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Original Article

Theoretical simulation on evolution of suspended sodium combustion aerosols characteristics in a closed chamber

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ABSTRACT

In the unlikely event of core disruptive accident in sodium cooled fast reactors, the reactor containment building would be bottled up with sodium and fission product aerosols. The behavior of these aerosols is crucial to estimate the in-containment source term as a part of nuclear reactor safety analysis. In this work, the evolution of sodium aerosol characteristics (mass concentration and size) is simulated using HAARM-S code. The code is based on the method of moments to solve the integro-differential equation. The code is updated to FORTRAN-77 and run in Microsoft FORTRAN PowerStation 4.0 (on Desktop). The sodium aerosol characteristics simulated by HAARM-S code are compared with the measured values at Aerosol Test Facility. The maximum deviation between measured and simulated mass concentrations is 30% at initial period (up to 60 min) and around 50% in the later period. In addition, the influence of humidity on aerosol size growth for two different aerosol mass concentrations is studied. The measured and simulated growth factors of aerosol size (ratio of saturated size to initial size) are found to be matched at reasonable extent. Since sodium is highly reactive with atmospheric constituents, the aerosol growth factor depends on the hygroscopic growth, chemical transformation and density variations besides coagulation. Further, there is a scope for the improvement of the code to estimate the aerosol dynamics in confined environment.

1. Introduction

With the mission of developing the technology of Fast Breeder Reactor (FBR) in India, design and development of Prototype Fast Breeder Reactor (PFBR) was initiated in the 80's at Indira Gandhi Centre for Atomic Research (IGCAR), India. The PFBR is a 500 MWe, sodium cooled, pool type, mixed oxide (MOX) fuelled reactor [1]. Fig. 1 shows the schematic diagram of PFBR system. The active core is contained in liquid sodium pool (1100 t) of main vessel having 12.9 m diameter. The main vessel consists of core, primary pumps, and intermediate heat exchanger etc. Argon cover gas of 800 mm height is sandwiched between sodium pool and top shield [2]. The reactor core is facilitated by two pumps, which drive sodium from the cold pool at the bottom through the core. The hot sodium flows through the intermediate heat exchangers, transfers its heat to the secondary sodium and finally returns to the cold pool, completing the flow circuit. The secondary sodium circuit is meant to transfer heat to the steam water circuit. The dimension of Steam Generator Building (SGB) is 19.6 m × 41 m × 48.1 m and it houses secondary sodium components. The Reactor Containment Building (RCB) is the tallest single containment rectangular building of 35 m × 40 m size and 54.5 m height above finished floor level and is provided as an ultimate safety barrier against the radioactive fission products released during the postulated accident to the environment. The details of other components of PFBR are explained elsewhere [3].

As a part of reactor safety analysis studies, generation, characterization and evolution of sodium aerosols are being pursued. In this context, the generation of aerosols is envisaged in three conditions: i) severe accidental condition such as Core Disruptive Accident (CDA) where liquid coolant along with cover gas mixed with

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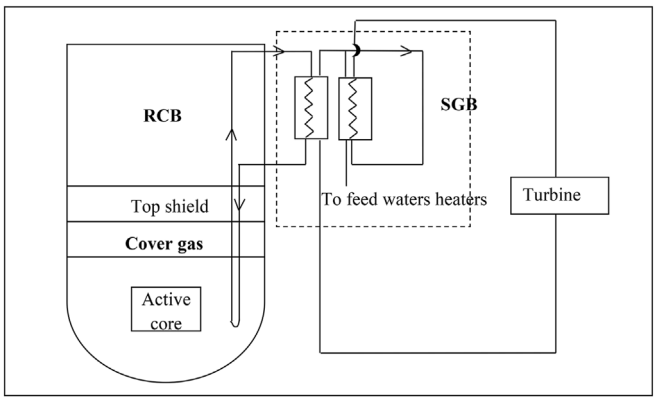

Fig. 1. Schematic diagram of PFBR.

