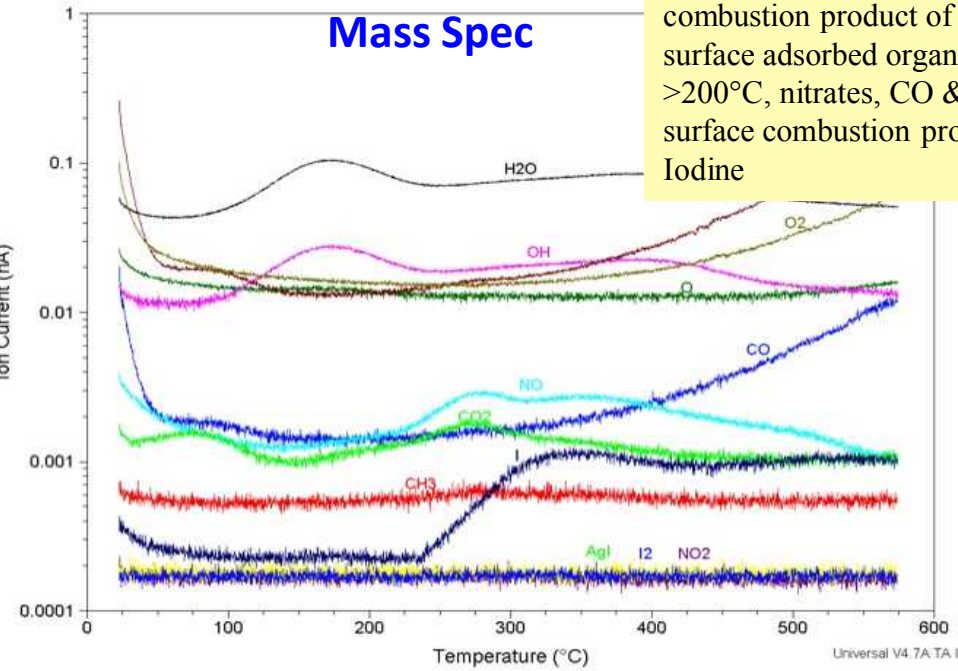


INL Deep Bed Sorption



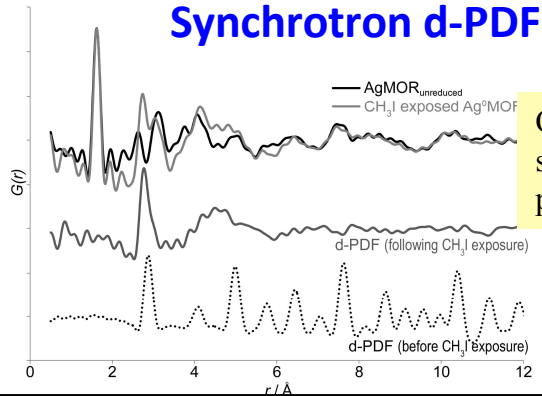
FTIR

No change
Pre and post
CH_3-I interaction,
No organic present



Mass Spec

< 200°C, surface waters and CO_2 removal (CO_2 combustion product of surface adsorbed organics)
>200°C, nitrates, CO & CO_2 surface combustion products, Iodine



