Supporting Information

Effects of Zeolitic Parameters and Irradiation on the Retention Properties of Silver Zeolites Exposed to Molecular Iodine

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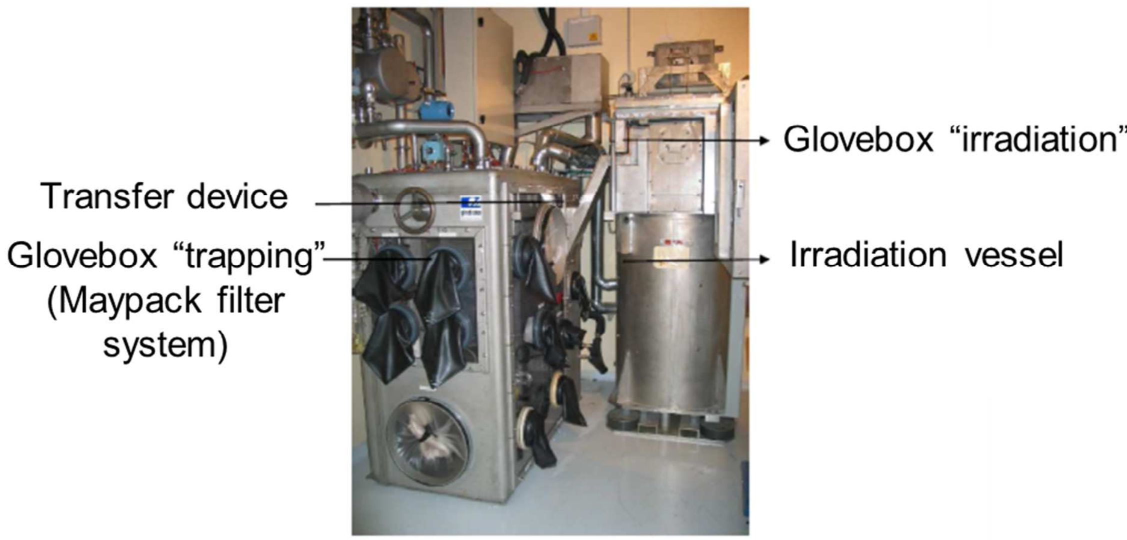
Figure S1: Photo of the EPICUR facility



The EPICUR facility consists of an irradiator with six ^{60}Co sources (T½ = 5.27 years, 893 TBq in September 2012), an irradiation vessel and a loop, allowing on-line measurements (*W.S. Megaw and F.G. May, The behavior of Iodine release in reactor Containers, J. Nucl. Energy, Parts. A & B; Vol: 16; issue 427, 1962*). The loop contains a sweeping gas and direct γ counting in a May-pack device. The dose rate at the filter location is about 2.2 kGy/h in order to mimic the effect of radiations delivered by fission products in the reactor containment during the accident. Fig. 3 gives the general scheme of the facility. The zeolite loaded with I_2 is placed in the irradiation vessel (Fig. S1 in ESI). This temperature-controlled irradiation vessel is connected through stainless steel pipes to the May-pack device (iodine filtration system) in such a way that the volatile species produced in the irradiation vessel are transferred to the May-pack device (gas flow path indicated by red arrows in Fig. 3). Prior to entering the May-pack, the gas flow from the irradiation vessel is diluted (green arrows) in order to provide at the May-pack level a fluid velocity and a low humidity ratio compatible with a good efficiency of the May-pack filters. Each gas flow (to the irradiation vessel and dilution) is regulated independently. The gas flow enters the May-pack by the upper side of each filter stage, so that the activity is deposited close to the NaI counters. The May-pack device is connected to a condenser and to a condensed steam vessel in order to collect water which is re-injected into the irradiation vessel at regular intervals if necessary. The irradiation vessel, the condenser, the condensed steam vessel as well as the piping are made of electro-polished stainless steel in order to minimize iodine deposits. The irradiation vessel, the May-pack device, as well as the gamma counters are placed in a glove box.

In order to quantitative account for the irradiation efficiency, zeolite integrated dose mapping was performed with different dosimetric films in the test configuration.

Table S1: Experimental matrix for irradiation tests.

Parameter	ZEO1	ZEO2
Pre-irradiation of the zeolite before I_2 adsorption	No	2.2 MGy
Temperature (°C)	120	
Pressure (bar)	3.5	
Gas composition	Moist air (R.H of 60%)	
Total flow rate ($g.h^{-1}$) Velocity ($cm.s^{-1}$)	20 0.4	
Irradiation duration (h)	30	
Average dose rate ($kGy.h^{-1}$) Z1 Z2 Z3	 1.6 to 2.0 2.0 to 2.6 2.6 to 4.3	
Iodine loading (mg/g of zeolite) Z1 Z2 Z3	 8.2 3.8 0.5	 9.3 3.1 0.4

Figure S2: Effect of preparation method and Si/Al ratio (A1&A2) and of the irradiation (A3&A4) on structural properties and silver speciation within Ag/Y zeolites.