vapors of fuel, fission products and structural materials get discharged from the primary system to the Reactor Containment Building (RCB) resulting in large amount of sodium aerosols, fission product aerosols and fission product noble gases [4]. Thus RCB would be bottled up with the aerosols and these aerosols may leak out through various leak path ways and result in environmental source term [5,6] ii) the sodium leakage from the secondary heat transport system leads to the generation of dense aerosols due to sodium fire in the SGB[7,8] where large quantity of released aerosols is envisaged [9] and iii) formation of sodium aerosols in cover gas region due to evaporation of sodium from the hot pool surface and condensation during normal operation of the reactor [10,11]. Among all these scenarios, the release of aerosols from the RCB is considered to be significant, resulting in exposures to the public in the emergency planning zone. The radiological impact analysis of a nuclear reactor in case of hypothetical severe accidental conditions is based on the estimation of the quantity of radioactive substances released to the environment [12,13]. To estimate the radioactive substances released to the environment, quantitative understanding of various aerosol processes and prediction of the temporal evolution of concentration of radioactive aerosols suspended in the containment is of vital importance and is required for assessment of radiological consequences.

The development of accurate aerosol behavior models to estimate the realistic aerosol in containment source term for SFR has been the focus of extensive theoretical studies and their validation with simulated experiments [14]. Even though the research regarding the CDA model is in progress since last 20 years, the status of the mid-1980s computer codes may be considered applicable today to a good extent [15]. Fermandjian and Dunbar (1985) reported the comparative study of the results given by some computer models such as AEROSIM (UK), MAEROS (USA), HAARM-3 (USA), AEROSOLS/A2 (France), AEROSOLS/B1 (France), and PARDISEKO-IIIb (FRG) along with the major drawbacks of aerosol modeling at that time. Later, Lhiaubet et al., 1990 validated the experimental data in a medium-scale facility (400 m^3) for a 90 min pool fire using these aerosol codes. The results showed that the evolution of suspended mass calculated with different codes is in good agreement with the experiment whereas the calculated aerosol deposition on the walls is diverging and always significantly lower than the measured values. By now, most of the aerosol modeling concepts implemented in the early codes are subsumed in codes such as ASTEC, MELCOR and CONTAIN [16]. Each of these codes has advantages and limitations regarding the accuracy of the physically realistic modeling of the actual aerosol behavior. Furthermore, it is also observed that the important aspects of the existing knowledge have not found their way into major computer codes, in particular, partitioning - or hosting - of radioactive contaminants by different phases (liquid sodium, vapor/gas phase, oxide aerosols, hydroxide droplets, etc.) is absent and the investigation of safety and potential source terms still requires development of additional modeling [15].

To describe the behavior of aerosols in containment, numerical tools such as method of moments (MOM), sectional method (SM), stochastic particle method and Monte Carlo method are employed. Among these, MOM is widely used due to its least computational time as well as relative simplicity of implementation [17]. HAARM-S (Heterogeneous Aerosol Agglomeration Revised Model-S) code is one such type based on MOM method. HAARM-S is a modified version of HAARM-3 program by studsvik for aerosol physics calculations with improved computational speed. It has been widely used for nuclear reactor safety analyses. In this code, all the aerosol agglomeration and deposition mechanisms, which are dependent upon the morphological properties of agglomerates, are corrected using experimental density correction factor [14]. HAARM-S code is developed by Battelle Columbus laboratories, Ohio and provided by NEA databank, USA. HAARM-S source code was written in FORTRAN–IV language. In the present work, the code has been updated to FORTRAN-77 to run compatible with Microsoft FORTRAN PowerStation 4.0 (on Desktop). The present paper gives details of the theoretical formulation and methodology of HAARM-S code. The aerosol characteristics (mass concentration and size) obtained using the code is validated with the experimental data conducted at Aerosol Test Facility (ATF). The paper provides results of comparison of measured and predicted aerosol mass concentrations and particle sizes and the effect of humidity on the mass and size characteristics. The code is run for two initial aerosol mass concentrations viz. 0.3 and 2.4 g/m^3 with average RH% condition (55–65%) that is prevailed in Kalpakkam and time evolution of mass concentration and size distribution is studied and compared with experimental results. The particle size growth to reach the saturation size is predicted for the two initial mass concentrations (0.5 and 3 g/m^3) along with three RH conditions viz. 20%, 50% and 90% and compared with earlier experimental study in our facility. The time evolution of aerosol concentration is studied based on the reason that the envisaged aerosol concentration and relative humidity in the containment during a severe accident of the reactor is in the range of 3–4 g/m^3 and 55–65% respectively [18]. Further, the sodium aerosol size growth is governed by both physical and chemical processes, hence three RH conditions and concentration difference of nearly one order is chosen for the study.

