Preliminary Analyses on Mean Charge of Radioactive Aerosols for Calculating Sticking Efficiency in Aerosol Dynamics

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1. Introduction

Coagulations and settling of aerosol particles are the key phenomena for the analysis of aerosol behaviors in the containment during severe accidents. In order to analyze such key phenomena, sticking efficiency is used, which is affected by charge distributions of the aerosol particles. Since there is a general belief that radioactive sources effectively neutralize the charged aerosol [1], the possible effects of significant charging on the aerosol particles are currently ignored. In the conventional calculations such as MELCOR, sticking efficiency is assumed to be 1 and independent of the charge of the aerosol particles.

However, there have been collected evidences that electrostatics can affect the behaviors of the aerosol particles [2]. Clement & Harrison have showed that the asymmetry in ion mobilities due to the emission of charged particles during radioactive decays can modify the aerosol agglomeration rate [3]. In order to analyze charge distributions of the aerosol particles, they derived a system of mean charge distribution equations and provided approximated steady-state solutions assuming that mass concentrations of aerosol particles in each section of size are independent of the time [4, 5]. However, it is difficult for the approximated solutions to apply when the mass concentrations of aerosol particles change with time. Moreover, it is also difficult to solve the system explicitly, since the system is very stiff due to large differences between magnitudes of mean charges and those of the number densities of ions. The differences are on the order of ~10^{20}.

In order to enhance such drawbacks, we develop a calculation module using a concept of separate calculations between mean charges of each section of aerosol size and the ion concentrations. Then, we apply the module to analyze mean charge distributions of Cs and I-131 aerosols to check if it is appropriate for the assumption on the sticking efficiency in the conventional calculations to be used in aerosol dynamics. We compare the results with those of the approximated steady-state solutions given by Clement & Harrison as well.

2. Charging Process of Radioactive Aerosols & Mean Charge Distribution Equations

2.1 Charging Processes of Radioactive Aerosols

Charged particles are emitted during the radioactive decays of aerosol particles, which leaves a net charge behind an aerosol particles. The emitted charged particles ionize the surrounding gas depending on their energy. In the case of α-decay aerosols, ionization and electron ejection are done by emerging α-particles, which makes the aerosol charged. The emitted electrons slow down then, rapidly become attached to molecules to form ions. The ions in the medium recombine or diffuse onto aerosols. The processes explained above are summarized in Fig. 1.

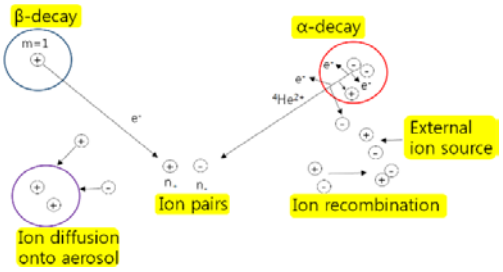

Fig. 1. Summary of charging processes of the radioactive aerosols

2.2 Derivation of Mean Charge Distribution Equations

Balance equations for the distribution of number concentration of aerosol particles of component k in section of l having a charge of j, $N_{j,l,k}$ are written as :

$$\begin{aligned}\frac{dN_{j,l,k}}{dt} &= \beta_{1,l,k}(j-1)\cdot n_{+}\cdot N_{j-1,l,k} - \beta_{1,l,k}(j)\cdot n_{k+}\cdot N_{j,l,k} \\ &+ \beta_{-1,l,k}(j+1)\cdot n_{-}\cdot N_{j+1,l,k} - \beta_{-1,l,k}(j)\cdot n_{k-}\cdot N_{j,l,k} \\ &- \eta\cdot\sum_{m} P_m\cdot\left(N_{j-1,l,k} - N_{j,l,k}\right),\end{aligned} \quad (1)$$

where

$$\begin{aligned}\beta_{1,l,k}(j)\ [m^3/s] &= \frac{e\mu_{+}}{\varepsilon_0}\frac{j}{\left\{\exp\left(2\cdot j\cdot\lambda(r_l)\right)-1\right\}}, \\ \beta_{-1,l,k}(j)\ [m^3/s] &= \frac{e\mu_{-}}{\varepsilon_0}\frac{j}{\left\{1-\exp\left(-2\cdot j\cdot\lambda(r_l)\right)\right\}},\end{aligned} \quad (2)$$

