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Experimental work on the adsorption behaviour of iodine oxide aerosols on surfaces in the containment during a severe nuclear accident

(AIAS project: Adsorption of Iodine oxide Aerosols on Surfaces)

1. Topic of research

1.1. Background

The AIAS (Adsorption of Iodine oxide Aerosols on Surfaces) project is a proposal for an experimental co-operation between two research institutions in Scandinavia: Chalmers Technical University of Technology (Göteborg, Sweden) and the VTT Technical Research Centre of Finland (Espoo, Finland). The goal of the co-operation is the investigation of the behaviour of iodine oxide aerosol particles in the containment during a hypothetical severe nuclear accident.

Iodine is a radiologically important fission product because it is biologically active and can exist in volatile forms both as inorganic and organic iodine. Volatile inorganic iodine species such as elemental iodine (I_2) and organic iodine compounds such as methyl iodide (CH_3I) can form during an accident. Since plant assessments show that iodine contributes significantly to the source term for a range of accident scenarios, research on the behaviour of iodine species is of great interest.

Previous experiments within the NROI (**N**ordic **R**esearch on **R**adiolytic **O**xidation of **I**odine) project have shown that the most radiological relevant and volatile gaseous iodine species (I_2, CH_3I) can react with radiolysis products from air and steam (O_3, NO_2, $OH^{\cdot}$). Under the presence of radiation in form of UVC radiation or gamma radiation collectable iodine oxide aerosols were formed and detected. Persistent iodine aerosol concentration was also measured in Phebus FP experiments even 36 h after isolation of the vessel. Such concentration can only be explained by formation of iodine containing particles within the containment.

The behaviour of these aerosols in the containment is not well understood and needs to be further investigated to achieve a better understanding on the iodine chemistry during a severe nuclear accident.

The goal is to investigate the adsorption/desorption and revaporisation behaviour of the iodine oxide particles on different surface materials in the containment, such as paints, and various reactive metal surfaces.

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1.2. Method

Iodine oxide aerosol particles labelled with ^{131}I will be produced and identified with a modification of the already existing EXSI (**EX**perimental **S**tudy on **I**odine Chemistry) facility, used in the previous NROI projects. The formed particles will be collected on different surface materials with an impactor, which is connected to the modified EXSI facility. The surfaces that will be studied are present in containments and potentially relevant during a severe accident. The materials considered are: Fresh and on different levels aged paints (aging parameters: heat, irradiation), chemically modified paints and structural metals (zinc, aluminium, stainless steel), electrical system materials (copper) and catalytic hydrogen recombiners (palladium/platinum alloys). They will be exposed to non- and radioactive iodine oxide particles and will be investigated with and without gamma irradiation (^{60}Co, 20 kGy h^{-1}). The interaction of the iodine aerosols with the paint surfaces will be compared with solutions of iodine oxides formed by reacting O_3 and I_2 in an organic solvent (Woodgate's method).[i]

After reacting the surfaces with the iodine oxide particles the samples will be pre-characterised by physical methods including XPS, ESCA, XRD, Raman spectroscopy and SEM. The binding of the iodine to the surface will be determined by radio-analytical means (HPGe measurements and autoradiography).

In the case of the palladium/platinum alloy surface the characterisation will include a test to determine if the catalytic surface has been inactivated (poisoned) by exposure to the iodine aerosol. This evaluation will be performed at Chalmers using a miniature chemical reactor and chemisorption equipment.

The release of iodine from the particles into the gas phase as well as into the water pool will be analysed in the FOMICAG (**F**acility set-up for **O**n-line **M**easurements of the **I**odine **C**oncentration in an **A**queous and a **G**as phase)
facility at Chalmers (see Figure 1).

