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Interactions between Sodium Aerosols and Fission Products: A Theoretical Chemistry and Experimental Approach

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Abstract : Safety requirements for Generation IV nuclear reactor designs, especially the new generation sodium-cooled fast reactors (SFR) require a risk-informed approach to model severe accidents (SA) and their consequences in case of outside release. In SFRs, aerosols are produced during a core disruptive accident when primary system sodium is ejected into the containment and burn in contact with the air; producing sodium aerosols. One of the key aspects of safety evaluation is the in-containment sodium aerosol behavior and their interaction with fission products. The study of the effects of sodium fires is essential for safety evaluation as the fire can both thermally damage the containment vessel and cause an overpressurization risk. Besides, during the fire, airborne fission product first dissolved in the primary sodium can be aerosolized or, as it can be the case for fission products, released under the gaseous form. The objective of this work is to study the interactions between sodium aerosols and fission products (Iodine, toxic and volatile, being the primary concern). Sodium fires resulting from an SA would produce aerosols consisting of sodium peroxides, hydroxides, carbonates, and bicarbonates. In addition to being toxic (in oxide form), this aerosol will then become radioactive. If such aerosols are leaked into the environment, they can pose a danger to the ecosystem. Depending on the chemical affinity of these chemical forms with fission products, the radiological consequences of an SA leading to containment leak tightness loss will also be affected. This work is split into two phases. Firstly, a method to theoretically understand the kinetics and thermodynamics of the heterogeneous reaction between sodium aerosols and fission products: I2 and HI are proposed. Ab-initio, density functional theory (DFT) calculations using Vienna ab-initio simulation package are carried out to develop an understanding of the surfaces of sodium carbonate (Na2CO3) aerosols and hence provide insight on its affinity towards iodine species. A comprehensive study of I2 and HI adsorption, as well as bicarbonate formation on the calculated lowest energy surface of Na2CO3, was performed which provided adsorption energies and description of the optimized configuration of adsorbate on the stable surface. Secondly, the heterogeneous reaction between (I2)g and Na2CO3 aerosols were investigated experimentally. To study this, (I2)g was generated by heating a permeation tube containing solid I2, and, passing it through a reaction chamber containing Na2CO3 aerosol deposit. The concentration of iodine was then measured at the exit of the reaction chamber. Preliminary observations indicate that there is an effective uptake of (I2)g on Na2CO3 surface, as suggested by our theoretical chemistry calculations. This work is the first step in addressing the gaps in knowledge of in-containment and atmospheric source term which are essential aspects of safety evaluation of SFR SA. In particular, this study is aimed to determine and characterize the radiological and chemical source term. These results will then provide useful insights for the developments of new models to be implemented in integrated computer simulation tool to analyze and evaluate SFR safety designs.

Keywords : iodine adsorption, sodium aerosols, sodium cooled reactor, DFT calculations, sodium carbonate
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