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An integrated model of tritium transport and corrosion in Fluoride Salt-Cooled High-Temperature Reactors (FHRs) – Part I: Theory and benchmarking

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HIGHLIGHTS

- A model was developed for use with FHRs and benchmarked with experimental data.
- Model results match results of tritium diffusion experiments.
- Corrosion simulations show reasonable agreement with molten salt loop experiments.
- This is the only existing model of tritium transport and corrosion in FHRs.
- Model enables proposing and evaluating tritium control options in FHRs.

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ABSTRACT

The Fluoride Salt-Cooled High-Temperature Reactor (FHR) is a pebble bed nuclear reactor concept cooled by a liquid fluoride salt known as "flibe" (^{7}LiF-BeF$_2$). A model of TRITium Diffusion EvolutioN and Transport (TRIDENT) was developed for use with FHRs and benchmarked with experimental data. TRIDENT is the first model to integrate the effects of tritium production in the salt via neutron transmutation, with the effects of the chemical redox potential, tritium mass transfer, tritium diffusion through pipe walls, tritium uptake by graphite, selective chromium attack by tritium fluoride, and corrosion product mass transfer. While data from a forced-convection polythermal loop of molten salt containing tritium did not exist for comparison, TRIDENT calculations were compared to data from static salt diffusion tests in flibe and flinak (0.465LiF-0.115NaF-0.42KF) salts. In each case, TRIDENT matched the transient and steady-state behavior of these tritium diffusion experiments. The corrosion model in TRIDENT was compared against the natural convection flow-loop experiments at the Oak Ridge National Laboratory (ORNL) from the 1960s and early 1970s which used Molten Salt Reactor Experiment (MSRE) fuel-salt containing UF_4. Despite the lack of data required by TRIDENT for modeling the loops, some reasonable results were obtained. The TRIDENT corrosion rates follow the experimentally observed dependence on the square root of the product of the chromium solid-state diffusion coefficient with time. Additionally the TRIDENT model predicts mass transfer of corrosion products from the hot to the cold leg (as was observed in the experiments with salts containing UF_4). In a separate paper the results of TRIDENT simulations in a prototypical FHR are presented.

1. Introduction

A unique model of TRItium Diffusion Evolution and Transport (TRIDENT), the first such model to integrate the effects of tritium transport, reactor coolant chemistry, and corrosion phenomena, was developed and benchmarked (Stempien, 2015). This paper highlights the important dependencies captured in TRIDENT, and reports the results of benchmarking comparisons.

Forsberg et al. have proposed a new type of reactor: one which combines the graphite-matrix, coated-particle fuel developed for gas-cooled reactors with the fluoride salt coolant used in the 1960s Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) (Forsberg et al., 2003). Originally called the Advanced High-Temperature Reactor (AHTR), this

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concept is now known as the fluoride salt-cooled high-temperature reactor (FHR). Interest from the U.S. Department of Energy (DOE) has enabled universities and national laboratories to engage in research and development related to FHRs (Forsberg et al., 2013; Holcomb et al., 2013). What distinguishes an FHR from a molten salt reactor (MSR) is that an MSR dissolves the fuel directly in the coolant, but an FHR utilizes a solid fuel with a clean (un-fueled) coolant-salt. The baseline FHR coolant is a lithium-fluoride beryllium-fluoride salt called flibe (0.67 mol fraction LiF – 0.33 mol fraction BeF_2) which utilizes lithium enriched in the Li-7 isotope. Flibe melts at 459 °C and boils at 1430 °C.

There are a number of potential core configurations and fuel types that might be compatible with an FHR, but the baseline commercial design (called the Mk1PB-FHR) utilizes a pebble bed core with online refueling (Andreades et al., 2014). While the FHR has numerous potential capabilities, configurations, and applications, in order to commercialize it, a compelling economic and technical case must be made if FHRs are to become viable candidates for next-generation power systems. The current commercial case for the FHR is that it enables a nuclear and renewable electric grid and increases revenue by 50% or more compared to base-loaded, light water reactor (LWR) nuclear plants (Forsberg et al., 2013, 2014a). This is a consequence of coupling the reactor to a Nuclear air-Brayton Combined Cycle (NACC) that enables the production of base-load electricity and peak electricity using auxiliary natural gas, stored heat, or hydrogen. The open-air NACC power cycle is similar to that used in natural gas combined-cycle plants. This specific design is based on the GE 7FB gas turbine. NACC heat exchangers have hot coolant salt on one side and compressed air on the other side; thus, any tritium diffusing through the heat exchanger will exit via the gas turbine exhaust. This is in contrast to closed power cycles (steam, supercritical carbon dioxide, etc.) where tritium can be trapped in the power cycle. Economic considerations favor an FHR with NACC but also impose added requirements for tritium control and thus increase the importance of modeling tritium behavior.

