# Preliminary Validation of Modified Transition Rate Matrix for Mason Equations of Hygroscopic Growth for Coupling with Multicomponent Sectional Equations

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## 1. Introduction

The analyses on the behaviors of the aerosol particles for estimation of the source term during the accident are performed by solving two sets of governing equations; the first equations are multicomponent sectional equations [1], which are based on the discretized sections on the size distribution of the particle. Although they offer high degree of freedom on modeling of evolution of the particles, they may require very huge computation burden when a large number of discretized sections is required to reduce numerical diffusion which is inherent on the numerical solutions of the hygroscopic growth.

In order to reduce such computation burden, hygroscopic growth is often treated separately by Mason equation in the conventional codes [2, 3]. However, the solutions of Mason equations are given with the change of radius in an individual particle. Therefore, for accurate and efficient analyses on the aerosol behaviors, especially, when hygroscopic growth and other aerosol dynamics occur simultaneously, it is essential to couple the two governing equations in rigorous manner.

As more rigorous and globally coupled scheme than those of the conventional schemes [4], some of the authors in this paper proposed a new formulation of transition rate matrix for Mason equation [5]. In this paper, for more stable numerical analyses than the one, we introduce a modified transition rate matrix based on the cumulative distribution on the number concentration of the aerosol particles in order to conserve the total number concentration and reduce the numerical diffusion. Validation of the modified transition rate matrix is done by the analytic solutions of the simplified Mason equations and the results of validation are compared to the one the some of the authors proposed [5].

## 2. Modified Transition Rate Matrix of Hygroscopic Growth for Coupling of Multicomponent Sectional Equations and Mason Equations

### *2.1 Mason Equation and Transition Rate Matrix*

In terms of change of mean diameter of aerosol particles, Mason equation for condensation & evaporation process is expressed as

$$\frac{dd_p}{dt} = \frac{4}{d_p} \cdot \frac{(S - S_r)}{a+b}, \tag{1}$$

where

$d_p$ : diameter of aerosol particle,

$S$ : saturation ratio,

$S_r$ : effective saturation ratio at the surface of the aerosol particle,

and other terms used in this Eq. are explained in Ref. 5.

The first step to construct the transition rate matrix is to obtain the change of mass concentration in each section due to hygroscopic growth. The interpolation is performed by Eq.(2) :

$$\frac{dQ_{l,k}}{dv} = b_{l,k} \cdot v^{slop_{l,k}}, \tag{2}$$

where

$Q_{l,k}$ : mass concentration of aerosol particles of component $k$ in section $l$,

$$slop_{l,k} = \frac{\ln\left(\dfrac{Q_{l+1,k} - Q_{l,k}}{v_{l+1,k} - v_{l,k}}\right) - \ln\left(\dfrac{Q_{l,k} - Q_{l-1,k}}{v_{l,k} - v_{l-1,k}}\right)}{\ln\sqrt{v_{l+1,k} \cdot v_{l,k}} - \ln\sqrt{v_{l,k} \cdot v_{l-1,k}}}. \tag{3}$$

Then, the fraction of the aerosol particles of component $k$ in section $l$ that remain in section $l$, $fr_{l,k}$, is calculated by Fig. 1 and Eq. (4). The elemental transition rate matrix, $A_{l,k}$ is obtained by Eqs. (5) and (6) .

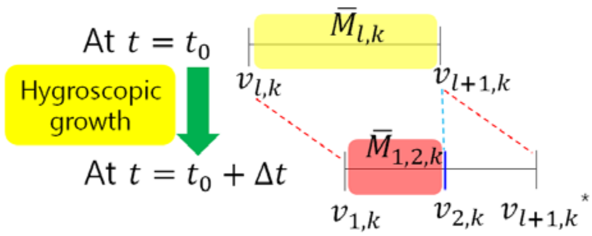


Fig. 1. Concept of change of mass concentration due to hygroscopic growth

$$fr_{l,k} = \frac{v_{2,k}^{slop_{l,k}+1} - v_{1,k}^{slop_{l,k}+1}}{v_{l+1,k}^{slop_{l,k}+1} - v_{l,k}^{slop_{l,k}+1}} = \frac{Q_{1,2,k}}{Q_{l,k}}, \tag{4}$$

$$A_{l,k} = \begin{bmatrix} -\lambda_{l,k} & 0 \\ c_{l,l+1}\lambda_{l,k} & 0 \end{bmatrix}. \tag{5}$$

where

$c_{l,l+1}$ : coefficient for conserving the mass of the particle after hygroscopic growth,

$$\lambda_{l,k} = -\frac{\ln(fr_{l,k})}{\Delta t}. \tag{6}$$

Then, the global transition rate matrix $A_m$, is obtained by Eq. (7) and the hygroscopic growth is analyzed by Eq. (8) :

$$\sum_{l=1}^{n} \sum_{k=1}^{n_{comp}} A_{l,k} = A_m, \tag{7}$$

$$\frac{dQ}{dt} = A_m Q. \tag{8}$$

*2.2 Modified Transition Rate Matrix*

During the hygroscopic growth, total number concentration of the aerosols should be conserved. It can be done by using the cumulative distribution on the number concentration of the aerosols. In addition, using the cumulative distribution can prevent the distribution after hygroscopic growth from negative overshooting which may occur for sharply peaked distribution of the aerosols due to a steep gradient.

