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**Andersson, Kasper Grann**

*Published in:*
Radioprotection

*Link to article, DOI:*
10.1051/radiopro/2016040

*Publication date:*
2016

*Document Version*
Peer reviewed version

# Physico-chemical properties of radionuclides emitted as particulate matter

K. G. Andersson

*Technical University of Denmark (DTU), Center for Nuclear Technologies (Nutech), Frederiksborgvej 399, Bldg. 201, DK-4000 Roskilde, Denmark*

**Abstract.** This paper presents work done to improve the representation in European decision support tools of physico-chemical forms of radiocontaminants released to the atmosphere from a major nuclear power plant accident. The task is to accommodate those types of scenarios where fuel particles are at play. A methodology with associated parameters has been developed to improve modelling of the aerodynamic particle characteristics, and resultant deposition relations on different surface types, as well as estimates of resultant post-deposition environmental contaminant mobility / forced decontamination.

Keywords: **NPP accident / emergency / decision support / atmospheric release / aerosols**

## 1. INTRODUCTION

The contaminant releases from the Fukushima and Chernobyl Nuclear Power Plant (NPP) accidents have been recorded to result in dispersion of radionuclides with different physico-chemical forms. Most contaminant aerosol size spectra measured immediately after the Chernobyl accident were recorded at distances of hundreds of kilometres (Andersson, 2015), and here the by far dominant physico-chemical form of more volatile element contaminants, such as Cs, was small, often condensation mode, aerosols with an aerodynamic diameter of about half of a micron. This was expected, as gravity would at such distances have depleted most larger particles from the plume. However, even in air as far away from Chernobyl as central Germany, the caesium aerosol distribution was found to be slightly bimodal, with a second fraction being several microns in aerodynamic diameter. This clearly demonstrated that different release processes were at play within a short time range. In fact, investigations have shown that at distances exceeding 50 km in some directions from the Chernobyl NPP, most of the deposited caesium was in the form of fuel particles. Also many other contaminants were released at Chernobyl, some of which were in part volatilised and other only released as fuel particles. In contrast, releases from the much less powerful Fukushima accident process were largely of volatile contaminant elements, where caesium (condensation particles) and iodine (gaseous and condensation particles) were of concern. This work is aimed at improving parameterisation in the ARGOS and RODOS NPP accident decision support systems (DSS) by considering physico-chemical forms of contaminants in relation to the release process. In the past, only the radionuclides released and the fractions of these released as gases were associated with the accident type.

## 2. MATERIALS AND METHODS

*Influence of physico-chemical form on environmental solubility*

It is known from extensive experimentation made in the Chernobyl 30 km zone that atmospherically dispersed fuel particles (uranium oxides with enclosures of other contaminants) degrade very slowly after deposition in the environment, leaching contaminants at a rate depending on soil pH and on whether they were oxidised during the release process. Kashparov

(see Andersson, 2015) has on the basis of his experimentation created time functions for the release of contaminants from fuel particles, according to particle oxidation and environmental (soil) pH, which can be used in accident decision support.
If material was initially oxidised, the dissolution rate constant after deposition in soil should be:

$$k\left(\text{years}^{-1}\right) = 0.6 \cdot 10^{(-0.15\,pH)} \text{ at } pH < 7.0 \text{ and } k = 0.05 \text{ at } pH > 7.0 \qquad (1)$$

If the material was NOT initially oxidised, the dissolution rate constant should be:

$$k\left(\text{years}^{-1}\right) = 40 \cdot 10^{(-0.45\,pH)} \text{ at } pH < 6.5 \text{ and } k = 0.05 \text{ at } pH > 6.5 \qquad (2)$$

This means that particularly if the material was not (or only partially) oxidised, it could well take years before most of the contamination is leached and can start to migrate freely and become available for uptake through food chains. In the DSS, it has in the past been assumed that all deposited contaminants move as if they were simple ions, readily available for all uptake.

*Physico-chemical forms from different accident types*

The definitions of the different 46 reactor accident scenarios in RODOS have been scrutinised by a reactor expert (see Andersson, 2015) with a view to identifying those scenarios where fuel particles might be released and whether these would be oxidised or not. None of the design basis accidents (DBA) could possibly lead to release of fuel matrix particles. Essentially no modifications are thus needed in any models to estimate the radiological consequences of those types of accidents. However, for 'Beyond DBA' scenarios, part of the released radioactive material could possibly be in the form of fuel matrix, following some limited degree of fuel melting (not the very powerful process that occurred at Chernobyl with a now obsolete reactor type). In connection with any considered severe accident there may be a release of fuel particles, with particularly high likelihood where the situation involves a reactor explosion. It was concluded that oxidation would be expected to occur in connection with all the scenarios. Including in the decision support modelling Kashparov's figures for leaching from oxidised particles in the decision systems may thus be tempting, but as oxidation may well not be complete, it could be wise to also enable visualisation of the effect on dose if particles were non-oxidised.

*Characterisation of radiocontaminants of possible concern*

A short list of radionuclides (Andersson, 2015), considered (by Slovakia, France, Germany, Finland, Czech Rep. and USA) for evaluation of radiological consequences in case of severe NPP accidents was used in selecting the radionuclides of greatest potential concern (Rb, Sr, Te, I, Cs, Ba, Zr, Nb, Mo, Ru, Sb, Ce, Pu, Np, Am and Cm).