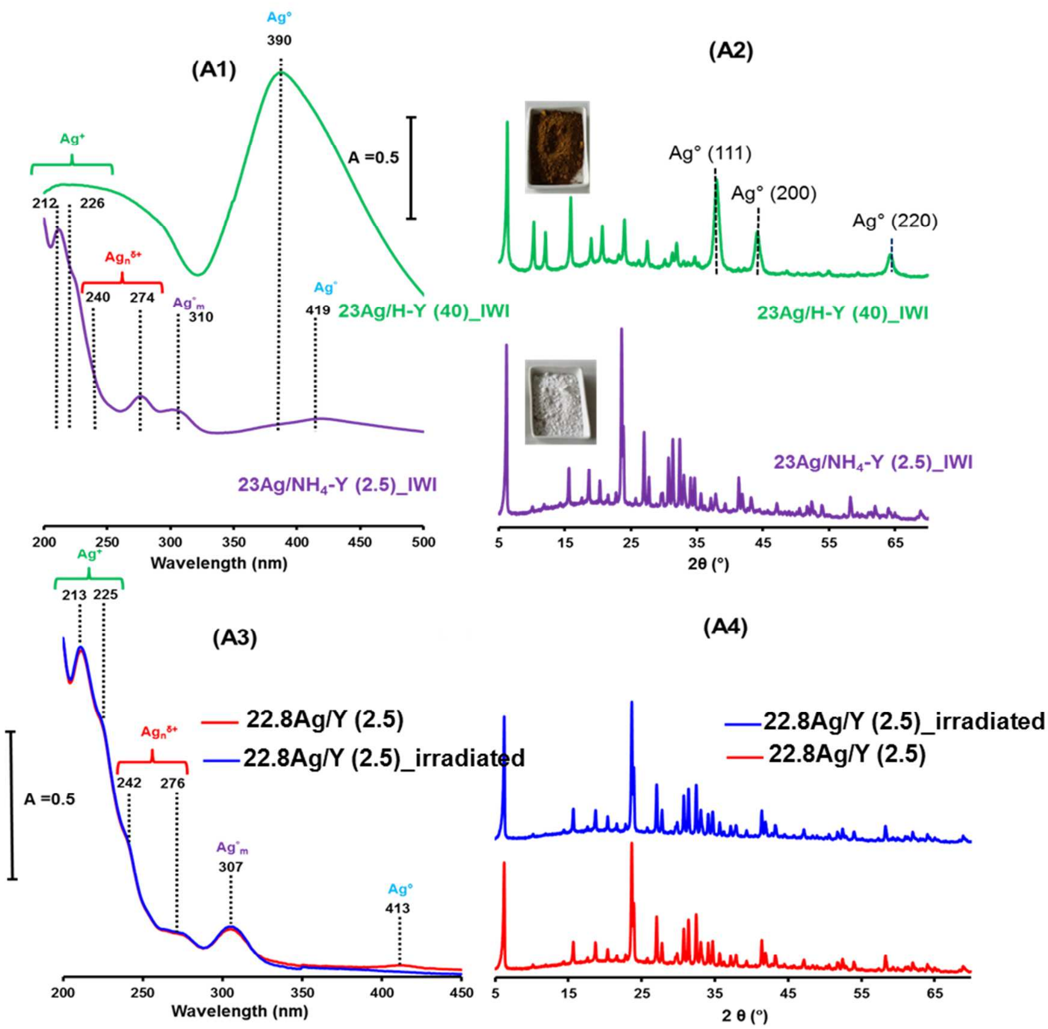
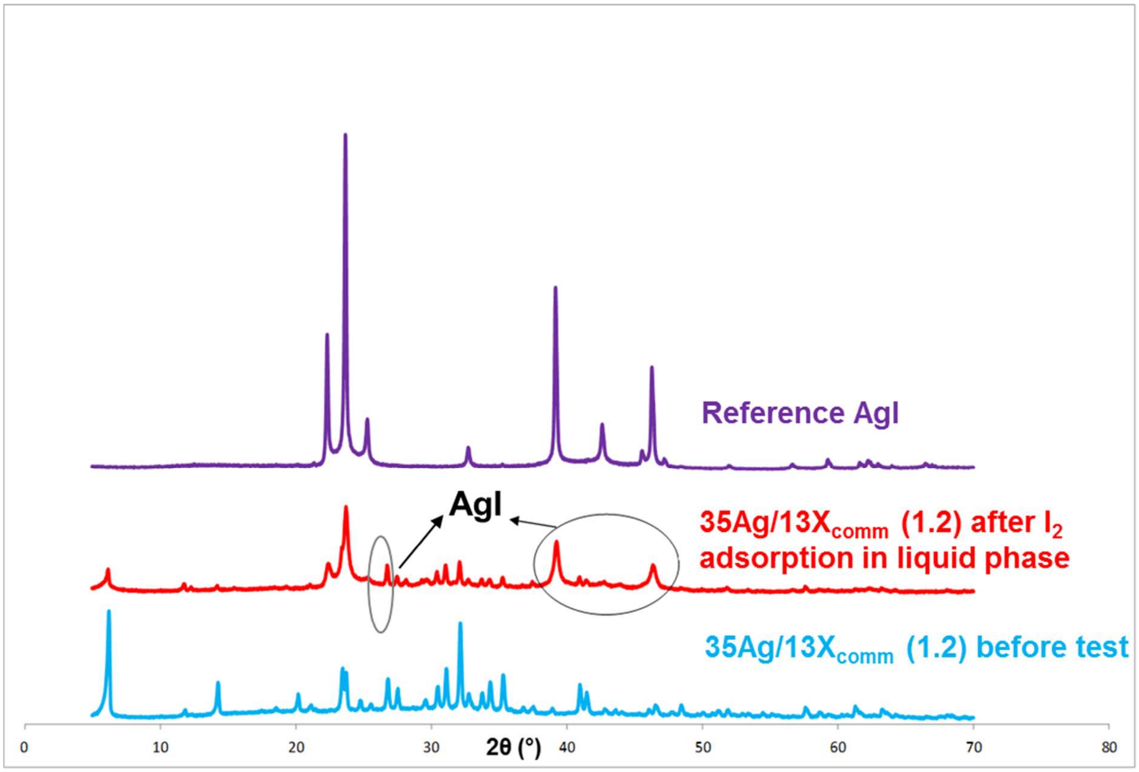