2. Materials and method

2.1. Theoretical simulation

The governing integro-differential equation describing the rate of change of particle concentration due to various agglomeration and deposition processes is given as

$$\frac{\partial n(x,t)}{\partial t} = \frac{1}{2}\int_0^x \varnothing(\xi, x-\xi)\, n(\xi,t) n(x-\xi,t) d\xi - n(x,t) \int_0^\infty \varnothing(x,\xi)\, n(\xi,t) d\xi - n(x,t) R(x) + S(x,t) \quad (1)$$

where, $x = \frac{4}{3}\pi r^3$ = Volume of particles with radius r;

$\xi = \frac{4}{3}\pi(r')^3$ = Volume of particles with radius r';t=Time;$n(x, t)$ = Size distribution function; $\phi(\xi, x)$ = Normalized collision kernel

predicting the probability of collision between two particles of volume x and ξ; $R(x)$ = Removal rate of particles; $S(x,t)$ = Source rate of particles introduced to the enclosed space of the vessel.

The first integral represents the formation rate of aerosols between the size x and $x+dx$ as a result of collisions between aerosols of volumes ξ and $x-\xi$ and $\phi(\xi, x-\xi)$ is the rate at which the particles of volumes ξ and $x-\xi$ combine to produce particles of volume ξ. The second integral represents the disappearance (loss) of particles in the size range between x and $x+dx$ due to collisions with all other particles.

In the code, the physical processes that are responsible for the coagulation of aerosols such as Brownian motion of the particles, gravitational settling and turbulent gas motion are modeled and these are assumed to be separable. The aerosol deposition mechanisms included in the code are gravitational sedimentation, Brownian diffusion to the walls and thermophoresis. In addition to these, some more assumptions are taken as given below:

a. The aerosols follow log-normal size distribution.
b. The gas bulk flow is zero and the particles are homogenously distributed over the simulation domain.
c. Impaction process (which may be significant only at high gas velocities) is neglected for the aerosol retention.

Eqn. (1) is multiplied by x^K and integrated over x and after rearrangement, it changes as

$$\frac{\partial X_k}{\partial t}=\frac{1}{2}\int_0^\infty d\xi\, n(\xi,t)\int_0^\infty d\zeta n(\zeta,t)\varnothing(\xi,\zeta)\left[(\xi+\zeta)^k-\xi^k-\zeta^k\right]-R_k(t)+S_k(t) \tag{2}$$

Where $X_k=\int_0^\infty n(x,t)x^k dx$ is the kth moment of the size distribution.

$R_k(t)=\int_0^\infty R(\xi)\xi^k n(\xi,t)d\xi$ and $S_k(t)=\int_0^\infty S(\xi)\xi^k d\xi$; $\zeta = x-\xi$;

Due to the simplification of equation (2) when k = 0, 1, 2, these moment equations are solved simultaneously for the unknowns X_0, X_1 and X_2. Then by assuming a lognormal distribution for the time dependent particle size distribution, the governing integro-differential equation is converted into three first order differential equations by methods of moment and solved numerically to obtain the first three moments. Hence, in terms of particle volumes, the number distribution is written as

$$n(x,t)=\frac{N(t)}{\sqrt{2\pi u(t)}}\exp\left(-\frac{ln^2\frac{x}{\bar{x}(t)}}{2u(t)}\right)\frac{1}{x} \tag{4}$$

where, $N(t)$ = total number concentration of suspended particles.
$\bar{x}(t)$ = geometric mean particle volume,
$u(t)$ = logarithmic variance.

The moments X_0, X_1 and X_2 are related to three parameters of the lognormal aerosol size distribution [i.e., N (t), $\bar{x}$ (t) and u (t)] by the following relations:

$$X_K(t)=N(t)\bar{x}(t)^k\left[\exp\frac{k^2}{2}u(t)\right] \tag{5}$$

$$X_0(t)=N(t)$$

$$X_1(t)=N(t)\bar{x}(t)\left[\exp\frac{u(t)}{2}\right]$$

$$X_2(t)=N(t)(\bar{x}(t))^2[\exp(2u(t))]$$

Alternatively, $N(t)=X_0$; $\bar{x}(t)=\frac{X_1^2}{\sqrt{X_2X_0^3}}$ and $u(t)=\ln\left(\frac{X_0X_2}{X_1^2}\right)$;

In addition to these equations, the code gives the mass and number of aerosols diffused, settled and leaked by solving additional differential equations.