In connection with the Chernobyl accident, it was found that contaminants of certain elements were only released to the atmosphere in the form of low solubility fuel particles, indicating that these would in general be expected to be highly refractory (undepleted). These comprised Zr-95, Nb-95, Ce-141/144, Np-237/239, Pu-238-242, Am-241/243 and Cm-242/244 (Andersson, 2015). The smallest 'fuel' particles (i.e. those that would travel to the greatest distances) were in April 1986 measured in air in Prague (Andersson, 2015) to have an activity median aerodynamic diameter (AMAD) of about 3-4 µm (measured for Nb-95 and Zr-95), and the larger AMAD of this particle group compared to all other Chernobyl aerosols measured here demonstrates the different origination process. From this knowledge combined with experience from the impact of explosive tests on a matrix of uranium dioxide, a likely initial fuel particle size distribution was estimated for prognostic modelling prior to actual assessment of the case-specific particle size distribution.

In the past, only two categories of airborne particles have been considered in the European DSS:

ca. 0.5 µm particles characteristic of condensation mode, and ca. 2-5 µm particles characteristic of the particles measured in the Chernobyl case at distances of hundreds of kilometres. However, considerable amounts of much larger particles were spread out over an area with distances of over a hundred kilometres in the Chernobyl case (Andersson, 2015), and although the release process may well be much less powerful in a future severe accident, large particles may with respect to some important radionuclides dominate the deposited contamination over very large areas.

*Importance of sizes of released airborne particles*

It is crucial to model the size of airborne particles adequately, as the deposition velocity to a grassed reference surface of a particle in the 10-20 µm range is several orders of magnitude greater than that of a ca. 0.5 µm particle (the size at which both the impact of gravity and Brownian diffusion on deposition reaches a minimum). Like Cs, also Te, Rb, Sb and Mo have generally been found to be more volatile than for instance Sr, and thus in a severe accident producing fuel particles generally to a lesser extent associated with these. To adequately take into account differences in airborne particle sizes, new wet and dry particle deposition data libraries have been compiled in the PREPARE project (Andersson, 2015) to be considered in the ARGOS and RODOS systems . The highly volatile iodine has in the Chernobyl case only been recorded as gas and small condensation particles. Iodine aerosol spectra at different distances after the Chernobyl accident thus show a perfect Gaussian distribution with no signs of bimodality (Andersson, 2015).

*Ruthenium – a special case*

Ruthenium is special in that it has a very high elemental boiling point (2700°C), which would in practically any conceivable incident scenario prevent it from being volatilised and depleted from fuel material, unless it can be oxidised to its highly volatile tetraoxide form.

*Natural migration and decontamination in inhabited areas*

As pointed out above, environmental dissolution of fuel particles may be slow, and this affects the migration and uptake to biota. However, also the mobility in inhabited areas and the effect of a number of dose reductive countermeasures that would generally be recommended for such areas, depend considerably on the particle size and environmental solubility. Parameters to accommodate this were also defined in the project (Andersson, 2015).

## 3. RESULTS AND DISCUSSION

As an example of the importance of adequately considering the physico-chemical forms of contaminants in connection with an NPP emergency releasing fuel particles to the environment, consider that all contaminants are in the DSS currently assumed to be on readily bioavailable form. However, the analyses by Kashparov show that contaminants bound in fuel particles will only very slowly (over years, depending on various parameters) be released and become mobile in the environment. In the earliest weeks after an accident, it will be observed that only a few percent of the contamination in a fuel particle contaminated area can thus be transferred, e.g., to crops, animals and humans, which can currently only be accommodated in the model by scaling radionuclide transfer factors down by accordingly (perhaps by a factor of 30). Countermeasure optimisation would then be made on the basis of DSS runs with these low transfer factors. A countermeasure may then be chosen that can reduce the transfer to crops over the following year by say 80 % (e.g., by adding a fertiliser). If for instance caesium is a dominant contaminant element, a large part of the contamination will over the next few years become strongly bound in

clay minerals (as also modelled in the current version of the DSS). The residual dose contribution estimated in the countermeasure optimisation process is thus little compared with the dose that can be saved. However, in reality, only few percent of the contamination has at that time been on a physico-chemical form allowing crop uptake, and the remaining perhaps 97% of the contamination will be released in the years to follow, giving a comparatively huge residual dose contribution. It also needs to be taken into account that large particles will be much more readily removed from impermeable surfaces than small ones, both by natural and forced removal processes.

## 4. CONCLUSIONS

A methodology has been derived to enable consideration in the European DSS of physico-chemical properties of radionuclides emitted as particulate matter. Comprehensive data libraries for inclusion in the DSS have been created. Primary issues are differences in particle size leading to different environmental deposition velocities, post-deposition environmental solubility, and influence of particle size on post-deposition environmental mobility and forced decontamination.

**Acknowledgement**
The research leading to these results has received funding from the European Atomic Energy Community Seventh Framework Programme FP7/2012-2013 under grant agreement 323287.

**References**
Andersson, K.G. (2015). Recommendations for taking into account different physico-chemical forms of contaminants in modelling deposition following a major NPP accident, PREPARE(WP4)-(15)-02. Deliverable D4.3/4.4 of the EURATOM FP7 project PREPARE.