



Experimental solubility measurements of lanthanides in liquid alkalis



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HIGHLIGHTS

- Measurements of solubility of Lanthanides in liquid sodium, cesium and sodium-cesium mixtures.
- The solubility of Nd, La, and Ce in liquid cesium is much higher than that in liquid sodium at the same temperature.
- The solubility in sodium-cesium mixtures decreases with sodium fraction increasing.
- The study prove the “liquid-like” lanthanide transport in metallic fuel during operation.

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ABSTRACT

In metallic nuclear fuel, lanthanide fission products play a crucial role in the fuel burnup-limiting phenomena of fuel cladding-chemical interaction (FCCI). The lanthanides have been hypothesized to transport by a ‘liquid-like’ mechanism out of the metallic fuel to the fuel peripheral to cause FCCI. By liquid fission product cesium and liquid bond sodium, the lanthanides are transported to the peripheral of the fuel through the porosity of the fuel. This work investigates the interaction between the lanthanides and the alkali metals by experimentally measuring the solubility of lanthanides within liquid sodium, and neodymium in liquid cesium and mixtures of cesium and sodium. The temperature dependence of the solubility is experimentally determined within an inert environment. In addition, the dependence of the solubility on the alkali metal concentration in liquid mixtures of cesium and sodium was examined. In quantifying the solubility, the fundamental understanding of this transport mechanism can be better determined.

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1. Introduction

With the emphasis from the Generation IV Forum, there has recently been a renewed interest in sodium-cooled reactors [1]. One of the current challenges facing sodium-cooled reactors is increasing the burnup of metallic nuclear fuel U-Zr, even though EBR-II Mark III fuel has been qualified for 10% heavy metal burnup [2]. A major concern for achieving higher burnup is the phenomenon of Fuel-Cladding Chemical Interaction (FCCI) [3]. In the operation of metallic fuel, swelling occurs which can cause the fuel to contact the cladding and resulting in intermetallic compounds to form between the cladding and the fuel. These intermetallic compounds effectively reduce cladding performance, causing reduced

lifetime of the cladding and the metallic fuel. When this interaction between fuel and cladding involves fission product lanthanides, FCCI and the degradation of the cladding is accelerated [3]. Thus the migration of the fission product lanthanides to the fuel peripheral has a vital role in phenomena of FCCI.

A proposed mechanism of the fission product lanthanides migration to the fuel peripheral is a ‘liquid-like’ transport phenomenon [4]. In this mechanism the fission product lanthanides transport is aided by liquid cesium and liquid sodium through the porosity of the fuel. The theory is based on the fact lanthanide fission products have a high solubility in liquid cesium while having a low solubility in liquid sodium. It is assumed the porosity of the fuel in operation, including the cracks due to fuel swelling, is filled with liquid cesium. Therefore, the fission products produced dissolve into the liquid cesium and diffuse to the peripheral of the fuel at the bond-sodium interface due to the temperature gradient

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present radially in the fuel. The fission products then precipitate out of the liquid due to their low solubility in liquid sodium. The “liquid-like” transport theory can account for most of the lanthanide transport phenomena identified by post-analysis of irradiated metallic fuel [4].

This work examines the mechanism of ‘liquid-like’ transport of the lanthanides by experimentally studying the solubility of lanthanides in liquid sodium and liquid cesium, and the data gathered supports modeling efforts [5] [6].

2. Materials and experimental procedure

Metal pieces of cerium, praseodymium, and neodymium were acquired from ESPI Metals with a purity of 99.9%. The sodium metal was from ESPI Metals with a purity of 99.95% while the cesium metal was from Alfa Aesar with a purity of 99.98%. These materials were used to measure the solubility using an inversion crucible method [7]. The tests were conducted within an argon-filled glovebox that had an O₂ and a H₂O concentration below 3 ppm.

Crucibles of stainless steel 304 or tantalum metal were used for experimentation. These cylindrical crucibles had a cap placed on each end of the cylindrical tube and at the center of the tube was a metal mesh, with a mesh of 300- μ m. Approximately 2 g of solid alkali metal and a 2 g solid piece of lanthanide were placed within one of the crucibles’ caps. The crucible was placed vertically, as shown in the left side of Fig. 1, within a furnace with the cap containing the alkali metal and lanthanide solid at the bottom of the crucible.

