First direct imaging of radioactive cesium atoms in environmental samples

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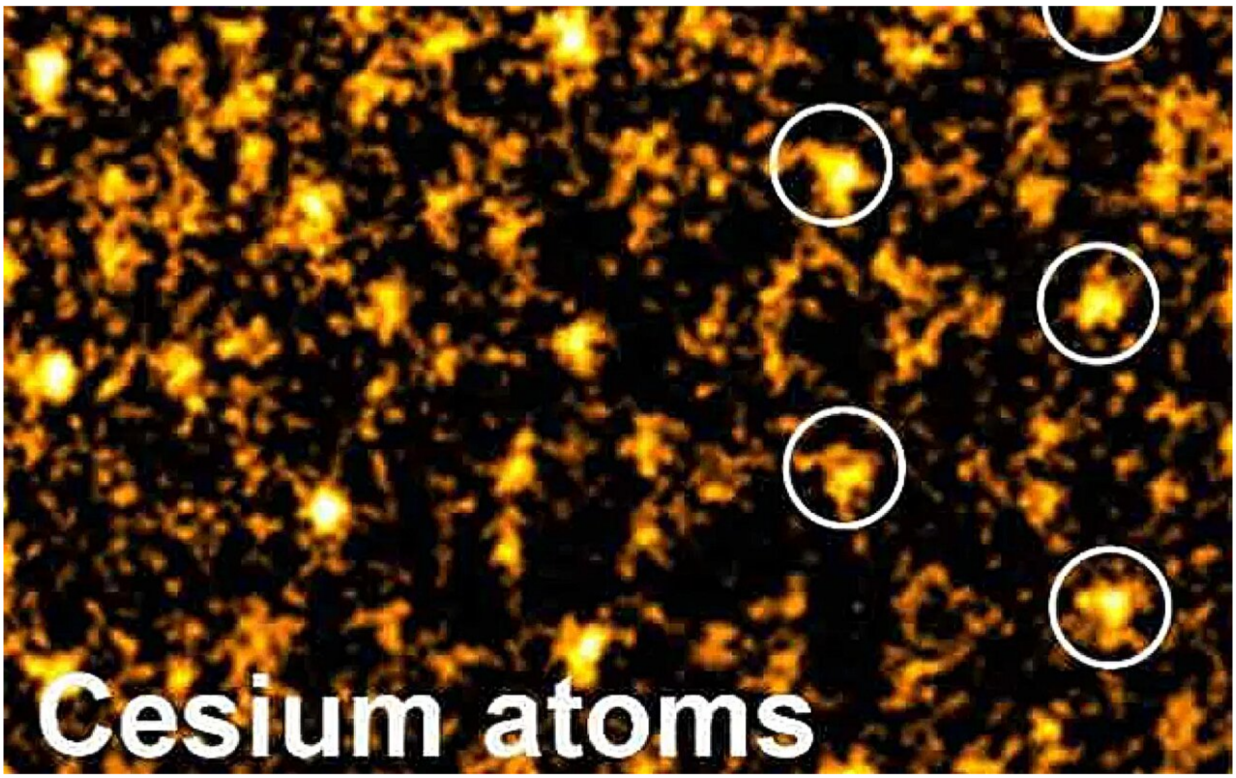

The Cs atoms in the image appear as bright spots (circled in the image). Approximately half of the Cs atoms in the structure are radioactive. Credit: *Journal of Hazardous Materials* (2024). DOI: 10.1016/j.jhazmat.2024.134104

Thirteen years after the nuclear disaster at the Fukushima Daiichi Nuclear Power Plant (FDNPP), a breakthrough in analysis has permitted a world first: direct imaging of radioactive cesium (Cs) atoms in environmental samples.

The analysis, completed by a team of researchers in Japan, Finland, America, and France, analyzing materials emitted from the damaged FDNPP reactors, reveals important insights into the lingering environmental and radioactive waste management challenges faced in Japan.

The study, titled "'Invisible' radioactive cesium atoms revealed: Pollucite inclusion in cesium-rich microparticles (CsMPs) from the Fukushima Daiichi Nuclear Power Plant" has just been published in the *Journal of Hazardous Materials*.

In 2011, after the Great Tōhoku Earthquake and Tsunami, 3 nuclear reactors at the FDNPP underwent meltdowns due to a loss of back-up power and cooling. Since then, extensive research efforts have focused on understanding the properties of fuel debris (the mixture of melted nuclear fuels and structural materials), found within the damaged reactors. That debris must be carefully removed and disposed of.

