

# Hierarchical materials to remove and confine radioactive Cs

Clément Cabaud<sup>a</sup>, Yves Barré<sup>a</sup>, Laurent De Windt<sup>b</sup>, Simerjeet Gill<sup>c</sup>, Micheal Moloney<sup>a,d</sup>, Nicolas Massoni<sup>d</sup>, Agnès Grandjean<sup>\*a</sup>

<sup>a</sup> CEA, DEN, DE2D, SEAD, Laboratory of Supercritical and Decontamination Processes, Univ. Montpellier, F-30207 Bagnols-sur-Cèze.

<sup>b</sup> MINES Paris Tech, PSL Research University, Centre de Géosciences, F-77300 Fontainebleau.

<sup>c</sup> BNL

<sup>d</sup> CEA, DEN, DE2D, SEVT, Research Laboratory for the development of conditioning matrices, Univ Montpellier, F-30207 Bagnols-sur-Cèze.

## Supplementary material

Total concentration (mg.L <sup>-1</sup> )			
Ca <sup>2+</sup>	80	HCO <sub>3</sub> <sup>-</sup>	360
Mg <sup>2+</sup>	26	SO <sub>4</sub> <sup>2-</sup>	12,6
Na <sup>+</sup>	6,5	Cl <sup>-</sup>	6,8
K <sup>+</sup>	1	NO <sub>3</sub> <sup>-</sup>	3,7

Table S1: Composition of the commercially available mineral water used for Cs sorption experiments.

	Content in g.kg <sup>-1</sup> as measured by ICP					Specific surface in m <sup>2</sup> .g <sup>-1</sup>
	Cs	Cu	Fe	K	Si	
Cs :Cu-HCF@silica before annealing	13.6±1.4	7.5±0.8	4.8±0.5	11.2±1.1	431±43	140.4
Cs :Cu-HCF@silica after 1000°C annealing in air	14.5±1.5	7.5±0.8	4.8±0.5	11.0±1.1	427±42	0.75

Table S2: Composition and specific surface of samples before and after thermal treatment at 1000°C in air.

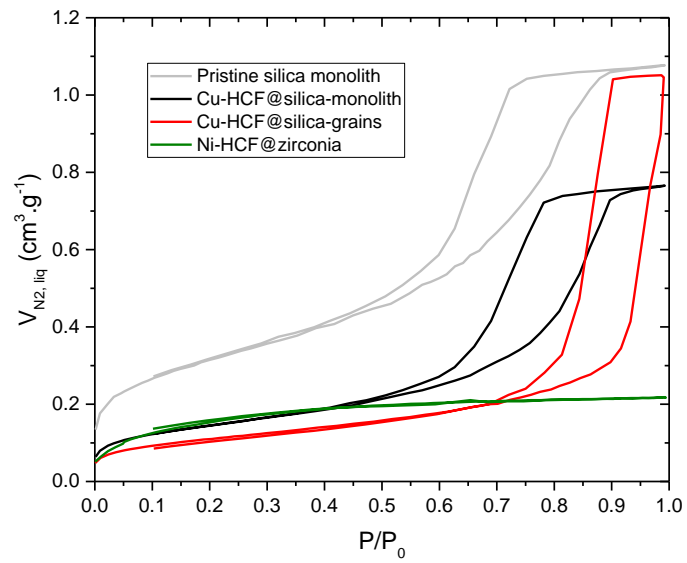


Figure S2: Adsorption isotherm silica-based monolithic samples of pristine silica monolith (grey), Cu-HCF@silica-monolith (black), Cu-HCF@silica-grains (red) and dense Ni-HCF@zirconia (green).

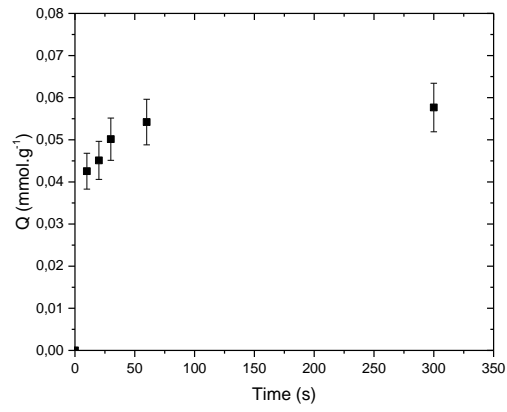


Figure S3: Kinetics of Cs sorption on ground silica monolith in freshwater ( $[Cs^+]_i = 3.9 \times 10^{-4}$  mol.L<sup>-1</sup>).

