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The study of cesium ionization at high gas temperature in a primary circuit

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HIGHLIGHTS

- A model of cesium ionization at high temperature is presented.
- The kinetics of ions is combined with neutral cesium-iodine chemistry.
- The formation of ions is driven by cesium thermal ionization and irradiation of a steam.
- At high temperature ion formation is dominated by cesium atoms thermal ionization.
- The insight on more realistic source term estimation is given.

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ABSTRACT

A kinetic model has been presented in developing a better understanding of the mechanism behind the ions formation at high gas temperature in a primary circuit in conditions of severe accidents. The formation of ions is triggered by the thermal ionization of cesium atoms and irradiation of a hot steam. The kinetics of ions is considered in combination with the neutral chemistry of cesium and iodine species. Modeling results are presented for temperatures 2000 and 1200 K and a dose rate of 1000 kGy/s. The evolution of the concentrations of the most abundant ions and neutral reaction products are calculated.

1. Introduction

The release of radioactivity outside a nuclear power station into environment is a main hazard of severe accidents with a nuclear reactor. The main sources of radioactivity are fission products (FPs), such as cesium and iodine. As the water rises through the core and evaporates during the dryout phase of the LWR accident, the steam is formed. Since the fuel temperature rises, fission products are released from the fuel and are then transported by the steam flow through the reactor core and primary circuit. Gas temperature gets higher in the reactor vessel upper plenum and in the adjacent part of the primary loop. During the main stage of the fission product release, temperatures are around 2500 K at the core outlet, 2000–1500 K in the upper plenum and near 700 K in the cold legs of the primary circuit (Sehgal, 2012). As a result, the most of FPs and structural materials emitted as vapors from the degraded core will be under high supersaturation levels when they are transported through the reactor coolant system (RCS). The supersaturated vapors will contribute to the formation and growth of aerosols via homogeneous (at once from the precursor vapors) and heterogeneous (e.g. on ions) nucleation. Further growth of formed clusters depends on their number concentration and the presence of condensable vapors (Williams and Loyalka, 1991; Kissane and Drosik, 2006; Kissane, 2008).

Cesium and iodine are among of the most important FPs because of their high potential radioactivity. In particular, their release is important for evaluation of the source term. The conditions in the RCS favor chemical interactions between different species residing in the gas phase. The formed compounds may have physical properties significantly different from the properties of initial elements. As noted in Sehgal (2012), Cousin et al. (2013), the role of chemistry, including high-temperature radiolysis reactions, is central in determining the transport of the emitted FPs and other materials out of the core and through the primary circuit. Radiolysis effects may have little impact in the core region where temperatures are so high that only simple atomic and radical species would exist. On the contrary, they may be important in cooler

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regions of the RCS where significant deposition of FPs occurs. Therefore, the analysis of radiation effect and evolution of gaseous cesium and iodine should be based on kinetic approach.

Presently, nuclear reactor safety codes do not account for any charged radioactive aerosol effects. Even if it is accepted, that the irradiative ionization creates the regions with high density of ions and highly charged aerosols, there is no consensus on its importance. It is assumed that charge-based effects are negligible, since charge is conducted away from aerosol particles to vapor phase. However, the electric charging process for radioactive aerosols may differ from that of non-radioactive aerosols. It is shown, that specific conditions, such as dilution in the atmosphere or space confinement, may noticeably increase the electrical charge of radioactive aerosols (Clement and Harrison, 2000).

Nucleation on charged clusters is called the ion induced nucleation. The vapor concentrations, required for activation of ions, are generally lower than those for neutral particles. As for now, there is no information of the fraction (if any) of ion-induced particle formation in RCS. This lack of knowledge is due to the incapability to detect or chemically identify the initial clusters (Kissane, 2008). Proposed candidates for initial clusters formation include refractory metal and FP volatile compounds. Aerosol dynamics models used in reactor accident scenarios tend to exclude the effects of radiation except radiolysis in high-radiation field (Cantrel et al., 2013). But charged particle interactions may affect the transport of radioactive particles away from the source. Aerosols also become charged due to thermal or chemical ionization. Therefore additional investigations are needed to clarify the importance of these phenomena.

Ionization in the thermal plasma is caused by collisions between gas particles. Easily ionizable atoms of alkali metals are the basic suppliers of free electrons and positive ions. Cesium is one of the main fission products released in the case of a core meltdown accident of a nuclear reactor. The ionization of Cs into Cs^+ liberates electrons at high temperature. Fission gas contains solid or liquid particles as a result of the volume condensation. The interaction between phases leads to the charging of particles and changes gas ionization degree. Cesium can rapidly react with other fission products or structural core materials. As the temperature decreases, the great affinity of Cs for steam leads to the production of CsOH in liquid and gas phases. Hence, we should focus more attention on the influence of high temperature on cesium ionization.