The first step to construct the modified transition rate matrix is to convert the mass concentrations in each section into number concentrations. Then we can make a cumulative distribution on the number concentrations. The next step is to re-map the solutions of the Mason equations into the initial fixed grid used in the multicomponent sectional equations. The procedure is shown in Fig. 2.

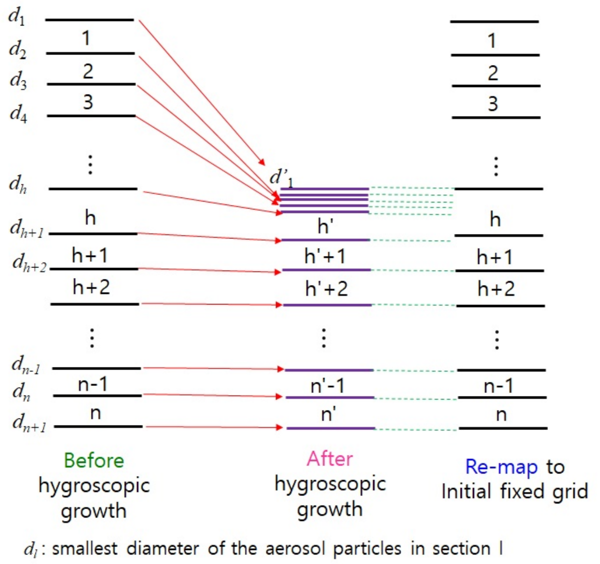


Fig. 2. Re-map of the sections after hygroscopic growth

Then, we calculate the transition rate during hygroscopic growth in terms of number concentrations. This is done by the following equations :
For the sections in which the smallest diameter of the particle after hygroscopic growth is smaller than $d'_1$,

$$\Gamma_{l,k} = -\frac{1}{\Delta t}\ln\left(\frac{N_{min}}{N_{l,k}}\right), \tag{9}$$

For the section in which the smallest diameter of the particle after hygroscopic growth is between $d_h$, and $d_{h+1}$,

$$\Gamma_{h,k} = -\frac{1}{\Delta t}\ln\left(\frac{N'_{h,k}}{\sum_{l=1}^{h-1} N_{l,k} + N_{h,k}}\right), \tag{10}$$

For the sections in which the smallest diameter of the particle after hygroscopic growth is larger than $d_{h+1}$,

$$\Gamma_{l,k} = \frac{\left(1+\Delta t\Gamma_{l+1,k}\right)N'_{l+1,k} - N_{l+1,k}}{\Delta t \cdot N_{l,k}}, \tag{11}$$

where
$N_{l,k}$ : number concentration of the aerosol particles of component $k$ in section $l$, before hygroscopic growth,
$N'_{l,k}$ : number concentration of aerosol particles of component $k$ in section $l$, after hygroscopic growth,
$N_{min}$ : minimum number concentration of the aerosol particles, set by user ($<10^{-8}$).

The number transition rate constants, $\Gamma_{l,k}$ 's are calculated in descending order of section number, i.e., $\Gamma_{n_{bin},k}$ is calculated first. Then using the $\Gamma_{n_{bin},k}$, $\Gamma_{n_{bin}-1,k}$ is calculated.

Using the number transition rates, modified transition rate matrix is constructed as Eq. (12) and similarly with the previous case, hygroscopic growth is analyzed by Eq. (13) :

$$A_n = \begin{bmatrix} -\Gamma_{1,k} & 0 & & & \cdots & & 0 \\ 0 & \ddots & & & & & \\ \vdots & & \ddots & & \ddots & & \\ c_{1,h}\Gamma_{1,k} & \cdots & c_{h-1,h}\Gamma_{h-1,k} & & -\Gamma_{h,k} & & \vdots \\ 0 & & & & c_{h,h+1}\Gamma_{h,k} & \ddots & \ddots \\ \vdots & & & & & \ddots & -\Gamma_{n,k} & 0 \\ 0 & & \cdots & & 0 & c_{n-1,n}\Gamma_{n-1,k} & 0 \end{bmatrix}, \tag{12}$$

$$\frac{dQ}{dt} = A_n Q. \tag{13}$$

## 3. Numerical Results

Validation of the modified transition rate matrix is performed by comparing analytic solutions of the simplified Mason equations written as Eq. (14) and the results of using the transition rate matrix discussed in section 2.1. Computation conditions for the validation are shown in Table 1.

$$\frac{dd_p}{dt} = \frac{P}{d_p}, \tag{14}$$

where
$P$ : a constant of the growth rate [$m^2$/sec].

