Radioactive cesium fallout on Tokyo from Fukushima concentrated in glass microparticles

June 27 2016

New research shows that most of the radioactive fallout which landed on downtown Tokyo a few days after the Fukushima accident was concentrated and deposited in non-soluble glass microparticles, as a type of 'glassy soot'. This meant that most of the radioactive material was not dissolved in rain and running water, and probably stayed in the environment until removed by direct washing or physical removal. The particles also concentrated the radioactive caesium (Cs), meaning that in some cases dose effects of the fallout are still unclear. These results are announced at the Goldschmidt geochemistry conference in Yokohama, Japan.

The flooding of the Fukushima Daiichi Nuclear Power Plant (FDNPP) after the disastrous earthquake on March 11 2011 caused the release of significant amounts of radioactive material, including caesium (Cs) isotopes 134Cs (half-life, 2 years) and 137Cs (half-life, 30 years).

Japanese geochemists, headed by Dr Satoshi Utsunomiya (Kyushu University, Japan), analysed samples collected from within an area up to 230 km from the FDNPP. As caesium is water-soluble, it had been anticipated that most of the radioactive fallout would have been flushed from the environment by rainwater. However, analysis with state-of-the-art electron microscopy in conjunction with autoradiography techniques showed that most of the radioactive caesium in fact fell to the ground enclosed in glassy microparticles, formed at the time of the reactor

meltdown.

The analysis shows that these particles mainly consist of Fe-Zn-oxides nanoparticles, which, along with the caesium were embedded in Si oxide glass that formed during the molten core-concrete interaction inside the primary containment vessel in the Fukushima reactor units 1 and/or 3. Because of the high Cs content in the microparticles, the radioactivity per unit mass was as high as ~4.4×10^{11} Bq/g, which is between 10^7 and 10^8 times higher than the background Cs radioactivity per unit mass of the typical soils in Fukushima.

Closer microparticle structural and geochemical analysis also revealed what happened during the accident at FDNPP. Radioactive Cs was released and formed airborne Cs nanoparticles. Nuclear fuel, at temperatures of above 2200 K (about as hot as a blowtorch), melted the reactor pressure vessel resulting in failure of the vessel. The airborne Cs nanoparticles were condensed along with the Fe-Zn nanoparticles and the gas from the molten concrete, to form the SiO2 glass nanoparticles, which were then dispersed.

Analysis from several air filters collected in Tokyo on 15 March 2011 showed that 89% of the total radioactivity was present as a result of these caesium-rich microparticles, rather than the soluble Cs, as had originally been supposed.

According to Dr Satoshi Utsunomiya: "This work changes some of our assumptions about the Fukushima fallout. It looks like the clean-up procedure, which consisted of washing and removal of top soils, was the correct thing to do. However, the concentration of radioactive caesium in microparticles means that, at an extremely localised and focused level, the radioactive fallout may have been more (or less) concentrated than anticipated. This may mean that our ideas of the health implications should be modified".

Commenting, Prof. Bernd Grambow, Director of SUBATECH laboratory, Nantes, France and leader of the research group on interfacial reaction field chemistry of the ASRC/JAEA, Tokai, Japan, said:

"The leading edge observations by nano-science facilities presented here are extremely important. They may change our understanding of the mechanism of long range atmospheric mass transfer of radioactive caesium from the reactor accident at Fukushima to Tokyo, but they may also change the way we assess inhalation doses from the caesium microparticles inhaled by humans. Indeed, biological half- lives of insoluble caesium particles might be much larger than that of soluble caesium".

More information: Conference Abstract: Plenary: Monday 27th June 11:45-12:45

Challenging Radionuclides in Environment at the Atomic Scale: Issues in Waste Disposal and Fukushima SATOSHI UTSUNOMIYA

Radionuclides are beneficial in many instances such as power generation, industrial, medical, and geochronological applications. Conversely, some fission products and actinides produced in nuclear reactors are radiotoxic and have long half-lives. These radionuclides need to be isolated and safely stored for geological periods; however, there have been instances where the release of these radionuclides has caused serious environmental issues. In such instances, the release of these radionuclides is governed by their interaction with inorganic, organic, and biological substances at the molecular scale in the Earth's surface and subsurface. Naturally-occurring nano-particles and microorganisms also play an important role in facilitating or retarding the migration of lowsolubility radionuclides. The migration processes at the nano- and atomic-scales have been illustrated by atomic-resolution

transmission electron microscopy (TEM). TEM is a powerful technique that enables us to investigate the structural and chemical properties of these particles at scales ranging from micron to sub-angstrom. This talk will address some of the observable microscopic phenomena that can impact the migration of radionuclides in surface and subsurface environments; physical and chemical alteration of nuclear waste and UO2+x, colloid-facilitated transport, microbial nanocrystallization of rare earth elements that are used as surrogate of trivalent actinides, the interaction between nanoparticles and microorganisms, and most recently contamination at Fukushima. At the present, 134Cs and 137Cs are important dose contributors, and their radioactivity will remain in soils, mainly fixed in the form of submicron-sized clay minerals. Some of the particles associated with Cs are transported while surface soils are run off. On the other hand, at the initial stage of Cs release from Fukushima Daiichi Nuclear Power Plant, low-solubility Cs-rich microparticles, which contain up to ~36 wt% of Cs as Cs2O, are responsible for ~90% of the radioactivity (rather than soluble forms of Cs such as CsOH). The interior of these particles exhibit evidence of various nanoscale processes in the molten core-concrete interaction (MCCI) that occurred subsequent to melt down in the primary containment vessel. Still, these particles play an important role in the dispersion of low-volatile radionuclides into the surrounding environment. The latter half of the talk will highlight various microscopic but critical phenomena in Fukushima as unveiled by state-of-the-art TEM investigations.

Provided by Goldschmidt Conference