$n_{k\pm}$: number concentrations of positive or negative ions produced from component k,
$\mu_{k\pm}$ $[m^2/(V\cdot s)]$: mobilities of positive or negative ions produced from component k,
e : magnitude of electronic charge, $1.6\times10^{9}C$
m : residual charge of the aerosol particles,
ε_0 : permittivity of free space,

$$\lambda(r_l) = \frac{e^2}{8\cdot\pi\cdot\varepsilon_0\cdot r_l\cdot k_0\cdot T}, \quad (3)$$

r_l (μm) : mean radius of aerosol particle in section l,
k_0 : Boltzmann constant,
T $[K]$: temperature of atmosphere.

For ion concentrations, we have

$$\frac{dn_{k+}}{dt} = -n_{k+}\sum_l\sum_j \beta_{1,l,k}(j)\cdot N_{l,k,j} + I_k\cdot\bar{\eta}_k\cdot Z_k - \alpha_k\cdot n_{k+}\cdot n_{k-} + q, \quad (4)$$

$$\frac{dn_{k-}}{dt} = -n_{k-}\sum_l\sum_j \beta_{-1,l,k}(j)\cdot N_{l,k,j} + I_k\cdot\bar{\eta}_k\cdot Z_k - \alpha_k\cdot n_{k+}\cdot n_{k-} + m_k\cdot\bar{\eta}_k\cdot Z_k + q, \quad (5)$$

where

I : number of ion-pairs produced per decay,

$\bar{\eta}_k$ [/s] : average radioactive decay rate per particles defined as

$$\bar{\eta}_k = \frac{\sum_l \eta(r_l) N_{l,k}}{\sum_l N_{l,k}\,(\equiv Z_k)}, \quad (6)$$

α_k [m^3/s] : ion-ion recombination coefficients.

By the conservation of ion concentrations and charge of aerosol particles, equations for mean charge distributions can be derived as

$$\frac{dn_{k+}}{dt} = -n_{k+}\sum_l \beta_{1,l,k}(J_{l,k})\cdot N_{l,k} + I_k\cdot\bar{\eta}_k\cdot Z_k - \alpha_k\cdot n_{k+}\cdot n_{k-} + q, \quad (7)$$

$$\frac{dn_{k-}}{dt} = -n_{k-}\sum_l \beta_{-1,l,k}(J_{l,k})\cdot N_{l,k} + I_k\cdot\bar{\eta}_k\cdot Z_k - \alpha_k\cdot n_{k+}\cdot n_{k-} + m_k\cdot\bar{\eta}_k\cdot Z_k + q, \quad (8)$$

$$\frac{dJ_{l,k}}{dt} = \beta_{1,l,k}(J_{l,k})\cdot n_{k+} - \beta_{-1,l,k}(J_{l,k})\cdot n_{k-} + \bar{m}_k\cdot\eta_k, \quad (9)$$

where

$$\bar{m}_k = \sum_m m_k P_{m,k}, \quad (10)$$

$P_{m,k}$: probability of leaving a net charge of m behind the aerosol particle component of k,

