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Theoretical assessment of particle generation from sodium pool fires

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HIGHLIGHTS

- Development of particle generation model for sodium-oxides aerosol formation.
- Development of partially validated numerical simulations to build up maps of saturation ratio.
- Nucleation of supersaturated vapours as relevant source of aerosols over sodium pools.
- Prediction of high concentrations of primary particles in the combustion zone.

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ABSTRACT

Potential sodium discharge in the containment during postulated Beyond Design Basis Accidents (BDBAs) in Sodium-cooled Fast Reactors (SFRs) would have major consequences for accident development in terms of energetics and source term. In the containment, sodium vaporization and subsequent oxidation would result in supersaturated oxide vapours that would undergo rapid nucleation creating toxic aerosols. Therefore, modelling this vapour nucleation is essential to proper source term assessment in SFRs. In the frame of the EU-JASMIN project, a particle generation model to calculate the particle generation rate and their primary size during an in-containment sodium pool fire has been developed. Based on a suite of individual models for sodium vaporization, oxygen natural circulation (3D modelling), sodium-oxygen chemical reactions, sodium-oxides-vapour nucleation and condensation, its consistency has been partially validated by comparing with available experimental data. As an outcome, large temperature and vapour concentration gradients set over the sodium pool have been found which result in large particle concentrations in the close vicinity of the pool.

1. Introduction

Generation IV (Gen. IV) designs are striving to reach the highest safety standards ever applied to Nuclear Power Plants (NPPs). Accidents with significant fuel damage (i.e., severe accidents) are being considered in the design phase so that they can be inherently avoided or prevented and/or mitigated by passive design features or by ad-hoc engineering safeguards with elimination of the need for offsite emergency response (GIF, 2002). This enlargement of design scope with respect to Gen. II and Gen. III reactors is a challenge for all the technologies involved in the systems chosen for Gen. IV since accident scenarios involve complex phenomena that have to be taken into account when demonstrating reactor safety. Sodium-cooled Fast Reactor (SFR) technology is more mature than other Gen. IV designs since several SFR reactors have been operated (i.e., BN-600, Phénix, Joyo, FBTR, etc.) and, as a result, the experience that has been gained currently amounts to >400 operational years. Nonetheless, in Europe, if France chooses to pursue SFR development, the new technologies to be used as well as higher safety standards to be put in place will require the construction of an "industrial demonstrator", called ASTRID, which might start operation around 2030 (CEA, 2015).

In SFR systems, sodium (Na) is used as a coolant due to its excellent properties. Its high thermal conductivity and low viscosity make it a good thermal-energy transport fluid with a broad temperature interval as a liquid (370–1156 K at atmospheric pressure) allowing its use as coolant in low pressure conditions close to the ambient conditions. In addition, it is also suitable for the fast-neutron economy since its absorption and scattering cross sections are very low; finally, its compatibility with structural materials is quite good (in comparison with heavy liquid metals like lead, lead-bismuth eutectic, …). However, despite all these advantages,

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Na shows a high chemical reactivity with oxygen, and in a less extent water, which entails a fire hazard risk in case of leaks into the containment.

In SFR safety studies the highly-unlikely event of a core disruptive accident must be considered. In accident conditions, core disruption by supercriticality could involve energetic destruction of fuel assemblies. Then, if this is assumed, the interaction between hot fuel and liquid sodium could lead to a vapour explosion which could create a breach in the primary system and contaminated liquid sodium at high temperature would be ejected into the containment. In these conditions, the vaporization of sodium and its possible instantaneous combustion with the oxygen available would result in the generation of supersaturated Na-oxide (Na_xO_y) vapours likely to nucleate and form suspended particles. The aerosols formed could act as a radioactive species carrier; this together with the potential harm associated with the chemical species resulting from the sodium oxides reaction with water vapour present in the atmosphere (sodium hydroxide aerosols) would be responsible to a great extent for the radiological and chemical impact of any potential source term. In addition, if the leak is major, the heat released during the combustion is substantial.

Several sodium fire studies (pool and spray fires) have been carried out since the 1970s, in facilities like PLUTON (Lhiaubet et al., 1990) and JUPITER (Malet et al., 1990) in France, CSTF in the USA (Hilliard et al., 1977, 1979; McCormack et al., 1978 and Souto et al., 1994), FAUNA in Germany (Cherdron and Jordan, 1980, 1983; Cherdron et al., 1985, 1990) or ATF in India (Subramanian and Baskaran, 2007; Subramanian et al., 2009; Baskaran et al., 2011). In all of them, the aim was to understand the sodium burning process and to study the sodium compound aerosols behaviour and their chemical composition. Since then a number of studies have intended to model the whole scenario with relative success.

