Ageing of Sodium Combustion Aerosols: a Theoretical and Experimental Study

E. Mathé[1,2], M.P. Kissane[1] and D. Petitprez[2]

[1]Institut de Radioprotection et de Sûreté Nucléaire (IRSN/PSN), Cadarache, 13115 Saint Paul-Lez-Durance, France
[2]Laboratoire de Physico-Chimie des Processus de Combustion et de l'Atmosphère (PC2A), UMR 8522 CNRS-Lille1,Université de Lille 1, 59655 Villeneuve d'Ascq, France
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Presenting author email: emmanuel.mathe@irsn.fr

In the context of the Generation IV initiative (www.gen-4.org) and, more immediately, the strategic choice France may make in the coming years regarding development of nuclear power, sodium-leak accidents in sodium-cooled fast-neutron reactor (SFR) facilities are the subject of renewed interest. The objective here is to model the aerosol contamination produced by a sodium fire in the containment during a SFR severe accident.

Assuming that an accident has occurred, it has previously been shown that the mass of oxide aerosols produced by a sodium spray fire can involve more than 60% of the ejected sodium (Mathé and Kissane, 2012). We are now interested in the physicochemical transformations of the sodium oxide aerosols since their toxicity diminishes as they age.

Theoretical Study

Sodium oxide aerosols produced inside the containment would consume ambient water vapour and carbon dioxide forming hydroxide, carbonate and bicarbonate species probably as successive concentric layers on the aerosols.

A single layer-transformation model exists (Cooper, 1980) but the theoretical approach used (diffusion in a porous medium) has not been validated.

Based on this model, a numerical simulation of the aerosol transformation for a realistic case was made. For a 1.8s spray fire producing 1t of sodium oxide aerosols in a containment with a relative humidity of 5%, we obtain the results of Figure 1.
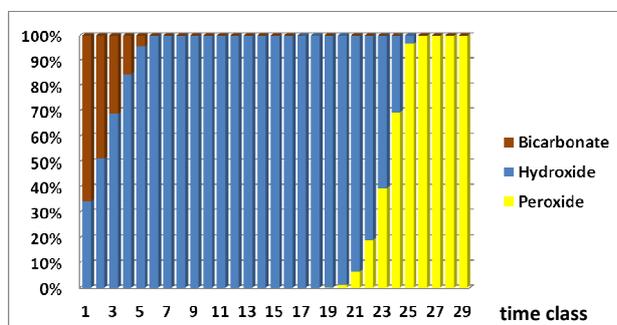

Figure 1. Evolution of the final composition of sodium aerosols for each injection-time class.

The first aerosols injected are highly transformed into bicarbonate until exhaustion of the carbon dioxide at which point the hydroxide prevails. Then, once the water vapour is fully consumed, the oxide aerosols can no longer evolve chemically.
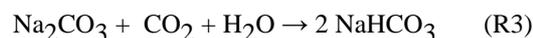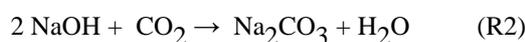
To validate this simulation we require gas diffusion coefficients in the different aerosol layers (taken here from Cooper, 1980). Analytical data being essentially nonexistent, we undertook an experiment to measure the different diffusion coefficients.

Experimental Study

To calculate the diffusion coefficients, we could measure the kinetics of gases consumed during the transformation of a given quantity of sodium oxide aerosol. The problem is that the gas consumption rate is too fast to measure. Instead, we developed experiments using thin pellets of sodium oxide and hydroxide. The different diffusion coefficients can be derived from the gas consumption rates and other experimental parameters.

First, we make the sodium oxide or hydroxide pellets in an inert preparation glove box and then we allow them to react in a second glove box with different atmospheres, i.e., specific water vapour and carbon dioxide concentrations.

H_2O and CO_2 consumption will be monitored versus the exposure time by conventional gas probes. Formation of O_2 will also be detected using an IR tuneable-diode laser spectroscopy technique. Kinetic parameters and diffusion coefficients will be calculated by fitting the experimental data to a model describing the chemical mechanisms (R1, R2 and R3).

$$2\,Na_2O_2 + 2\,H_2O \rightarrow 4\,NaOH + O_2 \qquad (R1)$$

$$2\,NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O \qquad (R2)$$

$$Na_2CO_3 + CO_2 + H_2O \rightarrow 2\,NaHCO_3 \qquad (R3)$$

The experimental results and Cooper's model will be compared: either a readjustment of Cooper's model will suffice or a new model will be developed.

Cooper, D.W. (1980) *Prediction of the Rates of Chemical Transformation of Sodium Fire Aerosols*, US NRC report NUREG/CR-1724.
Mathé, E. and Kissane, M.P. (2012) *Modélisation de la Production d'Aérosols par la Combustion de Sodium,* CFA 20121, 25-26 janvier 2012, Paris, ASFERA.