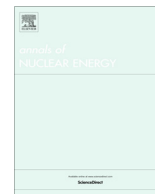




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## Experimental investigations on carbonation of sodium aerosol generated from sodium fire in the context of fast reactor safety

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## ABSTRACT

Carbonation of sodium aerosols is the most important aspects to be considered for the evaluation of chemical hazards as a part of fast reactor safety studies. The sodium oxide, immediately formed as the combustion product due to sodium fire, undergoes chemical changes to NaOH, Na<sub>2</sub>CO<sub>3</sub> and NaHCO<sub>3</sub> upon reactions with moisture and CO<sub>2</sub> prevailed in the atmosphere. Of which, hydroxide aerosols are highly corrosive and harmful, and it has stringent concentration limit for human exposure. Hence, in order to assess the condition for human intervention in the event of sodium fire, chemical composition of aerosols resulting from controlled sodium fires in a closed Aerosol Test Facility was investigated. The real time chemical species of aerosols generated from sodium fire and the effect of relative humidity (RH) and carbon dioxide concentration in air on carbonation have been studied. The experiments were carried out with the initial mass concentration of ~4 g m<sup>-3</sup>, RH between 20% and 90% and the CO<sub>2</sub> concentration in surrounding environment at 390 and 280 ppm. It is observed from the experimental study that aerosols are enriched with NaOH (0.8 mol fraction) in the beginning stage (samples collected during first few minutes after sodium fire) when surrounding atmosphere contains any of the following compositions – (i) ~90% RH and 390 ppm CO<sub>2</sub>, (ii) ~90% RH and 280 ppm CO<sub>2</sub> or (iii) 50% RH and 280 ppm CO<sub>2</sub> whereas they are almost equally distributed between NaOH and Na<sub>2</sub>CO<sub>3</sub> in the beginning stage when the atmosphere has any of the compositions (i) 50% RH, 390 ppm CO<sub>2</sub>, (ii) 20% RH, 390 ppm CO<sub>2</sub> or (iii) 20% RH, 280 ppm CO<sub>2</sub>. Carbonation of aerosols is completed between 20 min and 1 h just after sodium fire depending upon the prevailing atmosphere. The present study shows that highly humid condition promotes carbonation process. Faster the carbonation process the lesser would be the chemical hazard.

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### 1. Introduction

In sodium cooled fast reactor (SFR), leakage of hot sodium in the secondary heat transfer loop inside the Steam Generator Building (SGB), results in sodium fires of different kinds such as pool fire, spray fire and column fire (Cherdron and Jordan, 1982). The resulting sodium fire generates a lot of sodium aerosols, which mainly consist of oxides of sodium viz; Na<sub>2</sub>O, Na<sub>2</sub>O<sub>2</sub> and NaO<sub>2</sub> depending on the abundance of sodium and oxygen content at the time of reaction, and further react with the ambient carbon dioxide (CO<sub>2</sub>) and moisture to form other compounds of sodium such as sodium hydroxide, sodium carbonate or sodium bicarbonate (Clough and Garland, 1971). This type of secondary sodium leakage scenario

and its consequences are generic to all SFR. The key parameters in the sodium fire are the burning rate and release rate. The burning rate decides the quantity of sodium aerosols released from the fire. In a pool fire scenario, surface layer (area of the pool) is having contact with atmosphere and as burning progresses the top most layer becomes oxide which extinguishes the fire leaving molten sodium at the bottom. The duration of fire and area of pool give the aerosol concentration. Further, the generated aerosols would be expected to get distributed uniformly in the SGB volume ~20,000 m<sup>3</sup> and tend to settle down on the ground, wall and other components. According to the literature (Hilliard et al., 1979) 30% of the aerosols only released from the multiple path ways in the exhaust system to the atmosphere and dispersed according to metrological condition. With the passage of time the hydroxide form of aerosols get converted to carbonate form and finally to bicarbonate form. At this juncture, it is important to

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consider that, the hydroxide aerosols are highly corrosive and harmful to human tissues and it will cause damages to surrounding materials. According to Occupational Safety and Health Administration (OSHA) and American Conference of Governmental Industrial Hygienists (ACGIH), the inhalation Threshold Limit Values (TLV) for hydroxide is  $2 \text{ mg m}^{-3}$  and the TLV–TWA (Time Weighted Average) limit for carbonate and bicarbonate is  $10 \text{ mg m}^{-3}$  as dust exposure limits. As the carbonation progresses, the hydroxide species is getting converted to carbonate and then to bicarbonate which reduces the chemical toxicity, so that, human intervention can be decided after the sodium fire event. Hence, one should know how long these particles will remain in such chemical form and rate of change of chemical species for a given atmospheric condition prevailed in the SGB as well in the environmental condition of the surround place.

