

The long term fate of radioactive caesium after nuclear accidents

Adam J. Fuller

Samuel Shaw, Michael B. Ward, Sarah Haigh, Caroline L. Peacock, Stephen Stackhouse, Divyesh Trivedi, Fred Mosselmans, Andy Dent and Ian T. Burke



Nuclear accidents



Radioactive caesium

Main radionuclides: caesium-137 and caesium-134

Decays via β & γ emission

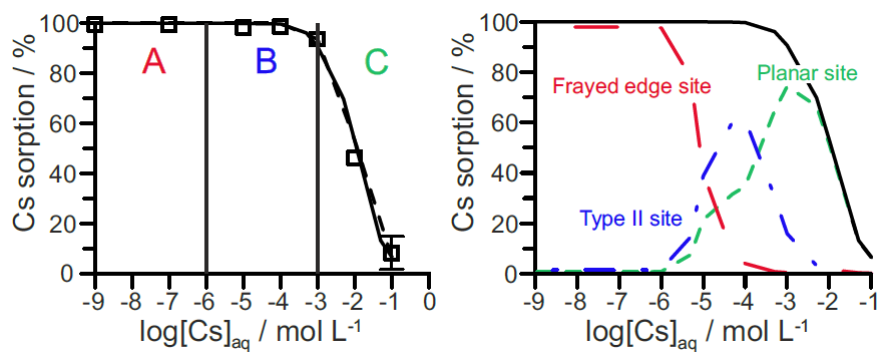
Difficult to shield against so contaminated ground gives a high dose

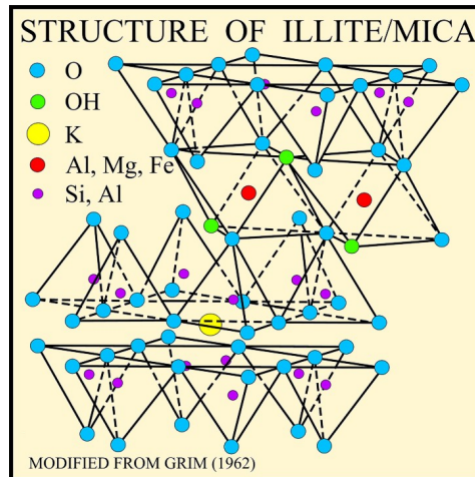
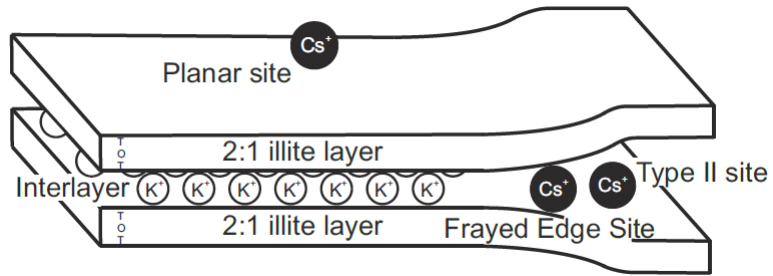
Caesium-134 has a half-life of 2 years

Caesium-137 has a 30 year half-life so it is a major long term contaminant

Does not accumulate in the body so main issue external radiation dose

Cs in the environment





Aim

Currently theories are based on extrapolation from macroscale solution experiments and computer modelling

Lack of direct evidence of specific sorption process

Aim:

Evaluate the validity of the Frayed Edge Site sorption theory by direct observation of Cs sorption to illite

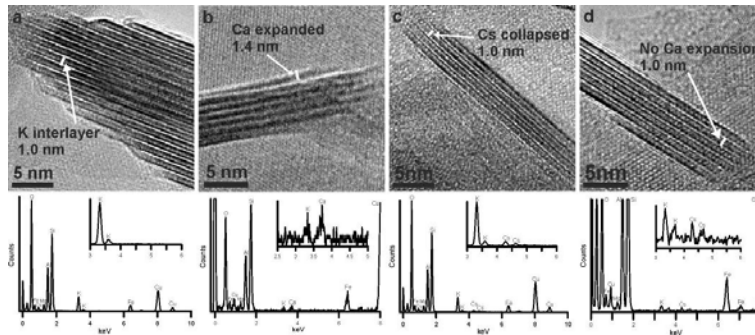
TEM Methods

- Illite suspended in CsCl solution (1 mol/L)
- Sampled at 4, 7, and 12 months
- Samples were then washed in NaCl and DIW to remove weakly sorbed Cs
- Samples were imaged using a FEI Titan ChemiSTEM
- Additional samples were suspended in CaCl for 1 month to investigate interlayer expansion processes
- Imaged using a Tecnai TF20 FEGTEM