Figure S3: Experimental illustration by XRD of AgI formation after exposure to I_2 in liquid phase for the commercial sorbent ($35Ag/13X_{comm}$ (1.2)).



S4: (A) Description of the adsorption isotherms models

1. *Langmuir model*

$$\frac{Q_e}{Q_{max}} = \frac{b \times C_e}{1 + b \times C_e}$$

Where

- Q_e : Adsorption capacity at the equilibrium (mg/g) ;
- Q_{max}: Adsorption capacity at the saturation of the adsorbent (mg/g);
- Ce: Iodine concentration at the equilibrium (mg/L) ;
- b: Langmuir constant (L/mg).

2. *Dissociatif Langmuir model*

$$\frac{Q_e}{Q_{max}} = \frac{\sqrt{b \times C_e}}{1 + \sqrt{b \times C_e}}$$

3. *Temkin model*

$$\frac{Q_e}{Q_{max}} = \frac{RT}{\Delta Q} Ln\,(K_0 C_e)$$

Where

- R : ideal gas constant (8.314 $J.K^{-1}.mol^{-1}$) ;
- T : Temperature (K) ;
- ΔQ : Adsorption energy variation ($J.mol^{-1}$) ;
- K_0 : Adsorption constant (L/mg).

S4: (B) Summary on I_2 adsorption isotherm modeling (T=25°C, 35Ag/13X$_{comm}$ (1.2)).

Isotherm model	R^2	Parameters of modeling
Langmuir	0.9947	Q_{max} = 280 mg/g b= 0.033 L/mg
Dissociatif Langmuir	0.9953	Q_{max} = 270 mg/g b= 1.187 L/mg
Temkin	0.9757	Q_{max} = 290 mg/g ΔQ = 38 kJ.mol^{-1} K_0 = 1288 L/mg

Figure S5: Characterization of 4.2Ag/FER (10.4) zeolite before and after I_2 adsorption in liquid phase by DR-UV-Vis (A) and XRD (B).

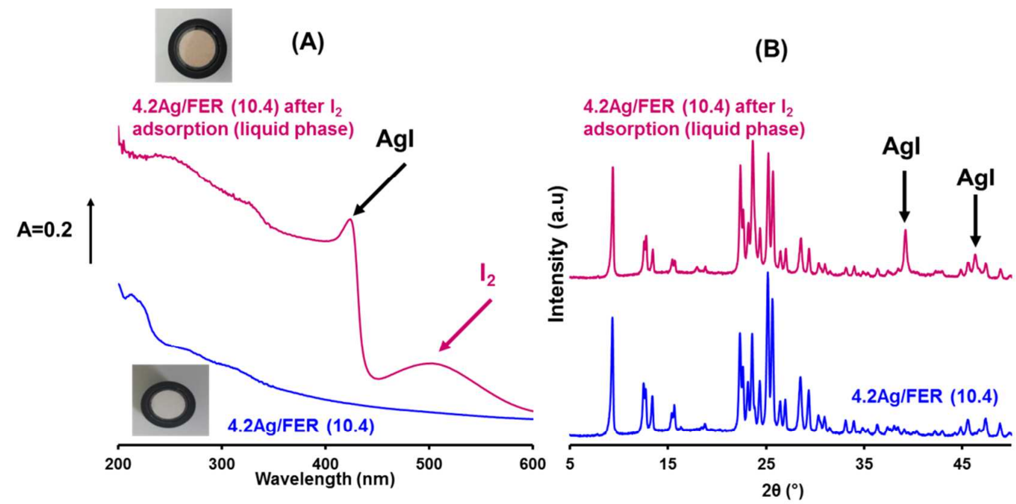


Figure S6: XRD (A) and DR-UV-Vis (B) characterization of 22.8Ag/Y (2.5) and 23Ag/H-Y (40)_IWI after I_2 adsorption in gaseous phase ($[I_2]_0$=1250 ppm, T=100°C).