$J_{l,k}$: mean charge of the aerosol particles of component k in section l, satisfying the following :

$$J_{l,k} = \frac{(n_{k-} - n_{k+})}{Z_k}. \quad (11)$$

$n_{k\pm}(0)$ and $J_{l,k}(0)$ are obtained by assuming that mean charge distribution and ion concentrations are at steady-state. In other words, we can obtain $n_{k\pm}(0)$ and $J_{l,k}(0)$ by setting LHS of Eqs. (7)~(9) as 0. For simplicity, Clement & Harrison proposed approximated steady-state solution of mean charge equations as

$$n_{k\pm} = \sqrt{\left(\frac{I_k\cdot\bar{\eta}_k\cdot Z_k + q}{\alpha_k}\right)} \equiv n_{k0}, \quad J_{l,k} = \frac{\varepsilon_0\eta_k(r_l)}{e\mu_- n_{k0}}. \quad (12)$$

According to Ref. [4], Eq. (12) is close to the exact solution when the ion-ion recombination processes are dominant.

2.3 Calculation Procedures for Mean Charge Distribution and Sticking Efficiency for Coagulation & Settling Kernels

Since the difference of magnitudes of ions in Eqs. (7) and (8), and those of mean charges in Eq. (9) are on the order of ~10^{20}, the system of Eqs. (7)~(9) is very stiff. Therefore, it is difficult to solve the system by Newton method with a single matrix. In this study, we decompose the equations for the ion concentrations, Eqs. (7) and (8) and those for mean charge distributions, Eq. (9). Eqs. (7) and (8) are solved via implicit Euler method with 2x2 Jacobian matrix. Since the mean charge of one section of the aerosol particle is not coupled with another, Eq. (9) for each section is separately solved by secant method. The aforementioned procedures are performed iteratively to obtain the converged solutions. Detailed calculation procedures are shown in Fig. 2.

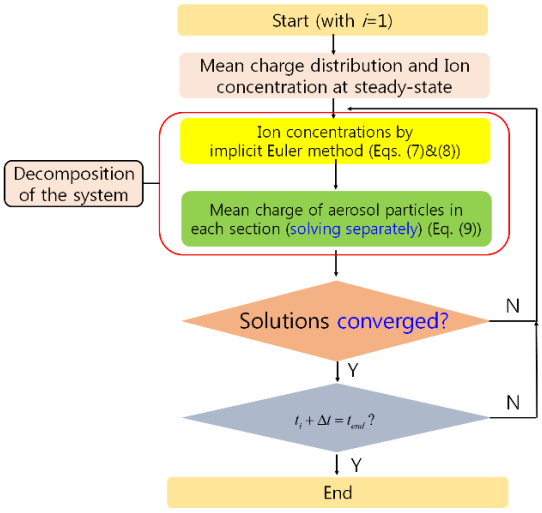

Fig. 2. Calculation procedure for mean charge distribution

Meanwhile, using the mean charges of the aerosol particles in each section, charge-dependent coagulation & settling kernels are calculated as

$$\beta(u_k, J_{l,k}, v_k, J_{p,k}) = \beta(u_k, v_k)\cdot f(Y), \quad (13)$$

where

$\beta(u_k,v_k)$: charge-independent coagulation & settling kernels for aerosol particles of component k,

$f(Y)$: sticking efficiency due to charged aerosol particles, expressed as

$$f(Y) = \frac{Y}{\exp(Y) - 1}, \quad (14)$$

$$Y = \frac{J_{l,k}\cdot J_{p,k}\cdot e^2}{4\cdot\pi\cdot\varepsilon_0\cdot k_0\cdot T\cdot(r_l + r_p)}, \quad (15)$$

u_k, v_k [kg] : mass of the aerosol particle of component k,

r_l, r_p [μm] : mean radius of aerosol particles in sections of l and p, respectively.

3. Numerical Results

Cs and I-131 aerosol particles are considered for verification of the modules. Detailed conditions used in the calculations are shown in Table 1. In the two cases, distribution of the mass concentrations of the aerosol particles are assumed to be constant over time. Properties and distribution of mass concentrations of the Cs and I-131 aerosols are shown in Table 2 and Fig. 3, respectively. Since the properties and the distributions

are constant during the calculation, the results of the time-dependent calculations should be the same as those of steady-state calculations.