The furnace was set to the temperature of interest, and heated for an equilibration time to allow the solution of liquid alkali metal and lanthanide solid to reach equilibrium. At the conclusion of the equilibration time the crucible was inverted. This caused the liquid alkali metal to flow through a 300- μ m mesh filter as it passed to the opposite end of the crucible. This mesh filter prevented the solid lanthanide piece from reaching the opposite cap, resulting in an alkali metal sample with the equilibrium concentration of lanthanide at this cap. The right side of Fig. 1 shows a schematic of the inside of the crucible after inversion. The crucible was cooled to allow the alkali metal to solidify and the cap with the alkali sample was subsequently removed from the crucible. The alkali sample in the cap was removed from the glovebox; the alkali metal was then neutralized in dissolving the metal in isopropyl alcohol. This solution was then diluted with a known quantity of deionized water to form an aqueous solution, with the concentration of lanthanides to

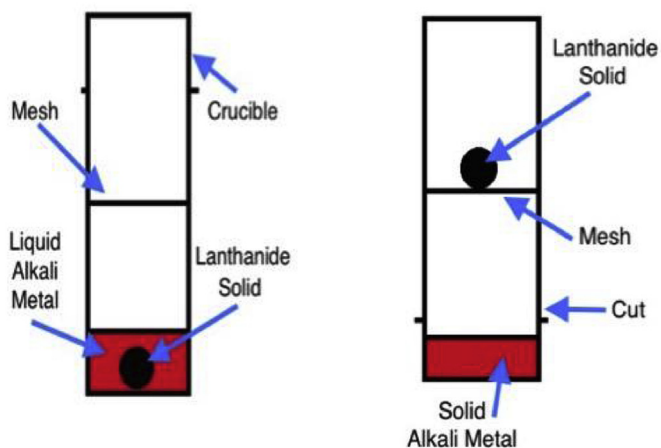


Fig. 1. A schematic of the inside of the experimental crucible. (Left: Before testing, Right: After inversion).

be measured in Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS) and the solubility calculated from this measurement.

3. Results

The time dependence of the lanthanide concentration in the alkali metal was studied to determine the equilibration time, how long the test should be heated to reach equilibrium. Then the solubility of lanthanides in liquid sodium was investigated for the temperature range from 450 °C to 550 °C while liquid cesium was investigated from 200 °C to 450 °C. Lastly the solubility of neodymium in mixtures of liquid cesium and liquid sodium was investigated at 450 °C. All the solubility data measured are given in the Appendix.

3.1. Time dependence

At 200 °C the concentration of neodymium in liquid cesium was measured as a function of heating time to assess the rate of equilibration if not rapid. Fig. 2 shows the time dependence of the concentration of neodymium in liquid cesium. In addition to the tests conducted, Fig. 2 also shows the impurity concentration of neodymium in the cesium metal received from the manufacturer. These impurity concentrations were the measurements we made of the material received from the companies, not from the analysis from the vendor. As soon as the metal was received we made samples and performed ICP-MS on the metal for the Ln concentrations.

Fig. 2 shows the concentration of neodymium when heating the solution over the time frame is a constant value of 1.7×10^{-5} at. %. This constant concentration shows the solubility of the neodymium in liquid cesium was reached prior to the earliest time measured of 1 h and the equilibration is rapid at 200 °C. Analysis of neodymium in liquid sodium at 450 °C showed similar results of the solubility concentration being reached prior to 1 h. To ensure equilibrium in the testing of the temperature dependence and the alkali concentration dependence, a time of 24 h was used for the heating in other tests.

3.2. Temperature dependence

Shown in Fig. 3 is the experimental data of the temperature dependence of the solubility of lanthanides in liquid cesium and liquid sodium. Also included are the average concentrations and standard deviation for the tests that were used to investigate the time dependence in liquid cesium and in liquid sodium.

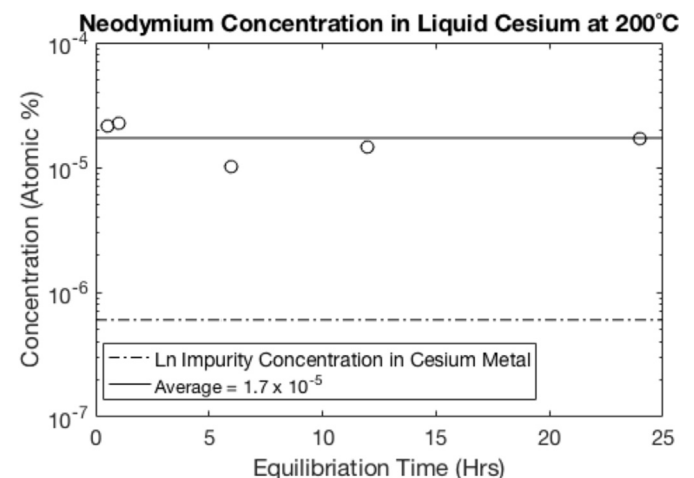


Fig. 2. Time dependence of the concentration of neodymium in liquid cesium.

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