The block diagram of HAARM-S code with basic structure is shown in Fig. 2. The program reads all the parameters from input.txt. In the input file, the data is divided into blocks identified with titles. The block TITLE consists of case identification and some constants defining calculation details. The maximum number of time steps and increment for time step index and at which output is desired is to be fed in this block. The block INITIAL gives the initial concentration and source rate of aerosols. The parameters to be provided in this block are the number concentration (particles/cm^3), geometric standard deviation, aerosol size (μm) and density of aerosols (g/cm^3). The COMPARTMENT block gives the geometries of containment. The vital inputs in this block are volume of the compartment, floor area to volume and wall area to volume ratio of the compartment. The LEAKAGE block gives the leakage flow to the next compartment at different time values for all compartments. The THERMAL block gives the thermo-hydraulic data such as temperature, temperature difference between gas and the compartment wall etc. for all compartments. The START card initializes the computation. The program initializes the data and calculates various parameters such as concentration and radius at each time step in each compartment till maximum time. The code gives four methods of integration: 0 for Adams - Moulton with variable incrementing, 1 for Runge-Kutta with fixed incrementing, 2 for Adams - Moulton with fixed incrementing and 3 for the MOLCOL method [19]. The output is written to the file 'output.txt'. Results are given in the output file for the quantities such as volume released, suspended mass concentration, settled mass, leaked mass, sigma of particle size, median particle radius, suspended particle concentration and source in the secondary compartment.

Table 1 gives the values of vital input parameters to get results in this paper. The initial size distribution of sodium combustion aerosols is mono-model with polydisperse distribution [20]. Based on the prior knowledge of experiments conducted at ATF, two parameters are considered-i) Geometrical Standard Deviation (GSD) as 1.1, ii) Formation of aerosol size at particular humidity. The aerosol radius is obtained from the modified cooper's relation given as

$$r=\frac{0.97\, r_o}{(1-RH)^{1/3}} \tag{6}$$

Where r_o is the dry particle radius (μm) and RH is the relative humidity. The dry particle radius measured in ATF at 20% RH is 0.45 μm. The above empirical relation is valid for the range of 20–90 RH%. In ATF, the volume of aerosol chamber (cylindrical with height 0.58 m and diameter 1.50 m) is 1.01 m^3 and the ratios of floor area to volume and wall area to volume are 1.74 and 2.67 m^{-1} respectively. The simulation is performed for maximum time of 21600 s. The density of sodium combustion aerosols is taken as 2.27 g/cm^3. The present simulation uses the MOLCOL method. The list of all input parameters used in HAARM-S code is given in supplementary material (Table S1).

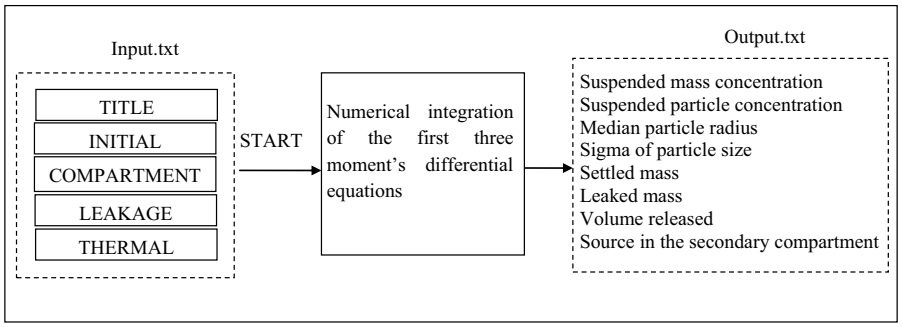

Fig. 2. Block diagram of HAARM-S code structure.

Table 1
Input parameters for HAARM-S code.

