## Density Functional Theory Study of the Surface Interactions between Sodium Carbonate Aerosols and Fission Products

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Abstract : The interaction of fission products (FP) with sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) aerosols is of a high safety concern because of their potential role in the radiological source term mitigation by FP trapping. In a sodium-cooled fast nuclear reactor (SFR) experiencing a severe accident, sodium (Na) aerosols can be formed after the ejection of the liquid Na coolant inside the containment. The surface interactions between these aerosols and different FP species have been investigated using ab-initio, density functional theory (DFT) calculations using Vienna ab-initio simulation package (VASP). In addition, an improved thermodynamic model has been proposed to treat DFT-VASP calculated energies to extrapolate them to temperatures and pressures of interest in our study. A combined experimental and theoretical chemistry study has been carried out to have both atomistic and macroscopic understanding of the chemical processes; the theoretical chemistry part of this approach is presented in this paper. The Perdew, Burke, and Ernzerhof functional were applied in combination with Grimme's van der Waals correction to compute exchange-correlational energy at 0 K. Seven different surface cleavages were studied of Y-Na<sub>2</sub>CO<sub>3</sub> phase (stable at 603.15 K), it was found that for defect-free surfaces, the (001) facet is the most stable. Furthermore, calculations were performed to study surface defects and reconstructions on the ideal surface. All the studied surface defects were found to be less stable than the ideal surface. More than one adsorbate-ligand configurations were found to be stable confirming that FP vapors could be trapped on various adsorption sites. The calculated adsorption energies (Eads, eV) for the three most stable adsorption sites for  $I_2$  are -1.33, -1.088, and -1.085. Moreover, the adsorption of the first molecule of  $I_2$ changes the surface in a way which would favor stronger adsorption of a second molecule of I2 (Eads, eV = -1.261). For HI adsorption, the most favored reactions have the following Eads (eV) -1.982, -1.790, -1.683 implying that HI would be more reactive than I2. In addition to FP species, adsorption of H2O was also studied as the hydrated surface can have different reactivity than the bare surface. One thermodynamically favored site for H<sub>2</sub>O adsorption was found with an Eads, eV of -0.754. Finally, the calculations of hydrated surfaces of Na<sub>2</sub>CO<sub>3</sub> show that a layer of water adsorbed on the surface significantly reduces its affinity for iodine (Eads, eV = -1.066). According to the thermodynamic model built, the required partial pressure at 373 K to have adsorption of the first layer of iodine is  $4.57 \times 10^{-4}$  bar. The second layer will be adsorbed at partial pressures higher than  $8.56 \times 10^{-6}$  bar; a layer of water on the surface will increase these pressure almost ten folds to  $3.71 \times 10^{-3}$  bar. The surface interacts with elemental Cs with an Eads (eV) of -1.60, while interacts even strongly with CsI with an Eads (eV) of -2.39. More results on the interactions between Na<sub>2</sub>CO<sub>3</sub> (001) and cesium-based FP will also be presented in this paper. Keywords : iodine uptake, sodium carbonate surface, sodium-cooled fast nuclear reactor, DFT calculations, fission products Conference Title : ICTCMME 2018 : International Conference on Theoretical Chemistry, Modeling and Molecular

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