Table 1. Computation conditions for the validation

| Case | $P$ | $\Delta t$ [sec] | $^*\tau = \frac{P\Delta t}{d_1^2}$ |
|---|---|---|---|
| 1 | 1.0e-16 | 1.0e+0 | 1.0e+0 |
| 2 | 1.0e-16 | 1.0e+1 | 1.0e+1 |
| 3 | 1.0e-14 | 1.0e+0 | 1.0e+2 |
| 4 | 1.0e-14 | 1.0e+1 | 1.0e+3 |
| 5 | 1.0e-10 | 3.0e-3 | 3.0e+3 |
| 6 | 1.0e-10 | 1.0e-2 | 1.0e+4 |

* τ : dimensionless time

In the calculations, the number of sections for the size distribution of the aerosol particles is 20. The smallest diameter of the aerosol is set to be 1.0e-8 m. and the largest diameter is set to be 1.0e-4 m. Size distribution of the aerosol particles are shown in Fig. 3.

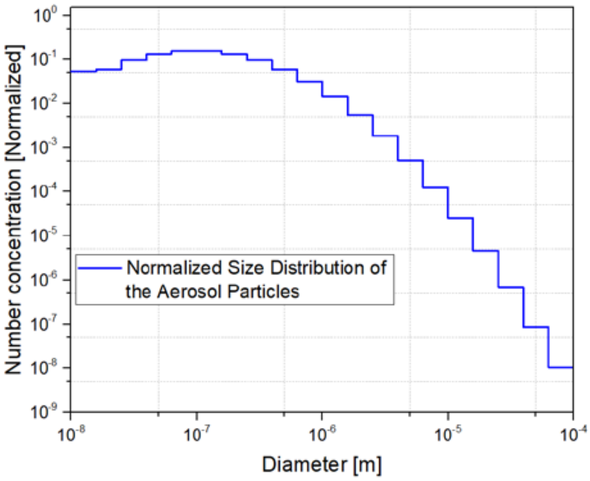


Fig. 3. Size distribution of the aerosol at initial

The total masses of the aerosol particles for cases are shown in Fig. 4. Size distribution of the aerosol particles for cases 1, 4, 5 and 6 are shown in Figs. 5~8, respectively.