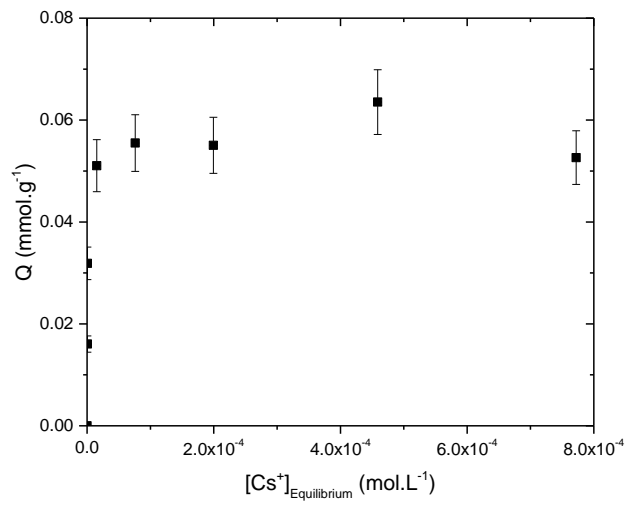


Figure S4: Sorption isotherm of Cs for ground monolith in freshwater.

Bed efficiency (BE) equation:

ES1

$$BE = \frac{\int_{V=0}^{V_R} [Cs]_i - [Cs]_{outlet} dV}{V_R * [Cs]_i}$$

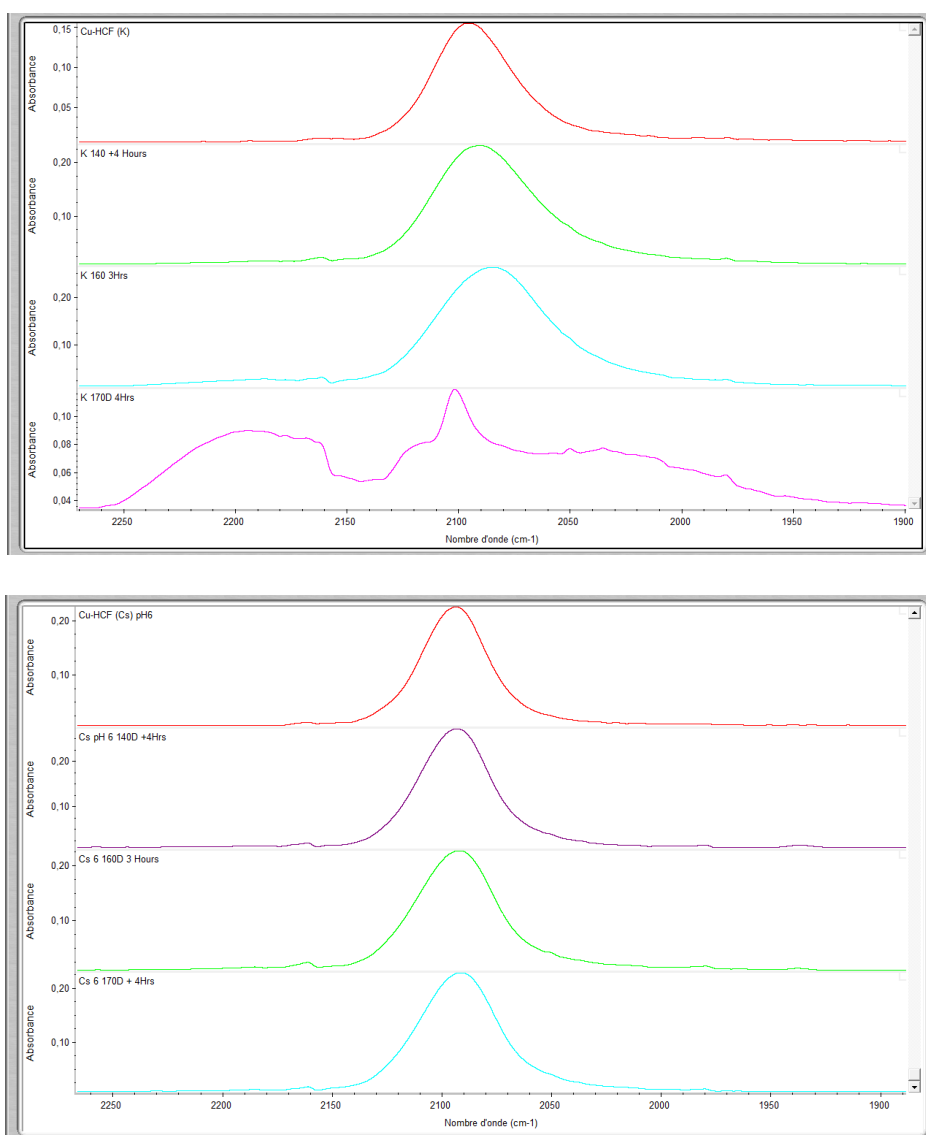


Figure S5: (Above) FT-IR showing the  $\nu(\text{CN})$  stretch of Cu-HCF which has been heated to 140, 160 and 170  $^{\circ}\text{C}$  for 4, 3 and 4 hours respectively (total 11 Hrs heating). Peak decomposition takes place at 170  $^{\circ}\text{C}$ . (Below) A Cs exchanged sample subjected to the same temperatures. No change is observed.