Table 1. Conditions considered for the calculations

Parameters	Data
Temperature [*K*]	298
Pressure [*Pa*]	1.01E+05
Number of sections	20
Minimum and maximum diameter of the particles [*m*] (min, max)	1.0E-08, 2.0E-05
Simulation time [*s*]	10

Table 2. Properties of Cs & I-131 aerosols [5]

Parameters	Cs	I-131
Decay rate of particles per unit volume	64	3.48E+04
Number of ions produced per decay	1.0E+04	7.7E+03
Ion-ion recombination coefficient	1.6E-02	1.0E-12

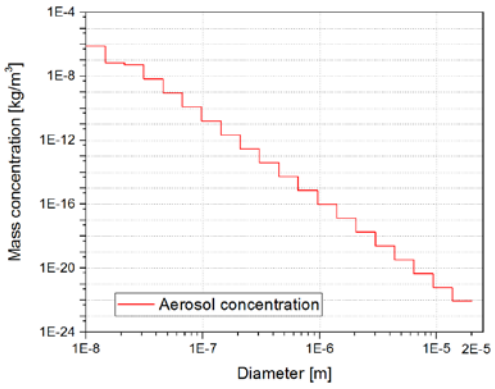

Fig. 3. Distribution of mass concentrations for Cs and I-131 aerosol particles

Mean charge distribution and the sticking efficiency of Cs aerosol particles in section 10 with those having various sizes are shown in Figs. 4 and 5, respectively.

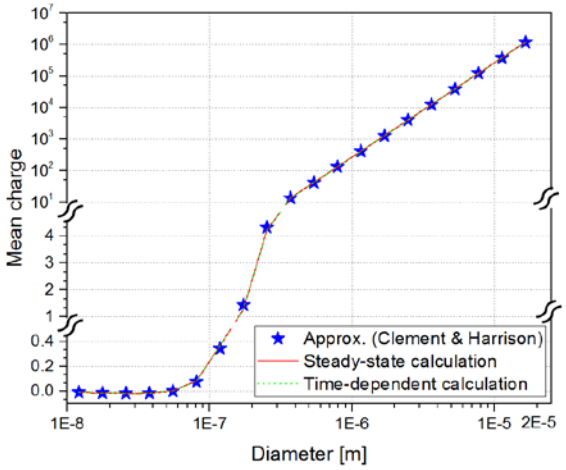

Fig. 4. Mean charge distribution of Cs aerosol particles

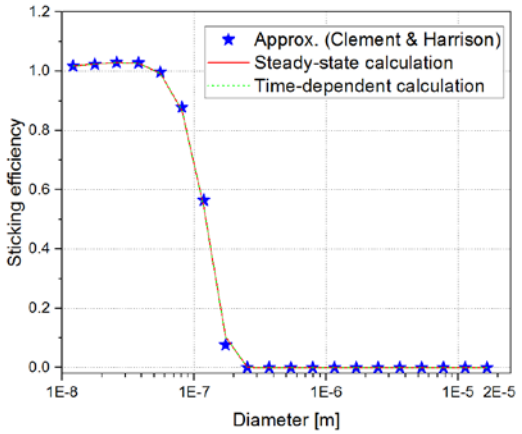

Fig. 5. Sticking efficiency of Cs aerosol particles in section 10 with those having various size

As shown in Figs. 4 and 5, mean charge distribution and sticking efficiency obtained from the time-dependent calculation show good agreement with those from the steady-state calculation and the approximated steady-state solution. Note that the sticking efficiency is not always 1 for all sizes, as shown in Fig. 5.

For I-131 aerosols, mean charge distribution and the sticking efficiency in section 15 with those having various sizes are shown in Figs. 6 and 7, respectively.