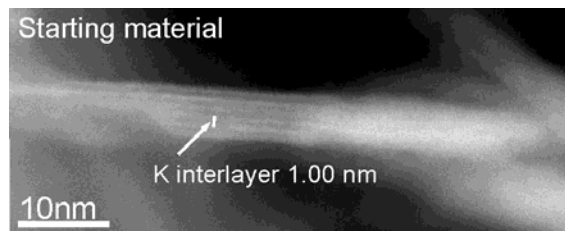


Sorption behaviour – initial

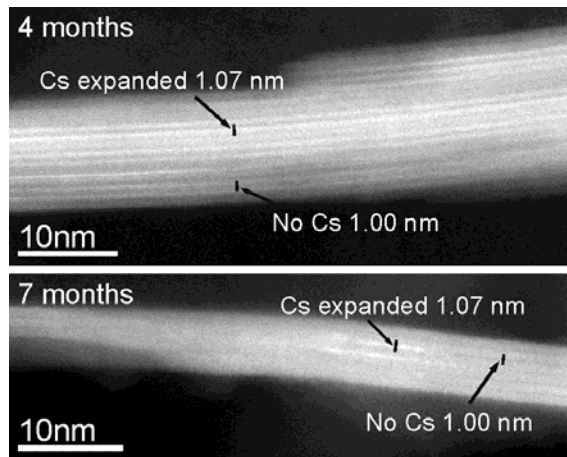
Caesium rapidly sorbs to frayed edges by exchange with calcium



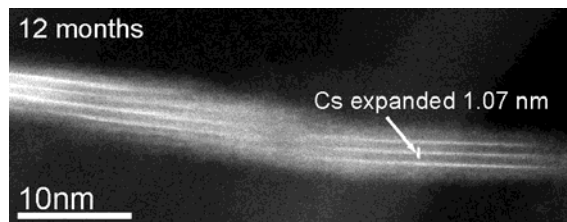
Sorption behaviour – long term



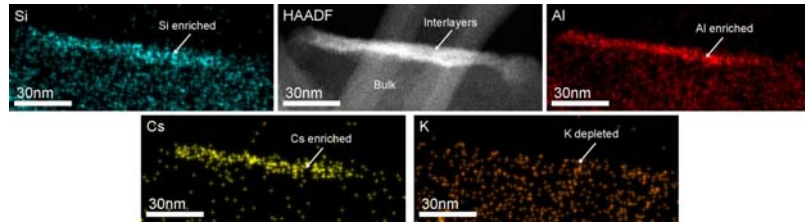
Sorption behaviour – long term



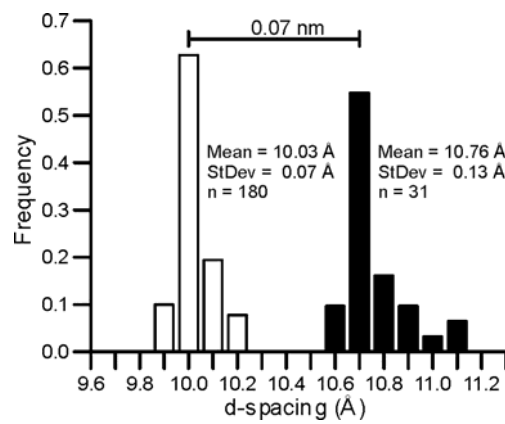
Sorption behaviour – long term



Sorption behaviour – long term



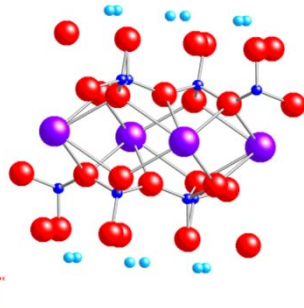
Sorption behaviour – long term



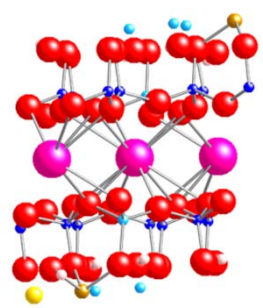
Sorption behaviour – long term

Red – Oxygen
Dark blue – Si
Light blue – Al
Purple – K

Pink - Cs

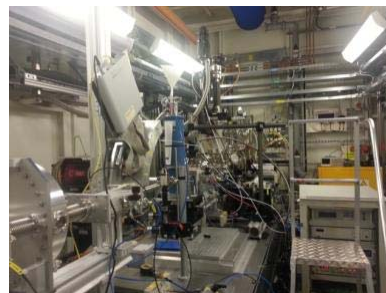


1.0 nm

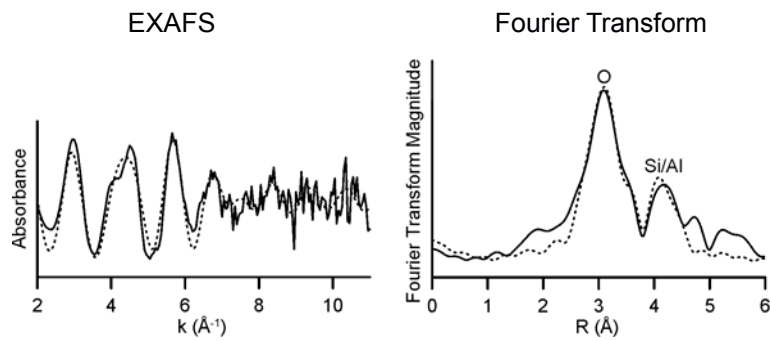


1.07 nm

EXAFS Methods



Sorption behaviour – long term



Sorption behaviour – long term

- Cs initially sorbs to expanded interlayers by cation exchange
- Cs moves further into interlayers over time by exchange with K
- Over the long term the concentration of irreversibly sorbed Cs will increase