S.No.	Parameter	Value	Explanation
1	XIN(1)	1.28E+06 for 2.4 g/m^3 1.9E+05 for 0.3 g/m^3	Initial concentration of particles (particles/cm^3)
2	SIGAIR	1.1	Aerosol geometric standard deviation based on radius
3	RAIR	0.592 for 2.4 g/m^3 0.55 for 0.3 g/m^3	Aerosol mass median radius, μm
4	RHO	2.27	Particle density (g/cm^3)
5	TMAXIN	21600	Maximum time (s)
6	HIN	1	Initial time step(s)
7	INDIN	3	The MOLCOL method
8	KMAX	999	Maximum number of steps
9	ISEQ	10	Increment for time step index at which output is desired
10	NCOMP	1	Number of compartments
11	TCR	1.00E-03	Ratio of thermal conductivity of the gas to that of the particle material
12	RDELTA	2.00E-03	Initial value for diffusion boundary thickness, cm
13	TOLIN	1.00E-03	Convergence criterion
14	VO	1.01E+06	Volume of compartment (cm^3)
15	AFO	1.74E-02	Floor-to-volume ratio of compartment (cm^{-1})
16	AWO	2.67E-02	Wall-to-volume ratio of compartment (cm^{-1})
17	DELT	90	Angle between flow direction and vertical downward direction
18	VOL1	1.01E+06	Volume of outlet compartment (cm^3)
19	AQ	2.70E+04	Cross sectional area (cm^2) of the compartment in flow direction
20	VIS	1.85E-04	Viscosity of air (dyne.s/cm^2) for compartment
21	TEM	298	Temperature (K) for compartment
22	RHOAI	1.18E-03	Density of air (g/cm^3) for compartment
23	GRADW	1	Temperature gradient in the gas at the compartment wall (K)
24	TET	298	Gas temperature (K) for compartment
25	PT	1.10E+06	Gas pressure (dyne/cm^2) for compartment

2.2. Experiments at ATF

Experiments on sodium aerosol characteristics are carried out in ATF. The details of ATF are explained elsewhere [21]. This facility consists of combustion cell to produce sodium combustion aerosols and a cylindrical aerosol chamber of volume 1 m^3 made up of 3 cm thick SS-304L. The vessel is properly earthed to avoid any static charge influence. Sodium aerosols are generated by combustion of sodium in combustion cell and these aerosols are passed in to the aerosol chamber for monitoring. The aerosol chamber is maintained at the ambient temperature of ~25 °C and atmospheric CO_2 content ~390 ppm, before the injection of aerosols. The suspended aerosol concentration at various time intervals is measured with the filter paper sampler. A closed face type filter paper sampler (0.047 m diameter) and a rotary pump with a capacity of 20 lpm coupled with rotameter are used for aerosol sampling. Aerosol sampling is carried out for 1 min at a flow rate of 10 lpm. An analytical balance with an accuracy of 0.1 mg (M/s AND Corporation, Model No: GR 202) is used for the gravimetric analysis of glass fiber filters (M/s Whatman). The suspended aerosol mass concentration measured in the first minute is taken as the initial mass concentration of aerosols. The uncertainty involved in the measurement of volume of the sampling is 2% for the rotameter (as per manual) and 1s in the time measurement. Further, fluctuations in flow rate are observed while sampling due to the loading of aerosols on filter paper which gives additional uncertainty in sampled volume of about 2%. Considering all these uncertainties, the total calculated uncertainty in measured aerosol mass concentration is ±5% [18]. Humidity of the chamber is measured by humidity sensor (M/s Rotronics, Model No: HC2-XD series) and the uncertainty associated with the measurement of humidity is ±0.8%. Set of experiments are conducted by generating aerosols in two different mass concentrations viz. i) 2.4 g/m^3 at humidity 60% and ii) 0.3 g/m^3 at humidity 50%. Each experiment is repeated thrice and the uncertainty associated with the results is ±5%.

3. Results and discussion

3.1. Code validation with experiments

The HAARM-S code is run for two experimental conditions-2.4 g/m^3 at humidity 60% and 0.3 g/m^3 at humidity 50%. The initial aerosol size (diameter) obtained from modified cooper's relation is taken as 1.184 and 1.10 μm for humidity 60% and 50% respectively.