Synchrotron d-PDF

Only AgI seen in MOR pores

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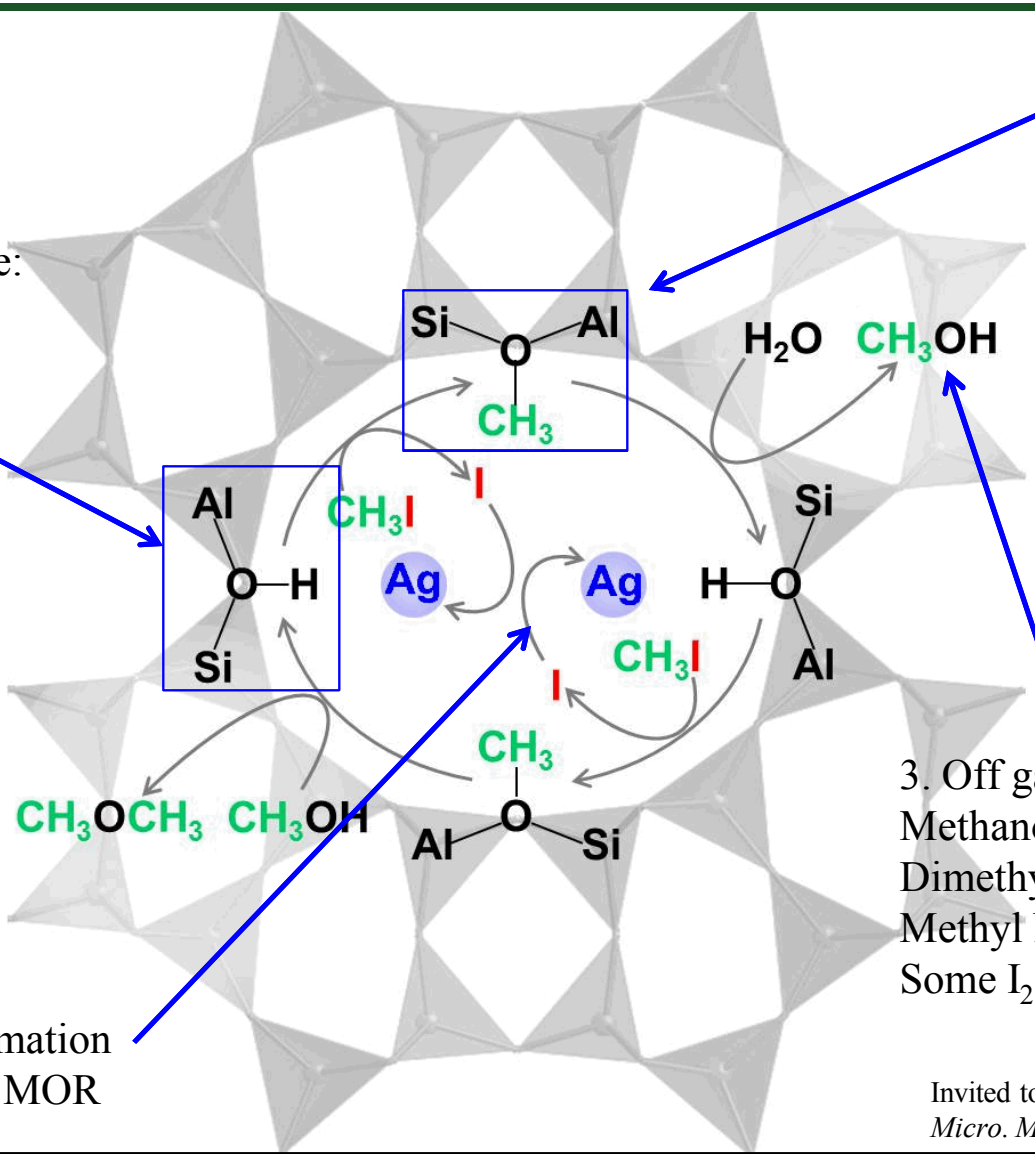
Mechanism Determined for Iodine Adsorption from INL Stream:
CH_3-I Cleaving and Iodine Sorption Over Ag-MOR

1. Original Zeolite Acid Site: H^+ charge-balances framework when Ag° formed

2. *SMS*: Surface Methoxy Species formation
Y. Jiang, et.al, *J. Am. Chem. Soc.*, 128 (2006) 11679.



3. Off gas, downstream:
Methanol
Dimethyl Ether
Methyl Nitrates
Some I_2 (noncaptured by Ag)

4. AgI formation in pore of MOR

Invited to Special Issue: Nenoff, et. al., *Micro. Meso. Materials*, in press, 2014.

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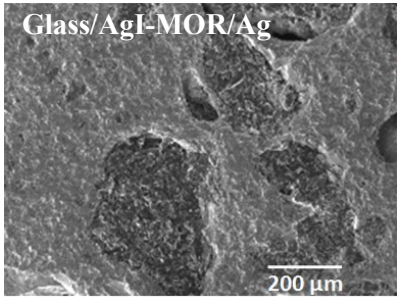
Determine Minimum Ag flake Addition to Glass Composite Material (GCM) for AgI-MOR – Milestone M3FT-14SN0312042

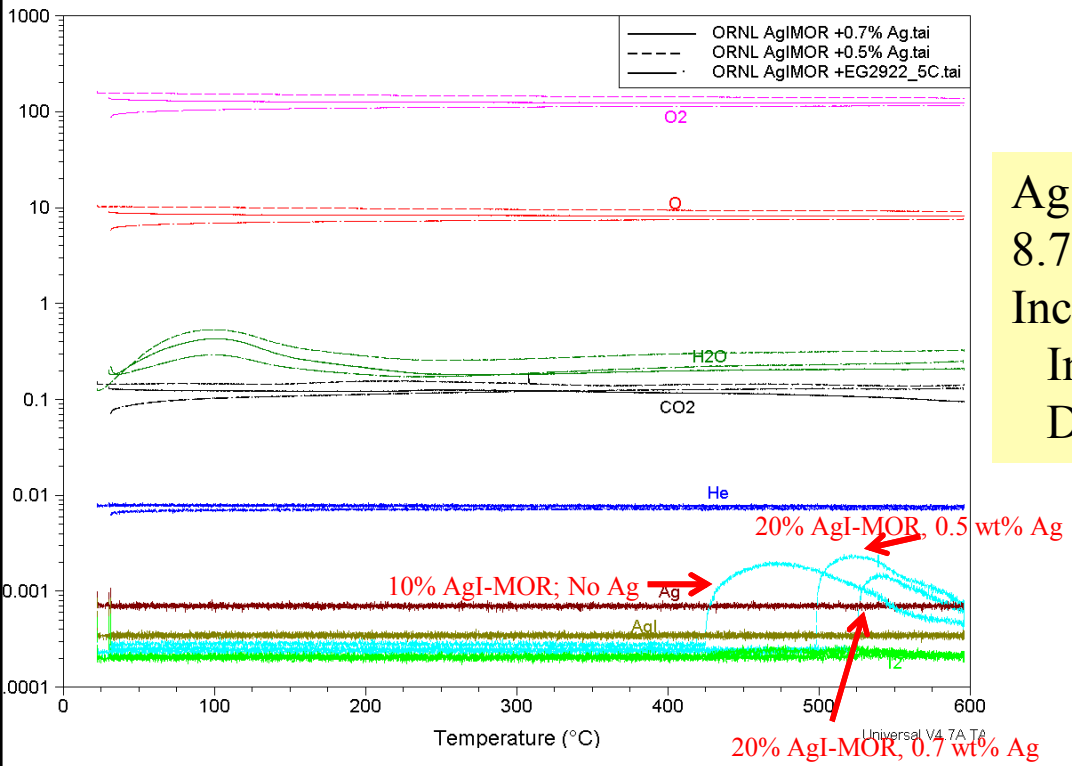
(T.M. Nenoff, T.Garino, M.A.Rodriguez, SNL)