The main objective of Phebus integral experiments (Kissane and Drosik, 2006) was to determine all significant phenomena during the FPs transport in RCS in conditions closed to LWR severe accidents. However, additional experimental results are required to develop more adequate and detailed models, especially for iodine chemistry at low temperature and cesium retention at high temperature. Calculations with basic models of aerosol formation and deposition (Kissane, 2008; Allelein et al., 2009) strongly underestimate cesium deposits in the hot vertical line for FPT1 test (Kissane and Drosik, 2006). Several hypotheses were tested. The suggestion of Cousin et al. (2013) is to use the underestimated Nusselt number for coolant laminar flow in the upper plenum with an irregular geometry.

Thermodynamic and kinetic models for the I, Cs, Mo, B, O, H chemical system were developed and implemented in the SOPHAEROS module in the code ASTEC. These developments highlight the role of kinetic limitations in formation of gaseous iodine in the RCS. These limitations were tested in Phebus FPT3 test where RCS chemistry was influenced by the degradation of a boron carbide control rod.

However, proper understanding of severe accident phenomena is still very important, that was recently justified regarding the consequence of the severe accidents in Japan. So it is necessary to further reduce the uncertainties of simulation results and to investigate new phenomena not regarded in detail before. To be sure that the research conducted on severe accidents is efficient and focusing on relevant topics, the Severe Accident Research Priority (SARP) work was started in the first SARNET FP6 project. The concluding results of the work in the SARP group at the end of the SARNET2 FP7 activity were summarized in Klein-Hesling et al. (2014). The defined ranking table includes the level of safety importance and the knowledge level, based on the consensus found in the SARP group. The issue 4.4 of the table in Klein-Hesling et al. (2014) concerns the high temperature chemistry in the RCS and its impact on the source term to the environment. The prediction of the FP species exiting the RCS should be improved to provide the best estimate of FP source into the containment. The 4.5 issue concerns the chemistry of FP in the containment and its impact on the source term to the environment. Both issues are classified with a high priority.

In this work we consider a possibility of cesium atoms thermal ionization followed by vapor condensation on formed ions. No documented studies have examined the thermal ionization effect in comparison with the gas radiolysis in reactor accident scenarios. Moreover it was previously assumed that ionizing radiation leads to charge neutralization. The significance of the suggested mechanism is evaluated for conditions of the PHEBUS FTP1 experimental test. We take into account the gas-phase ion kinetics and neutral cesium-iodine chemistry (Xerry et al., 2012). The analysis is done in a box approach as a first step to estimate the phenomenon importance. Neither of existing codes considers this effect in application to FP behavior in conditions of severe accidents. However, in the source at FPs release during, for example, a LOCA this phenomenon certainly takes place. In spite of its short timescale of few milliseconds, it can modify the cesium speciation and hence the cesium deposition at high gas temperature. Thus, the accurate modeling of this phenomenon is needed in order to reduce uncertainty in evaluations of high-temperature chemistry of fission products.

The layout of this paper is as follows: in Section 2 we discuss equilibrium ionization of cesium atoms in a hot steam which serve mainly as a thermal bath. Section 3 describes estimations for vapor condensation on ions formed. In Section 4 the kinetic model is presented with some results of the transient analysis in Section 5. These results are discussed in the Section 6.

2. Thermal ionization of cesium atoms

Above a temperature of about 1500 K the evaporation of fission products is complete. In mixture of hydrogen and steam the predominant cesium containing gaseous species are CsOH, CsI and Cs. In a steam dominating atmosphere the Cs form is primarily CsOH. For iodine the predominant chemical forms are I, I_2, HI and CsI. At high temperature the ionization of an atom by electron impact is a basic process (Smirnov, 2001; Sodha, 2014)

$$e + A = 2e + A^+$$

In this process the incident electron interacts with a valence electron, transfers to it a part of its kinetic energy and causes the detachment of the valence electron from the initially neutral atom.

The alkali metal atoms like cesium and sodium have a low ionization potential. In the core and upper plenum the temperature is around 2000–2500 K. At such temperatures, the ionization of the carrier gas (steam) is low. Hence it is natural to suppose the applicability of the Saha equation to thermal plasma which is a heated steam containing positive ions Cs^+, some negative ions and free electrons.

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