ON INDOOR ENVIRONMENT CONTAMINATION BY CESIUM-RICH RADIOACTIVE MICROPARTICLES RELEASED FROM THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT: A REVIEW

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Abstract. *Advanced nuclear technologies in conditions of geological disaster can lead to unpredictable ecological hazards. Through unknown processes Cesium-rich micro-particles CsMPs formation took place during melting inside the reactor. Not much has been documented on the occurrence of CsMPs inside buildings. After Fukushima disaster new ways of radiation contamination of the natural environment by CsMPs were identified.*

Key words: *Cesium-rich micro-particles, radioactive materials, indoor CsMPs*`

It was difficult to predict three disasters happened simultaneously in Japan. As a result of the processes, developed at the Fukushima Daiichi nuclear power plant, the catastrophe destroyed the complex and interdependent security systems. Clearly, the study of contamination process will help to increase security of power generation facilities in Japan and around the world. The most serious environmental consequences of the accident, including surface pollution by radioactive cesium, have not yet been explored. The big challenge of modern environmental technologies is to control cesium nanoparticles in nature. Conclusions about reactions, developed in the Fukushima reactors, have been based on indirect studies. It is believed that after the shutdown of the cooling system, the temperature in the reactor rise to 2,200 K and the radioactive Cs was released from the irradiated fuel. All CsMPs (2.0-3.4 μm in size) contain SiO2 glass and Zn-Fe-oxide nanoparticles associated with a wide range of Cs concentrations. Uranium U traces are also associated with Zn-Fe-oxides. Cesium-rich micro-particles of CsMPs generated at the Fukushima Nuclear Power Plant were first discovered in the atmosphere 170 km southwest of the plant. These particles are solid body objects. Their formation took place during melting inside the reactor and they carry important information about the physical and chemical properties of the radioactive materials inside the reactor. CsMPs formation process studies use a high resolution electron microscope combined with conventional radio-analytical techniques and study of chemical and structural properties on an atomic scale [1,2]. Interestingly, the process of formation of CsMPs at Fukushima is markedly different from the presented above process of microparticle formation. It takes place as a result of interaction of the melted active zone of the reactor in experiments [3,4,5] with concrete particles (a process known as molten core concrete interaction - MCCI).

Due to the extremely high radioactivity per unit mass, ~ 10^{11} Bq/g, CsMPs particles can be an important source of radiation in the Fukushima environment. In addition, CsMPs are important carriers, through which volatile radionuclides, such as uranium (U), reach the environment [2]. Usually instruments used for radiological measurements shall be selected based upon the instrument's detection capability for each known or potential radionuclide. In the case of man-made CsMPs, the produced nano-particles are radiation sources, but it is impossible to monitor their concentration by usual radiation detectors. The CsMPs detection is not the only technical problem: besides expensive specific equipment and highly technical qualification of the staff, one need to know the particles complex migration path in the media. Inhalation of CsMPs is associated with serious health risks especially indoors. It is very important to determine during analysis, if artificial nanoparticles and specifically

CsMPs retain their properties (size, original structure, reaction properties) in water, air, soil or sediment. Accidentally produced man-made CsMPs pose a significant environmental challenge in terms of both discovery and modeling of further migration pathway. CsMPs are now known to have been transported long distances from Fukushima. In March 2011, ~520 PBq of radionuclides were released into environment during the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. In 2017 Caesium-rich micro-particles emission first time was discovered via the atomic-resolution electron microscopy. CsMPs analysis served as a window into the meltdown event process at the Fukushima Daiichi Nuclear Power Plant. In 2023 the article "Occurrence of radioactive cesium-rich micro-particles (CsMPs) in a school building located 2.8 km south-west of the Fukushima Daiichi Nuclear Power Plant' was published [6]. So the chain of discoveries during last years point to the need of study the new radioactive nano-particles formations. Through analysis of 20 soil samples with the QCP method, the first regional-scale quantitative map of CsMP abundance and RF values around the FDNPP was presented in the article "Abundance and distribution of radioactive cesium-rich microparticles released from the Fukushima Daiichi Nuclear Power Plant into the environment" (Chemosphere, Volume 241, February 2020, 125019). Researchers from Kyushu University, the University of Helsinki, IMT Atlantique, the University of Nantes and Stanford University published a study in the journal Chemosphere that revealed the presence of significant amounts of highly radioactive and poorly soluble cesium-rich microparticles (CSMPS) in an abandoned school building near the Fukushima Daiichi nuclear power plant. These microparticles, which entered the building during the March 2011 accident, can pose a health hazard if inhaled. The results highlight the importance of considering indoor CsMPs .

