Molecular Dynamics Simulation of Oxygen-containing NaCl-KCl-UCl_3 Molten Salt

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1. Introduction

Molten Salt Reactors(MSRs) are designed to operate under natural circulation conditions of the fuel salt. The thermophysical properties of the fuel salt (e.g., density and viscosity) are therefore critical parameters to ensure stable circulation. However, during operation, insoluble fission products or impurities dissolved from structural alloys are generated. Furthermore, oxygen may remain in fuel salt after salt purification, and it can also ingress from external sources. Because quantitative data on oxygen effects are scarce, we performed molecular dynamics (MD) simulations to assess their impact.

While the necessity of calculating molten salt properties is becoming more important, universally used simulation tools like classical molecular dynamics have limitations related to atomic potential development when it includes impurities. Therefore, we employed Matlantis [1] to simulate oxygen-containing fuel salts, that based on machine-learning interatomic potentials.

2. Methods and Results

2.1. Simulation Details

In this work, we analyzed NaCl-KCl-UCl_3 molten salt with oxygen contents between 0 and 4 wt%. Initial ionic configurations were generated with PACKMOL. Molecular dynamics (MD) simulations were performed in NPT ensemble at 823 K Nosé–Hoover thermostat using 1 fs timestep until the bulk density converged.

2.2. Results

The density of oxygen-containing NaCl-KCl-UCl_3 melts decreases as the fraction of oxygen increases, as shown in Fig. 1. The degree of density decrease is larger than expected, we therefore investigated structural changes via cluster analysis.

In molten NaCl-UCl_3, uranium-chloride network formation has been widely reported [2][3]. Cl^- anions coordinate to U^{3+} cations, and then U^{3+} cations form linkages by sharing Cl^- anions. Isolated U^{3+} cations with no sharing to other uranium classified as monomer, and dimer, trimer, tetramer and polymer mean that structures contain 2, 3, 4, 5 or more U^{3+} cations. In our systems, we consider that both chloride (Cl^-) and oxide (O^{2-}) can coordinate to U^{3+}, and act as bridging ligands.

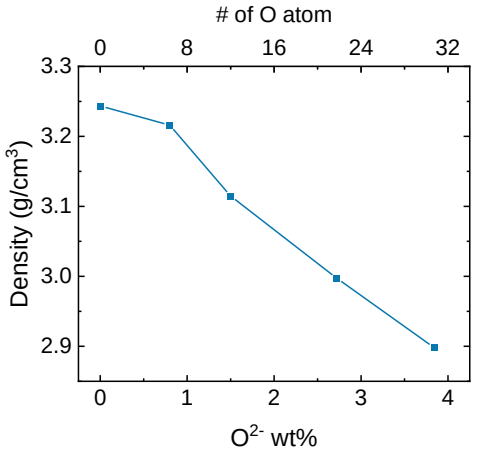

Fig. 1. Density of NaCl-KCl-UCl_3 as a function of oxygen content (wt%).

Fig. 2 shows the fraction changes of uranium networks with increasing oxygen content (wt%); polymer decreases, whereas monomer, dimer, and trimer increase. In other words, oxygen ingress led to depolymerization, and this causes volume expansion, and consequently, the density decreases.

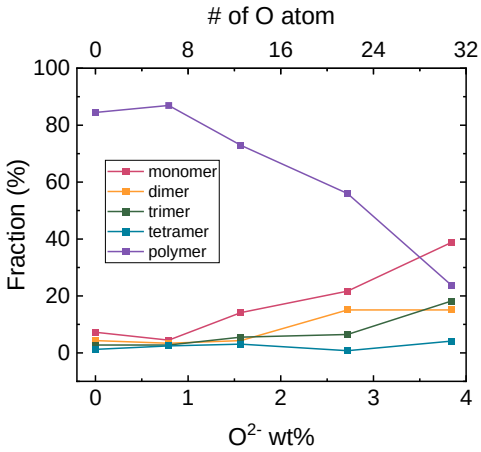

Fig. 2. Fraction of U monomer, dimer, trimer, tetramer, and polymer at different wt% of oxygen.

3. Conclusion

Thermophysical properties are key to MSR performance, yet impurities introduced during reactor operation–fission products, moisture, and oxygen–can modify them.

Our simulations of oxygen-containing NaCl-KCl-UCl_3 show that even small amount of oxygen additions alter U-(Cl/O) connectivity, driving depolymerization and a measurable reduction in density.

We plan to quantify the link between oxygen content, network depolymerization, and density using radial distribution functions (RDFs) and cluster-structure analyses.

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