Towards this sodium fire experiments were conducted in Aerosol Test Facility and aerosols released from such experiments were analysed for chemical characterization following a novel high resolution conductometric titration technique (Subramanian et al., 2009). Experiments were conducted in controlled atmospheres with relative humidity 50% and 65%, keeping  $\text{CO}_2$  concentration same as present in ambient air (390 ppm). The initial mass concentration of sodium aerosols was kept  $\sim 2 \text{ g m}^{-3}$ . The information obtained from this experimental study could overcome some difficulties which were encountered by earlier investigators. Such difficulties were (i) the techniques adopted for chemical assay of sodium aerosols had not been described clearly by the investigators (based on the literature survey), (ii) based on experimental investigations reported by Cherdron and Jordan (1982), Hofmann et al. (1978), and theoretical prediction by Clough and Garland (1971) a comprehensive conclusion on chemical species of sodium aerosols starting from the sodium fire to the formation of bicarbonate species for a given relative humidity could not be obtained. Such obstacles were overcome by adopting high resolution conductometric titration technique for simultaneous measurement of various species of sodium aerosols with progress of time. Recently Herranz et al. (2013) reported a review work on comparison of LWR and SFR in-containment source term. They mentioned a list of reactions during sodium combustion and their conversion in open atmosphere in a tabular form. Based on the old experimental work by previous investigators and one of our latest publications they mentioned the conversion of oxide and hydroxide to carbonate and bicarbonate species with prevailing atmospheric conditions. Mathe and Kissane assumed that during an accident the mass of oxide aerosols produced by a sodium spray fire could involve more than 60% of the ejected sodium (Mathé and Kissane, 2012). Further they were interested in the physicochemical transformation of the sodium oxide aerosols since their toxicity diminishes as they age. Hence they initiated experimental and theoretical investigations on ageing of sodium combustion products (Mathe et al., 2013). They planned experimental investigations to verify Cooper's model on chemical speciation of sodium aerosol particles. It is surprising to note that in addition to our earlier work, a few old publications deal with chemical characterization of sodium aerosols released from sodium fires. Probably due to rare information regarding appropriate analytical techniques for simultaneous determination of different sodium compounds present in sodium aerosol samples no further experimental studies have been taken up in this field. Since India is engaged in promoting sodium cooled fast breeder reactors in the second stage of nuclear program (Chetal et al., 2006) we focused our attention to have a detailed investigations on chemical characterization of sodium aerosols which will be useful for environmental safety analysis at fast reactor sites.

In SFR, the reactor containment building (RCB) houses the nuclear island and it is confined. The reactor vessel is carrying

liquid sodium in which reactor is immersed. The reactor vessel is covered with roof top and sodium is protected by argon cover gas layer. The sodium (viz. secondary sodium) flows in tube into the reactor vessel, takes the heat from the primary sodium, comes out of RCB and transfers the heat to steam in stream generator located in the Steam Generator Building (SGB). The SGB is accessible and ventilated as once through mode. The major component in SGB are the secondary sodium pump, the steam generator and turbine connectivity components. The scenario presented here is the crack developed in the secondary sodium carrying pipe, leaking hot sodium into the SGB atmosphere. The sodium flows down to the lower part of the surface where trays are kept (underneath of entire pipe length). The hot sodium gets filled in the trays and starts burning as pool fire. In the event of large leakage say up to few tons, the hot sodium comes out of the tray and spreads over the floor under the secondary sodium circuit of SGB and continues to burn, releasing large quantity of aerosols. These aerosols are released through exhaust systems in SGB. Such a large leakage is classified as category 4 sodium leakage, in which sodium burning rate is  $\sim 40 \text{ kg h m}^{-2}$  and estimated release rate is  $\sim 50 \text{ g/s}$  (Indira et al., 2006). The experimental results obtained from our earlier work (Subramanian et al., 2009) would not correlate the scenario of sodium fire in SGB since, in the event of large leakage of sodium in SGB, the sodium aerosols would be released into the atmosphere and get dispersed according to the atmospheric condition and reaching the site boundary as per the prevailing wind speed (Johnson et al., 1979). In this case the maximum plume central line concentration at 50 m distance is nearly  $4 \text{ g m}^{-3}$  (based on GPM calculation) (Baskaran et al., 2007). The relative humidity conditions at Kalpakkam (coastal site) are found to vary from 50% to 90% depending upon the seasons; whereas the RH% in the inland region ( $\sim 2 \text{ km}$  inside) varies from 20% to 80%. Moreover from environmental safety point of view it is desirable to investigate the conditions which favour release of aerosols in carbonate or bicarbonate form rather than harmful sodium oxides or sodium hydroxide. In view of these requirements, in the present paper, we mainly reported carbonation of sodium oxide aerosols in various experimental conditions with relative humidity at 20% to 90%,  $\text{CO}_2$  concentrations at 390 ppm and 280 ppm and initial aerosol mass concentrations  $\sim 4 \text{ g m}^{-3}$ .

## 2. Experimental

### 2.1. Experimental set up

The experiments reported in this work were conducted in a specially designed Aerosol Test Facility (ATF) (Baskaran et al., 2004). The schematic of the experimental set up is presented in Fig. 1. The major components of ATF are (i) combustion cell made of SS-304L, in which, 10 g of sodium is heated using electrical bunsen burner under argon environment, (ii) aerosol chamber (having volume  $1 \text{ m}^3$  with height = 60 cm and diameter = 150 cm), (iii) auxiliary systems such as pneumatic control system, data acquisition system for temperature, pressure and RH during experiments, (iv) filter paper samplers to collect samples for chemical analysis as well as mass concentration of aerosol. The aerosol chamber is maintained with ambient temperature (25–28 °C) and required RH and  $\text{CO}_2$  content, before injection of aerosols.

#### 2.1.1. Filter paper sampling system

A filter paper sampling system is integrated with aerosol chamber for collection of representative samples for chemical analysis and measurement of aerosol mass concentration. The system uses 47 mm dia. filter paper sample holder loaded with Whatman (GF/A) glass microfiber filter circles and necessary flow arrangement

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