The FHR possesses a number of appealing characteristics including coolant outlet temperatures of at least 700 °C and a number of inherent and engineered passive safety features. Major inherent safety features are due to the design of the coated particle fuel, which is stable to greater than 1600 °C, and the high boiling point of the coolant, which exceeds 1400 °C. These properties would enable the FHR to operate at atmospheric pressure with hundreds of degrees of margin to fuel damage and coolant boiling (Forsberg et al., 2014b). Major engineered passive safety features include the use of a pool-type vessel and a natural circulation decay heat removal system activated passively by a fluidic diode.

No FHR has ever been built, and uncertainties will need to be addressed before either a test or commercial reactor can be constructed. The MSRE operated at ORNL in the 1960s using uranium fuel dissolved in the LiF-BeF_2 coolant (Haubenreich and Engel, 1970). The issue of tritium production in this coolant was only realized at the end of the MSRE program. Tritium production rates, tritium release rates, and tritium distribution throughout an FHR are currently unknown. Corrosion rates are uncertain because they couple to tritium behavior (Calderoni et al., 2009) and are sensitive to the presence of impurities within the coolant (Keiser et al., 1977). In order to address these uncertainties, a unique model of TRItium Diffusion Evolution and Transport (TRIDENT) integrating the effects of tritium transport, reactor coolant chemistry, and corrosion phenomena was developed and benchmarked (Stempien, 2015). This paper highlights the important dependencies captured in TRIDENT and reports the results of benchmarking comparisons. Results from TRIDENT simulations of a prototypical pebble-bed FHR are presented in a separate article.

2. TRIDENT reactor system model description

A high-level flow chart for the TRIDENT model is given in Fig. 1. Tritium is born in the coolant as dissolved TF (see Section 3). Depending on the chemical redox condition of the coolant (defined by the fluorine potential described in Section 4) tritium will speciate into both TF and T_2 via chemical reactions. The fluorine potential in the coolant determines the thermodynamic driving force for corrosion and the relative amounts of TF and T_2 in the system. Both TF and T_2 can be adsorbed on graphite in the core depending on the mass transport parameters and graphite capacity for tritium. In FHRs, tritium behavior is intimately linked with corrosion (see Sections 4 and 6). Tritium born in the coolant as TF can corrode structural metals, and a byproduct of these reactions is $T_{2(g)}$. If corrosion products are deposited/precipitated in the system, the reaction is the reverse of the corrosion reaction, and TF is generated as T_2 is consumed. Corrosion control is based on mitigating selective chromium attack by TF, and the radiological management of tritium is based on preventing unwanted diffusion/migration of T_2 through system boundaries. TRIDENT couples the redox potential to TF/T_2 speciation, allowing it to realistically predict tritium buildup in the reactor, relevant corrosion reactions, tritium trapping on graphite, and tritium diffusion through system barriers. If desired, TRIDENT can simulate different engineered mechanisms for TF and T_2 removal from the coolant (such as gas-stripping, permeation windows, sorption on beds of carbon, etc. discussed in a separate paper). Only T_2 can diffuse through metal. Thus, only T_2 is allowed to diffuse out of the primary system through the heat exchangers in the model. TRIDENT is capable of simulating single loop reactors or two-loop reactors having both a primary and secondary system. Any tritium not escaping through the heat exchangers or not removed by engineered systems is then returned to the core through the core coolant inlet.

3. Tritium production in FHRs

Any beryllium or lithium-based salt reactor coolant will generate tritium due to neutron transmutation. Eqs. (1)–(5) show the major pathways for tritium production in flibe. Although the baseline FHR coolant is flibe enriched to 99.995 wt% in Li-7, the

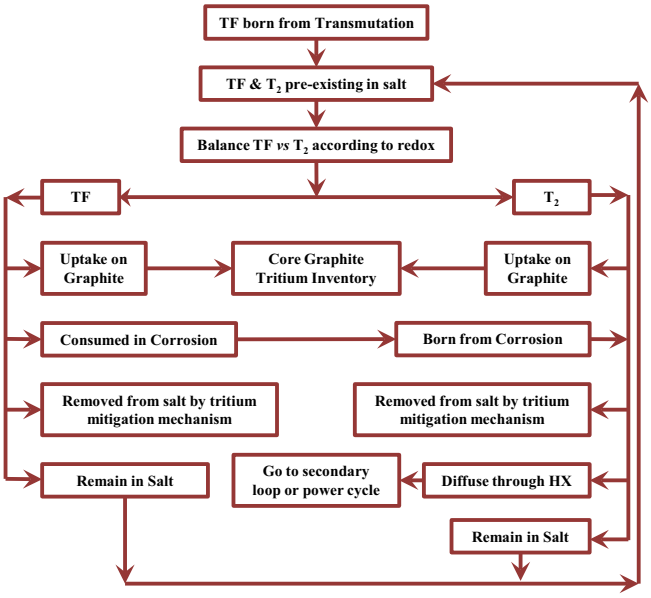

Fig. 1. TRIDENT model flow chart.