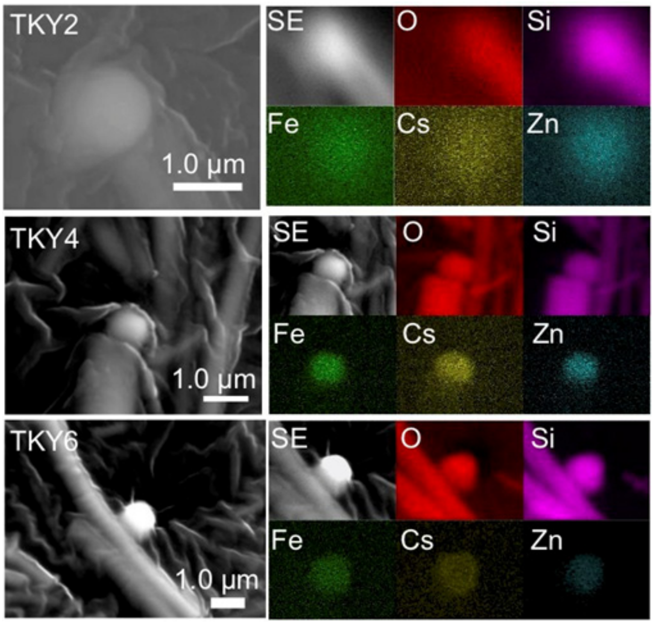

Fig.1. Secondary electron images of CsMPs discovered in atmospheric particles trapped on a Tokyo air filter from March 15, 2011, with major constituent elements displayed (Utsunomiya et al. 2019).
Photo from internet source https://safecast.org/2019/08/fukushima-cesium-enriched-microparticle-csmp-update/.

Fig. 1 presents secondary electron images of CsMPs discovered in atmospheric particles trapped on a Tokyo air filter from March 15, 2011, with major constituent elements displayed. These tiny particles with a radius of only a few micrometers are mainly composed of silica glass, Fe and Zn. It is important to note that the content of radioactive Cs in CsMP exceeds that of any other form of Cs-labeled radioactive fallout released from damaged reactor units 1-3. Studies by (Utsunomiya et al. 2019) show that these particles were temporarily formed during FDNPP meltdown events when molten fuel escaped from the primary containment and interacted with the underlying concrete base. While previous research has focused primarily on CsMP found outdoors, the most recent scientific investigations demonstrate that CSMPS can significantly penetrate buildings, creating a potential risk indoors. This finding highlights the need for buildings in the exclusion zone to be thoroughly cleaned before they can be safely reintegrated for public use.

Levels of Cs radioactivity in dust samples collected from school floors ranged from 340 to 4040 Bq/m^2 (corresponding to 125-1490 Bq/m^2 in 2023), with 4.5 to 38.9% of the radioactivity coming from CsMPs. The highest concentration of CsMPs was found at the entrance to the school, with up to 2,481 particles per m^2, indicating that these radioactive particles penetrate deeply into the building. In contrast, lower but still significant amounts of CSMPS were found on the second floor, up to 1273 particles per m^2. Interestingly, the amount of CsMPs and the fraction of Cs radioactivity associated with CsMPs in samples collected outdoors were significantly lower, indicating a discrepancy between indoor and outdoor pollution levels. Researchers believed that the CSMPS present in the premises were formed by dry deposition as the school had remained untouched since the accident. On the other hand, samples taken outdoors recorded both dry and wet deposition of Cs precipitation. It is likely that outdoor CSMPs were partially washed away or blown away by rain and wind, leaving predominantly soluble Cs species associated with dust and soil particles [8].

The investigation of CsMPs infiltration into environment and its further possibly global migration is one of main ecological and eco-toxicological challenges of modern science.

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