Original Prep: AgI-GCM *
AgI-MOR ground in mortar and pestle, then sieved to <150 µm.
Mixed at 20wt% with 80 wt% glass powder and **additional 5wt% Ag flake (to capture any escaping Iodine during GCM sintering)**.
Uniaxially dry pressed at 70 Mpa in steel die.
Heated at 2°C/min in air to 550°C for 1hr to sinter.

*J. Amer. Ceram. Soc. **2011**,94(8), 2412-2419; US Patent:



Complete Physical Encapsulation of AgI by Glass in composite



Ag flake Addition Studies to 8.7 wt% Iodine Loaded AgI-MOR (ORNL):
Increase from Zero (to < 5 wt%) indicates:
- Increased temperature of Iodine release,
- Decreased amount of Iodine release.

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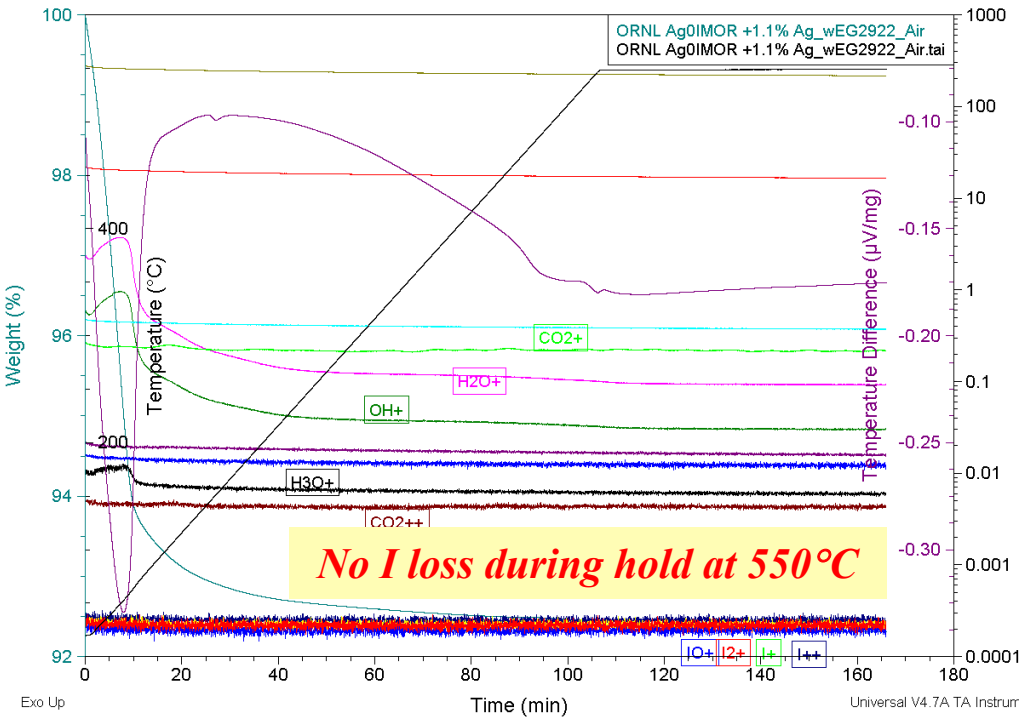
AgI-MOR GCM Optimized Per extra Ag Flake Addition:
- Sintering in Air: 1.1 wt% Ag Flake Addition
- Sintering in Inert Atmosphere (eg. He): *NO Ag Flake*

Sintering in Air: 5°C/min to 550°C for 1 hr
20 wt% ORNL AgI-MOR + 80 wt% glass + 1.1 wt% Ag Flake



Sintering in Inert:
20 wt% ORNL AgI-MOR +
80 wt% glass; NO Ag

Preliminary Results
GCM sintered in *inert atmosphere*
1) No need for Ag flake
2) No oxidation of Ag in AgI-MOR
3) Transport of AgI from bulk into MOR pores
(added physical retention of Iodine)

On-Going GCM Optimization Research:
- Durability studies on GCM sintered in inert with no added Ag
- Optimize maximum loadings (AgI-MOR) possible for durable waste forms
- Study the effect of particle size of iodine loaded AgI-MOR on final GCM waste form (eg., phase purity, durability)