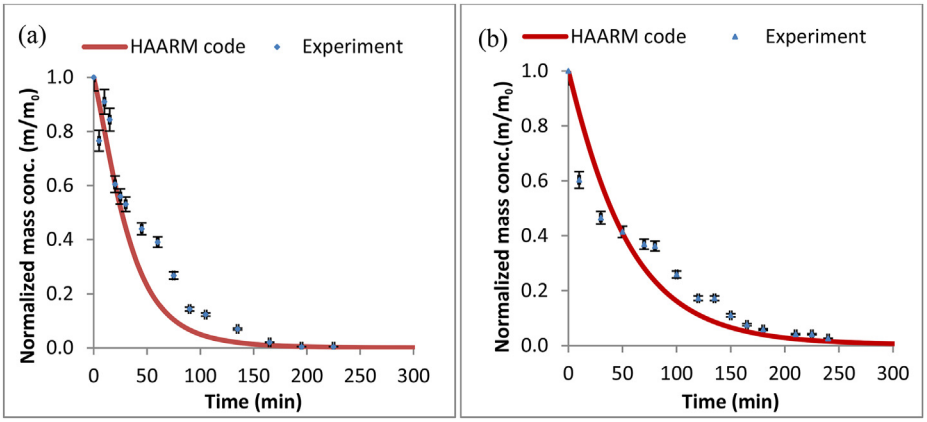

Fig. 3. Decay of aerosols mass concentrations for two initial concentrations: (a) 2.4, (b) 0.3 g/m^3.

Fig. 3(a) and (b) show the decay of normalized simulated and measured aerosols mass concentrations with progress of time for specified initial mass concentrations and with humidity. The figures also include ±5% uncertainty in experimental concentrations. It is observed that the aerosol mass concentration decay is faster in initial phase compared to later phase. This is due to the fact that, during the initial period, the aerosols undergo rapid coagulation followed by settling due to high initial concentrations. As the time progresses, the aerosols coagulation and deposition rate become slower when suspended mass concentration is sufficiently low. The decrease of mass concentration is attributed to various aerosol processes viz. Brownian coagulation, diffusion towards wall, gravitational agglomeration followed by settling etc. The dominating process for aerosol deposition is gravitational settling due to micron sized aerosols. It is also observed that, the suspended mass concentration decays faster when the initial mass concentration is high. It is inferred from the figures that, there exists difference in the simulated and experimental concentrations, which is below 30% during initial period (up to 60 min) and around 50% in the later period. This could be due to the chemical nature and hygroscopic growth of sodium aerosols [20], which is not accounted in the code. Beyond 180 minutes, the HAARM-S code predicted values are 50% less than the experimental values, but by this time the suspended mass concentration becomes one order less and hence the difference in the estimation is inconsequential. Further, the phase density of sodium combustion aerosols is taken as bulk material density, which is more than the measured density. In fact, the density of aerosols particle also changes over a period of time, which is not considered in the simulations [22].

3.2. Effect of humidity on aerosol size growth at different aerosol mass concentrations

The effect of humidity on the evolution of aerosol size is studied with two initial concentrations viz. 3 and 0.5 g/m^3 using HAARM-S code. The two concentrations are chosen in such a way to delineate the effect of concentration over size growth. The code is run for three humidity conditions - 20, 50 and 90% corresponding to lower and higher side w.r.t normal atmospheric condition of 50%. Initial aerosol size (Mass Median Diameter) obtained from modified cooper's relation is calculated to be 0.94, 1.10 and 1.88 μm for humidity conditions 20, 50 and 90% respectively. Fig. 4 (a) & (b) show the theoretical aerosol size growth with the progress of time for concentrations 3& 0.5 g/m^3 respectively and various humidity conditions- 20%, 50% and 90%. For 3 g/m^3, the size of aerosol increases from 0.94 to 1.52 μm for 20% RH case, 1.10 to 1.56 μm for 50% RH case and 1.88 to 2.00 μm for 90% RH case whereas for 0.5 g/m^3, the size of aerosol increases from 0.94 to 1.14 μm for 20% RH case, 1.10 to 1.22 μm for 50% RH case and insignificant change of aerosol size for 90% RH case. The size growth of aerosol is more for higher initial concentration when compared to lower concentration. This is due to the fact that, coagulation rate of particles is more effective in the presence of higher aerosol mass concentration. It is also observed that, in the case of 3 g/m^3, the rate of growth of aerosol size (up to 20 minutes) is found to be faster at 20% and 50% RH condition than 90%. This is due to different initial aerosol sizes at different humidity conditions and competitive mechanisms of coagulation and settling process of aerosols with time.