However, many uncertainties remain concerning the physical and chemical state of the fuel debris and this greatly complicates retrieval efforts.

Attempts to understand the chemistry of radioactive cesium results in a world first

A significant amount of radioactive Cs was released from the damaged Fukushima Daiichi reactors in particulate form. The particles, termed Cs-rich microparticles (CsMPs), are poorly soluble, small (

Prof. Satoshi Utsunomiya from Kyushu University, Japan, led the current study. He explained that the CsMPs "formed in the bottom of the damaged reactors during the meltdowns, when molten nuclear fuel impacted concrete."

After formation, many CsMPs were lost from the reactor containment into the surrounding environment.

How the image was created?

Detailed characterization of CsMPs has revealed important clues about the mechanisms and extent of the meltdowns. However, despite abundant Cs in the microparticles, direct atomic scale imaging of radioactive Cs in the particles has proven impossible.

Prof. Gareth Law, a study collaborator from the University of Helsinki, explained that "this means we lack full information on the chemical form of Cs in the particles and fuel debris."

Utsunomiya said, "While Cs in the particles is present at reasonably high concentrations, it is often still too low for successful atomic scale imaging using advanced electron microscopy techniques. When Cs is found at a high enough concentration, we have found that the electron beam damages the sample, rendering resulting data useless."

However, in the team's previous work using a state-of-the-art high-resolution high-angle annular dark-field scanning transmission electron microscope (HR-HAADF-STEM), they found inclusions of a mineral called pollucite (a zeolite) within CsMPs.

Law explained that "in past analysis we showed that the iron-rich pollucite inclusions in the CsMPs contained >20 wt.% Cs. In nature, pollucite is generally aluminum-rich. The pollucite in the CsMPs was clearly different to that in nature indicating it formed in the reactors.

"Because we knew that most of the Cs in CsMPs is fission derived, we thought that analysis of the pollucite could yield the first ever direct images of radioactive Cs atoms."

Zeolites can become amorphous when subjected to electron beam irradiation, but that damage is related to the composition of the zeolite, and the team found that some pollucite inclusions were stable in the electron beam.

Learning this and informed by modeling, the team set about painstaking analysis that saw Utsunomiya, graduate student Kanako Miyazaki, and the team finally image radioactive Cs atoms.

Utsunomiya explained, "It was incredibly exciting to see the beautiful pattern of Cs atoms in the pollucite structure, where about half of the atoms in the image correspond to radioactive Cs. This is the first time humans have directly imaged radioactive Cs atoms in an environmental sample.

"Finding concentrations of radioactive Cs high enough in environmental samples that would permit direct imaging is unusual and presents safety issues. While it was exciting to make a scientific world first image, at the same time it's sad that this was only possible due to a nuclear accident."

More than an imaging breakthrough

Utsunomiya emphasized that the study's findings are broader than mere imaging of radioactive Cs atoms. "Our work sheds light on pollucite formation and the likely heterogeneity of Cs distribution within the FDNPP reactors and the environment."

Law said, "We unequivocally demonstrate a new Cs occurrence associated with the materials emitted from the FDNPP reactors. Finding Cs containing pollucite in CsMPs likely means it also remains in the damaged reactors; as such, its properties can now be considered in reactor decommissioning and waste management strategies."

Collaborator Emeritus Prof. Bernd Grambow from Subatech, IMT Atlantique Nantes University, added, "We should now also begin to consider the environmental behavior or Cs-pollucite and its possible impacts. It likely behaves differently to other forms of Cs fallout documented thus far.

"Also, the effect on human health might have to be considered. The chemical reactivity of pollucite in the environment and in body fluids is certainly different than that of other forms of deposited radioactive Cs."

Finally, reflecting on the study's significance, Prof. Rod Ewing from Stanford University underscored the pressing need for continued research to inform debris removal strategies and environmental remediation. "Yet again, we see that the pain-staking analytical efforts of international scientists really can unlock the mysteries of nuclear accidents, aiding long-term recovery efforts."

More information: Kanako Miyazaki et al, "Invisible" radioactive cesium atoms revealed: Pollucite inclusion in cesium-rich microparticles (CsMPs) from the Fukushima Daiichi Nuclear Power Plant, *Journal of Hazardous Materials* (2024). DOI: 10.1016/j.jhazmat.2024.134104

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