The pioneering work by Beiriger et al. (1973), who developed a tool mainly focused on fire energetics (the SOFIRE code), was followed by others who made specific hypotheses and approximations. In the SOFIRE model, the formation of a thin vapour layer over the surface of the pool is not considered and the Na-O_2 reactions take place on the pool surface. Through an extensive review of the physico-chemical processes involved in the liquid sodium pool combustion, Newman (1983) established that pool combustion could occur via surface or gas-phase reaction depending on the ignition temperature. Based on this work, Sagae and Suzuoki (1985) developed an analytical combustion model in which sodium vapour diffuses from the pool surface to a flat flame where the Na-O_2 reactions take place. In this model the combustion rate depends on heat and mass transfer of Na and O_2 to the flame (flame sheet approach). The vapour-phase sodium-combustion model has been included in numerous combustion codes as the SPM code by Miyake et al. (1991), SOPA code developed by Lee and Choi (1997) or more recently in multi-dimensional numerical analysis codes such as the SPHINCS code (Yamaguchi and Tajima, 2003a) or AQUA-SF code (Takata et al., 2003).

However, as said above, all these computer codes have mostly focused on energetics and thermal-hydraulics (Murata et al., 1993) so that particle modelling has received much less attention. For example: the SOFIRE code (Beiriger et al., 1973) assumes that all Na_xO_y produced becomes particles of a given size with no consideration of particle formation kinetics or primary-particle size; the CONTAIN-LMR code (Murata et al., 1993) requires the user to fix the initial particle size in the analysis and this becomes the size at which any particle is nucleated.

This paper presents the main features and bases of a Particle Generation (PG) model from sodium pool fires whose main output variables are particle generation rate and size. This consists of a suite of individual models of the following phenomena involved: Na vaporization (diffusion layer approach), O_2 transport by air natural circulation (3D flow pattern modelling), Na-O_2 chemical reactions (instantaneous reactions and energy input) and vapour-to-particle conversion (Na_xO_y vapours nucleation by Classical Nucleation Theory and condensation by kinetic theory of gases). Although direct model validation is not feasible due to the lack of experimental data on Na_xO_y vapour nucleation, a partial model validation of sodium vaporization, air convection and sodium reactions has been carried out against available experimental data.

Consequently, the scope of the presented model is the estimation of the particle generation by homogeneous and heterogeneous nucleation during an in-containment sodium pool fire in the near field of the pool. The main output of the model is the number concentration and size of the primary particles resulting from the Na pool combustion. Therefore, any further consideration of particles growth by agglomeration, chemical reaction with steam and/or other gases (like CO_2) and transport to other regions in the far-field of the pool, are out of the scope of the present paper. Any of these additional phenomena would be essential for a full prediction of in-containment source term behaviour and, as such, they would be encapsulated in specific models in a lumped parameter code addressing SFR severe accidents, in which the present work would be embedded as a particle source into the containment atmosphere.

This work has been mostly carried out in the frame of the 7th Framework Programme of the European Commission via the JASMIN project.

2. Conceptual scenario

Liquid-sodium-pool combustion can proceed via surface reaction or gas-phase reaction depending on the initial pool temperature. Newman (1983) showed that when the pool temperature is lower than the ignition one (623–723 K), surface reaction dominates, forming a grey-purple product close to the sodium surface that acts as a floating layer on the surface of the liquid sodium without any combustion; however, for sodium pool temperatures higher than the ignition one, this grey layer wets or decomposes allowing Na vapour to vaporize. Assuming instantaneous combustion, this would occur in the gas phase in a layer called the flame region located close to the pool; it is in this region where the sodium oxide vapours would form, nucleate and create particle growth.

The gas-phase reaction region is sketched in Fig. 1, in which the Na pool is displayed as a hot flat plate. As observed, the pool nearby can be split into three layers: sodium vapour, right over the pool; a flame region, above and adjacent to the Na layer; and the bulk gas overlying the flame and feeding the flame region with oxygen by natural convection (Miyake et al., 1991; Newman and Payne, 1978; Newman, 1983).

At steady-state burning, the pool does not reach its boiling point but remains close to the commonly found equilibrium pool temperature of 1000–1010 K (Newman and Payne, 1978). On the other side, vapour pressure of sodium is approximately 1 kPa at 800 K and very nearly 20 kPa at 1000 K. In these conditions, sodium readily evaporates and this vapour will be transported by diffusion to the flame region in which Na vapour reacts with the available oxygen forming Na-oxides and making the local temperature rise to 1200–1300 K. Namely, sodium oxidation is sustained by Na diffusion and by O_2 brought to the region by turbulent convective flows in the gas over the pool surface. As sodium vapour supply by evaporation becomes relatively large and chemical reactions are known to be fast, sodium combustion will be governed by Na vapour supply by evaporation and oxygen transport into the reaction region. In the sodium vapour layer, a vertical laminar flux will exist from the sodium pool surface to the flame region,

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