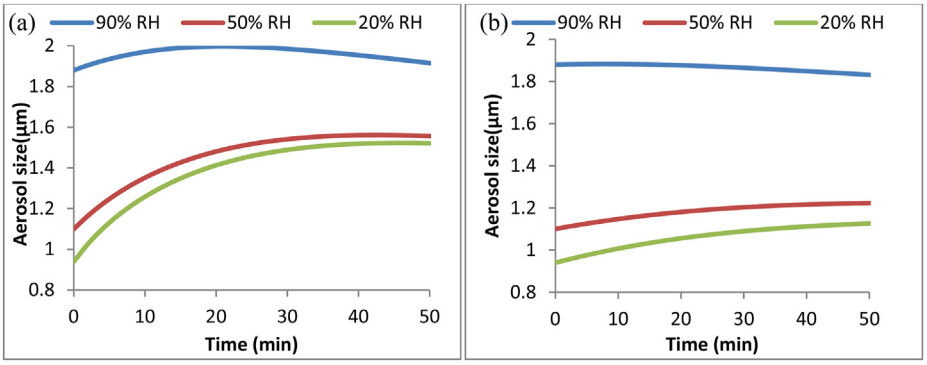

Fig. 4. HAARM code calculated aerosol size growth with the progress of time for mass concentration (a) 3 g/m^3 and (b) 0.5 g/m^3 and at humidities-20%, 50% and 90%.

Table 2
Experimental and simulated aerosol sizes (diameter) for two mass concentrations 3 and 0.5 g/m^3.

Humidity	Concentration (g/m^3)	Experiment			HAARM code			Deviation (%) Exp/Code
		Initial size ((μm)	Final size (μm)	Ratio	Initial size (μm)	Final size (μm)	Ratio	
20	3	0.90	1.50	**1.67**	0.94	1.52	**1.62**	3.0
50	3	1.10	1.70	**1.55**	1.10	1.56	**1.42**	8.4
90	3	1.80	2.40	**1.33**	1.88	2.00	**1.06**	20.3
20	0.5	0.92	1.45	**1.58**	0.94	1.14	**1.21**	23.4
50	0.5	1.10	1.60	**1.45**	1.10	1.22	**1.11**	23.4
90	0.5	1.82	2.40	**1.32**	1.88	1.88	**1.00**	24.2

The simulated aerosol size enlargement for two mass concentrations 3 and 0.5 g/m^3 is compared with the experimental results reported in Kumar et al. [10]. The experimental and theoretical initial and saturated sizes along with their ratios are presented in Table 2. The deviation of experimental ratios with theoretical ratios is also given. The experimental and calculated ratios match reasonably well with a maximum deviation of 24.2%. Nonetheless, HAARM-S code calculated results are underestimated in comparison with the measured values. This difference could be attributed to the dependence of aerosol size growth on coagulation, hydration of sodium hydroxide aerosol (hygroscopic nature) and particle density variations due to chemical conversion of sodium combustion aerosols [23,24]. Further, it is observed that the ratio of saturated size to initial size decreases as humidity increases for both the concentrations i.e., more is the humidity, lesser is the aerosol size growth. At higher humidity condition, the initial particle size is large which undergoes agglomerated settling resulting faster depletion of concentration and hence non-availability of the aerosols resulting in lesser size growth [25]. Another factor leading to this effect is the vapor pressure where the partial pressure of the water vapor inside the particle becomes equal to saturated water vapor of the surroundings [26], hence size growth is limited.

Comparison of the simulated and experimental aerosol size growth factors (ratio of saturated to initial aerosol size) shows that the simulated ratios are significantly lesser than the experimental ratios for 50% and 90% RH conditions. There is a clear discrimination between the experimental observation and simulation by coagulation growth. It is to be noted here that the measured initial size of aerosols is independent of initial aerosol mass concentration for fixed humidity. In the case of 0.5 g/m^3, the measured ratio is 1.58, 1.45 and 1.32 whereas the simulated ratio is 1.21, 1.11 and 1.00 for the three humidity conditions. The difference between the measured values and simulated values shows that particle growth has occurred due to processes other than coagulation i.e., sodium aerosols are hygroscopic. Hence it undergoes both hygroscopic and chemical conversion (particle density variations due to chemical conversion of aerosols) with progress of time in addition to the physical coagulation process [20]. It is observed from the recent study by Ref. [25], using AEROSOL/LM module and found that the average relative error between measured and predicted aerosols mass median diameter did not exceed 11% for 3 g/m^3 initial mass concentration. Similar result is observed using HAARM-S code (Table 2) for 3 g/m^3, but in the case of 0.5 g/m^3, the relative deviation between average value of measured and simulated particle size would be 24%.