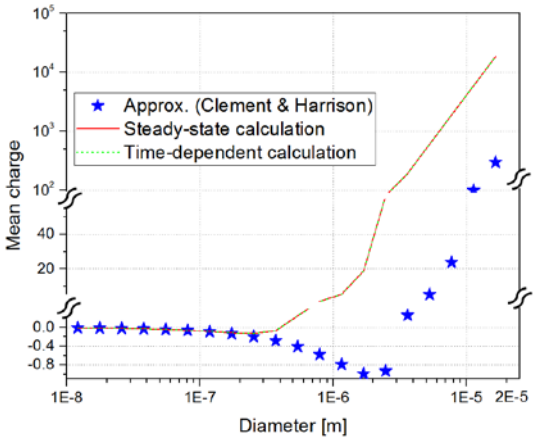

Fig. 6. Mean charge distribution of I-131 aerosol

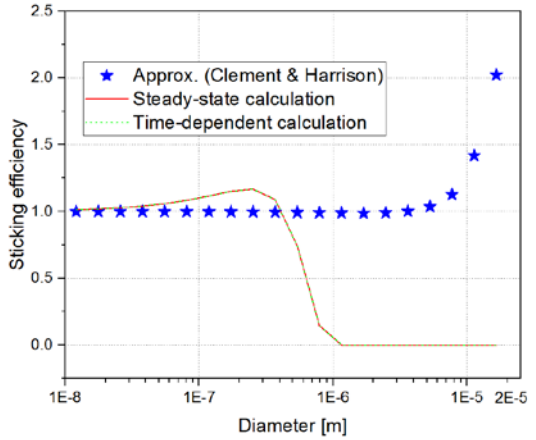

Fig. 7. Sticking efficiency of I-131 aerosol particles in section 15 with those having various size

In this case, similarly with the Cs case, as shown in Figs. 6 and 7, mean charge distribution and sticking efficiency obtained from the time-dependent calculation show good agreement with those from the steady-state calculation. However, there are large discrepancies between results from the time-dependent & steady-state calculations and the approximated steady-state solutions. For I-131 aerosols, as shown in Table 2, ion-ion recombination process is not dominant, in contrast to those of Cs aerosols.

Similarly with the sticking efficiency of Cs aerosols, those of I-131 aerosols are not always 1 as well. If such aspects on the sticking efficiency are considered in the calculations of aerosol dynamics, behaviors in coagulations and settling will be different from those without considering mean charge distributions.

4. Conclusions

In this study, we developed the calculation module for mean charge distributions of the radioactive aerosol particles in order to perform stable and accurate calculations when the mass concentrations of the aerosol particles changes with the time. Using the module, we performed the analyses to check if it is appropriate for the assumption on the sticking efficiency in the conventional calculations to be used in aerosol dynamics. We also compared the results with those of the approximated steady-state solutions given by Clement & Harrison as well.

Compared to the mean charge distributions for Cs aerosols obtained the approximated steady-state solutions, the results obtained from the module showed good agreement in terms of mean charge distribution and the resulting sticking efficiency. However, there were large discrepancies in the case of I-131 aerosols, since ion-ion recombination process is not dominant as that of Cs aerosols.

In contrast to the assumptions used in the conventional calculations of aerosol dynamics such as MELCOR, we showed that the sticking efficiency is not always 1. They are component- and size-dependent. If such aspects on sticking efficiency are considered in the calculation of aerosol dynamics, coagulations and settling rates will be different from those used in the conventional calculations. Therefore, as a future work, the module will be coupled with a calculation module of the aerosol dynamics to analyze behaviors of aerosol particles in the containment. The results of the analyses will be compared with those from conventional calculations. In addition, in order to calculate the charge distributions for the various radioactive aerosols, construction of database on the properties for other radioactive aerosols will be done as well.

ACKNOWLEDGEMENTS

This work was supported by the Nuclear Safety Research through Korea Nuclear Safety Foundation (KORSAFe), granted financial resource from Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 1305001).

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