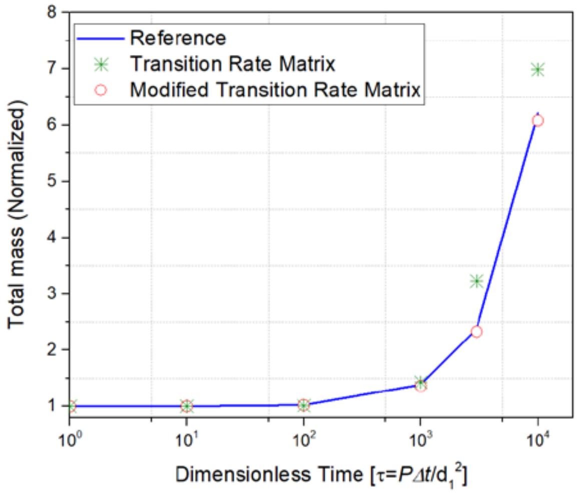


Fig. 4. Total mass of the aerosols for various cases

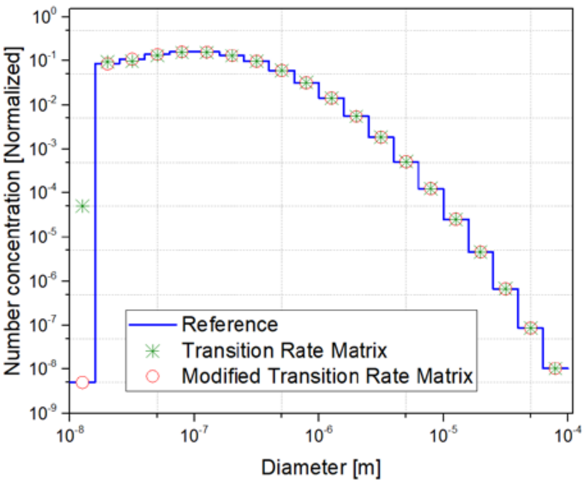


Fig. 5. Size distribution of the aerosols for case 1

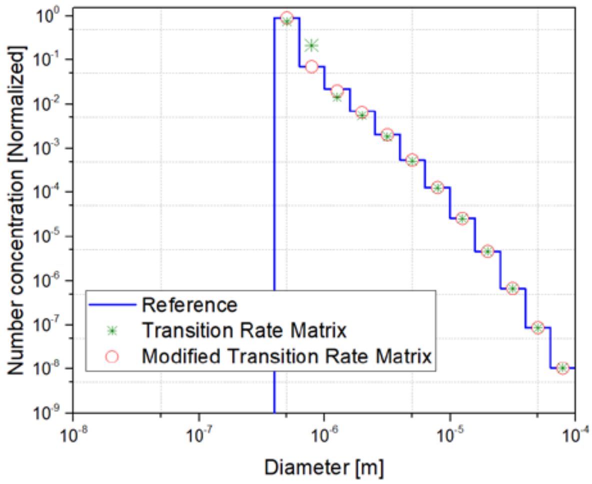


Fig. 6. Size distribution of the aerosols for case 4

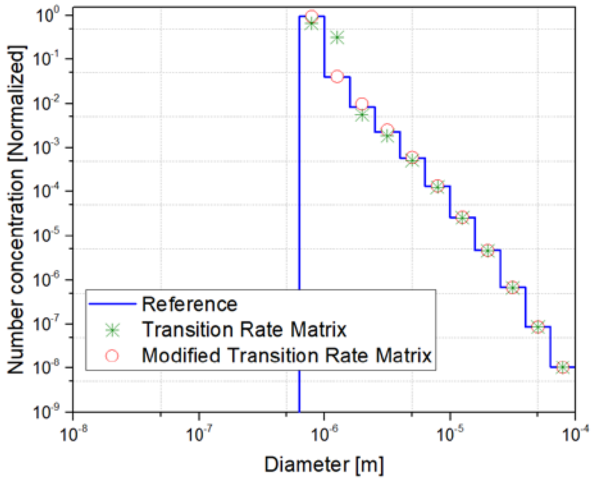


Fig. 7. Size distribution of the aerosols for case 5

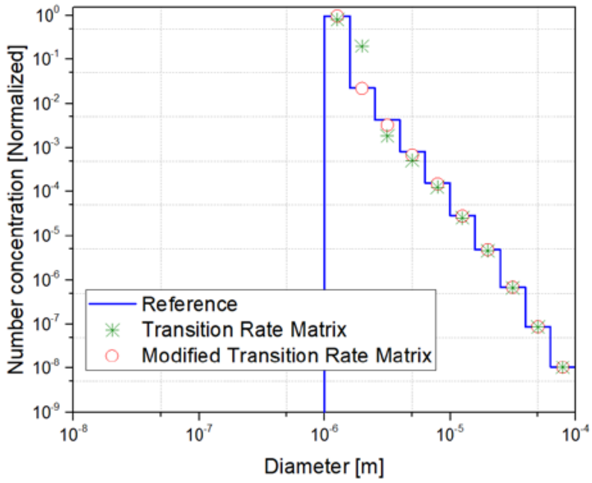


Fig. 8. Size distribution of the aerosols for case 6

As shown in Figs. 4~8, regardless of the dimensionless time, the results of the modified transition rate matrix show excellent agreement with the analytic solutions of the simplified Mason equations. Meanwhile, the results of transition rate matrix show that there are numerical diffusions when the dimensionless time is longer than

1.0e+3. Therefore, the modified transition rate matrix provide more accurate and stable results for the various dimensionless time.

## 4. Conclusions

In this paper, we proposed the modified transition rate matrix for more stable numerical analyses on the behaviors of the aerosol particles than the one some of the authors proposed in previous work [5]. The modified transition rate matrix was constructed by using the cumulative distribution of the number concentrations in order to conserve the total number concentration, which reduces the numerical diffusion of the aerosol particles.

The modified transition rate matrix was applied to the simplified Mason equation, which we can obtain analytic solutions easily. For the various dimensionless time, the modified transition matrix show excellent agreement with the analytic solutions. Meanwhile, the transition matrix show good agreement with the analytic solutions when the dimensionless time is less than $10^3$. However, there are numerical diffusion in the size distribution of the aerosols when the dimensionless time is higher than $10^3$.

As future work, with the modified transition rate matrix, we will perform validation on the coupling scheme of Mason equations with the multicomponent sectional equations. The validation will be performed not only with the analytic solutions of the simplified coupling scheme but also with the experimental results on the aerosol behaviors in which hygroscopic growth and other aerosol dynamics occur simultaneously.

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