When aerosols are suspended in a closed chamber, many competing processes occur simultaneously. The suspended concentration decreases with the progress of time due to several mechanisms such as gravitational settling, wall plating and ventilation if it exists. Since these removal mechanisms are dependent on particle size, Kumar et al. [10] developed a theoretical model. The first order differential equations to predict the decay of mass and number concentration of sodium aerosols in closed chamber are solved by finite difference method. Details of the simulation are explained elsewhere [20]. In this simulation, coagulation due to Brownian motion and aerosol decay due to gravitational settling, wall plating and ventilation is considered. Even though the particles are considered as monodisperse, the time varying particle decay rate constant (λ) and coagulation rate (K) is considered at every time step in the simulation. Table 3 shows the comparison of growth factors of aerosols by HAARM-S code and Kumar et al. It is observed that the HAARM-S code saturated aerosol size is in line with the theoretical simulation. The maximum deviation between the ratios obtained by two methods is less than 10%. This deviation could be attributed to assumption of monodisperse aerosols by Kumar et al. whereas HAARM-S code assumes the particles as lognormal distribution with geometric standard deviation. Another difference is, Kumar et al. considered particle coagulation due to Brownian motion only whereas HAARM-S code takes coagulation due to Brownian motion, gravitation and turbulent gas motion.

4. Summary and conclusion

Characterization and evaluation of behavior of aerosols for SFR is of vital importance for the reactor safety assessment. In this context, a study on evolution of sodium aerosol characteristics is carried out with the variation of mass concentration and humidity. The theoretical simulation using HAARM-S code is carried out and successfully validated with the observations. The maximum difference between measured and simulated mass concentrations is

Table 3
Comparison of aerosol sizes simulated by Kumar et al. [20] with HAARM code for two mass concentrations.

Humidity	Concentration (g/m^3)	Aerosol Size by theoretical simulation-[20]			Deviation of R_1 with HAARM code (%)
		Initial	Final	Ratio (R_1)	
20	3	0.94	1.59	**1.69**	4.3
50	3	1.10	1.71	**1.56**	9.9
90	3	1.88	2.10	**1.12**	5.7
20	0.5	0.94	1.11	**1.18**	2.5
50	0.5	1.10	1.11	**1.02**	8.1
90	0.5	1.88	1.91	**1.02**	2.0

below 30% at initial period (up to 60 min) and 50% at later period, where the concentration is insignificant. A comparison of the measured and simulated aerosol sizes has been carried out for various humidity and mass concentration of aerosols. The growth factor of aerosol size (ratio of saturated to initial) is found to be more in lower humidity condition and enhances in high concentration condition. The deviation in the growth factor of aerosol size is found to be more in higher humidity condition and lower concentration. The observed and simulated growth factors match reasonably well with a maximum deviation of 24%. Further, the present simulated size growth factors are compared with Kumar et al. and maximum deviation is found to be less than 10%.

The simulated growth factors (HAARM-S) are underestimated in comparison with the measured values. This difference could be attributed to the dependence of aerosol size growth on coagulation, hydration of sodium hydroxide aerosol (hygroscopic nature) and particle density variations due to chemical conversion of sodium combustion aerosols. The simultaneous incorporation of physical processes like hygroscopic growth and chemical transformation in the code is complex and involves thorough understanding of the mechanisms. Moreover, the improvement of the code to include these parameters is in progress and possible in future course of action. HAARM-S code can be used to simulate the behavior of sodium fire aerosols in a large containment. The code can be applied to predict the behavior of aerosol (sodium and fission product) bottled-up condition in RCB, during CDA. The evolution of these aerosol characteristics in RCB will help to estimate the in-containment source term, aerosols transport through cracks and environmental source term.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2021.12.029.

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