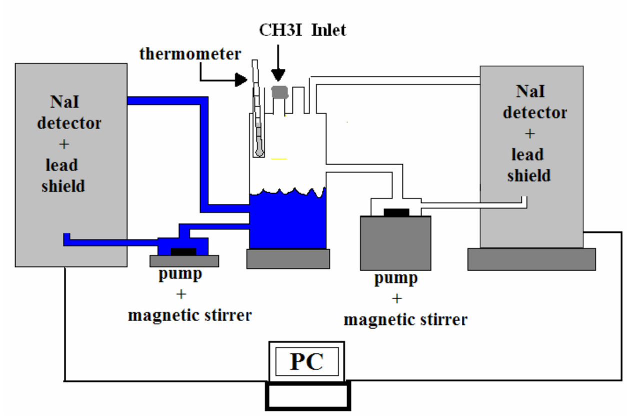

Figure 1: Schematic of the FOMICAG set-up (Chalmers)

The set-up represents a lab-scale of a BWR with the relative proportions of the Swedish BWR Oskarshamn 3. It allows a study of iodine species released under various conditions from the iodine oxide aerosol adsorbed on different surfaces.

It consists of a central vessel with a removable lid with four openings through which samples can be introduced to the system. The vessel is filled up to one-third with an aqueous solution of desired pH and composition during an experiment. The samples can be placed through one of the openings in the lid with a glass hook into the gas or aqueous phase.

The vessel is connected with pipes to one aqueous and one gaseous loop. The phases are circulated by two centrifugal pumps to ensure mass transfer. The system can be flushed with different atmospheres such as air, nitrogen and argon. Both aqueous and gas phase can be temperature controlled by heating plates and heating bands. In the centre of both loops NaI(Tl) scintillation detectors are placed detecting the evolution of ^{131}I concentration in gas and aqueous phase on-line.

Through another opening in the lid also additional chemicals like gaseous methyl iodide, chlorine etc. can be added to the system (both aqueous and gas phase) during the experiment and their influence on the chemical system can be studied.
By using the set-up a complex matrices of experiments can be performed by varying the following parameters:

- Composition of the gaseous atmosphere: air, nitrogen, argon etc.
- Composition of the aqueous phase: pH variation
- Temperature variation in both phases
- Sample placement: Aqueous phase or gaseous phase
- Adding of additional gaseous and liquid chemicals during the experimental through openings in the lid to change the chemical system during an experiment
- Sampling of the gas phase for e.g. HPGe analysis are possible through septa being attached to lid openings
- Sampling of the aqueous phase allow specifications of it e.g. pH measurements, ion chromatography of formed iodine species

After investigating the distribution of released iodine from the samples over a defined time period for certain conditions the samples will be past-examined to analyse remaining deposits and possible surface changes by analytical methods in analogy to the pre-examination of the samples (see above).
Both non-radiated and with the gamma source (^{60}Co, 20 kGy h^{-1}) irradiated surfaces in order of severe accident conditions will be investigated.

The experimental work will be performed at Nuclear Chemistry, Chalmers (SE) and at Fine Particles laboratory, VTT Technical Research Centre of Finland (FI) by the PhD students Sabrina Tietze (Chalmers, SE) and Teemu Kärkelä (VTT, FI). The aerosol production, adsorption and aerosol analysis will be performed using the EXSI facility at VTT (mainly non-radioactive samples). Radioactive samples will be produced with required parts from the EXSI facility at Chalmers Technical University.

The desorption/release experiments in the FOMICAG facility as well the analytical examination will be performed by Sabrina Tietze at Chalmers.

1.3. Reporting plan

The project is planned to be set for 2011. The experimental work is planned for spring/summer 2011 within an at least six-month period.

The results from this project will be presented within peer-reviewed scientific papers.

The final reports of the project will be submitted until March 2012.

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2. Organisation and supervision of the project

The experimental work will be performed at Chalmers Technical University in Göteborg, Sweden and at VTT Technical Research Centre of Finland in Espoo under the supervision of Professor Christian Ekberg (Nuclear Chemistry, Chalmers) and Ari Auvinen (VTT, Fine Particles).

The project will be supported by co-supervision by Dr. Henrik Glänneskog (Vattenfall, Sweden) and Dr. Mark Foreman (Nuclear Chemistry, Chalmers).

Required parts of the EXSI facility and samples produced at VTT in Finland will be transported to Chalmers (Sweden).

The experimental work will be performed in collaboration with Teemu Kärkelä (Fine Particles, VTT) and Sabrina Tietze (Nuclear Chemistry, Chalmers).

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[i] R.C Cambie, R.C. Hayward, B.G. Lindsay, A.I.T. Phan, P.S. Rutledge and P.D. Woodgate, *Journal of the Chemical Society Perkin I Transactions*, 1976, 1961-1966.