SOLUTION OF A GENERAL POPULATION BALANCE EQUATION BY THE LAPLACE TRANSFORM TECHNIQUE

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Abstract. *The Laplace transform technique with numerical inversion is used to solve an integro-partial-differential equation related to the mathematical modeling of the physical problem to study convective processes with birth and death rates of particles or aerosols. Such model is governed by the population balance equation (PBE), in which is taken into account the nucleation, growth and coagulation processes. From these solutions, numerical results were obtained and compared with those available in previous works in the literature permitting a critical evaluation of the present solution methodology.*

Keywords*: Population balance equation, Laplace transform with numerical inversion, method of characteristics.*

1. INTRODUCTION

The dynamic behavior of a population of small particles is a subject of interest in the fields of atmospheric physics, crystallization, and colloid chemistry. In all such systems, particles grow through collisions and coalescence with other particles (coagulation) and through accretion of material in the medium containing the particles (Ramabhadran et al., 1976). The mathematical description of system where nucleation, growth, and aggregation occur is referred to as the population balance (PB). The PB often takes the form of a nonlinear integro-partial-differential equation and rarely analytically tractable (Litster et al., 1995). Precipitation and crystallization are widely studied problems in modern chemical engineering. Several phenomena are involved, such as mixing at various scales, nucleation, crystal growth, aggregation, and breakage (Marchisio et al., 2003). The influence of mixing on this kind of process has been studied for more than two decades, leading to different and sometimes contradictory results (Baldyga et al., 1995; Barresi et al., 1999; Kim and Tarbell, 1996).

Particularly, for atmospheric aerosols, the most important phenomena are coagulation and heterogeneous condensation. Because of the strong dependence of aerosol properties such as scattering, on particle size, it is desirable to understand in as much details as possible how a size distribution evolves under the influence of the two processes. The size distribution of an aerosol is described by its size distribution density function, which is governed for a general population balance equation. For simulations of atmospheric aerosol dynamics including turbulence transport and dispersion, numerical solution of the equation will ultimately be necessary. However, analytical solutions for certain limiting cases of a spatially homogeneous can be valuable in understanding the qualitative structure of the behavior in more complex situation particles (Ramabhadran et al., 1976; Peterson et al., 1978; Gelbard and Seinfeld, 1978).

In this context, the objective this work is to obtain a solution via Laplace transform with numerical inversion for a general equation that governs the size distribution of aerosols. The solution should be useful for describing the dynamic behavior of any system of particles which will be occurring coagulation and condensation. Also, analytical solutions for previously situations treated in the literature will be obtained in order to permit a critical comparison of the present solution methodology.

2. ANALYSIS

The spatial and chemical state of a homogeneous particulate system is described by the particle density function of the size distribution, n(v,t), where n(v,t)dv is the number of particles per unit volume of fluid in the range of volume v of v+dv. The drive for a similar system in which an individual particle may grow by adding material through the fluid phase (sink or loss of material), in which particles can collide and coagulate, are described by the general population balance equation (Ramabhadran et al., 1976; Drake, 1972) in the form:

$$\frac{\partial n(v,t)}{\partial t} = -\frac{\partial}{\partial v}\left[I(v,t)n(v,t)\right] + \frac{1}{2}\int_0^v \beta(v-\tilde{v},\tilde{v})n(v-\tilde{v},t)n(\tilde{v},t)d\tilde{v} - n(v,t)\int_0^\infty \beta(v,\tilde{v})n(\tilde{v},t)d\tilde{v} + S\left[n(v,t),v,t\right] \quad (1.a)$$

Where I(v,t)=dv/dt, is the change in the rate of volume of a particle of volume v for the transfer of material between the particles and fluid phase, $\beta(v,\tilde{v})$ is the coagulation coefficient for particles of volume v and $\tilde{v}$, S is the rate for net addition of new particles in the system. The initial and boundary conditions for Eq. (1.a) are usually given as:

$$n(v,0)=\frac{N_0}{v_0}e^{-v/v_0}; \quad n(0,t)=0 \tag{1.b,c}$$

The first term on the right side of Eq. (1) represents the growth rate of particles by mass transfer to individual particles. The second term represents the rate of accumulation of particles in the size range (v, v+dv) by the collision of two particles of volume $v-\tilde{v}$ and $\tilde{v}$ to form a particle of volume v (assuming conservation of volume during coagulation). The third term represents the rate of loss in the range of particle size (v, v+dv) by collision with all other particles. The last term represents all sources and sinks of particles. Equation (1) provides a wide variety of physical context, similar to colloid chemistry, atmospheric dynamics of aerosols, crystallization kinetics and biological population dynamics. In this paper, we will consider applications without the last term of Eq. (1.a), i.e., S[n(v,t),v,t]=0.

2.1. Solution methodology via Laplace transform technique

For solving the mathematical model given by integro-partial-differential equation (1), the Laplace transform technique with numerical inversion was employed. Therefore, for this purpose, three test-cases were considered, namely, i) test-case 1: $\beta(v,\tilde{v})=\beta_0$ and I(v,t)=0; ii) test-case 2: $\beta(v,\tilde{v})=\beta_1(v+\tilde{v})$ and I(v,t)=0, and iii) test-case 3: $\beta(v,\tilde{v})=\beta_0$ and I(v,t)=σv.

2.1.1. Test-case 1

The Laplace transform procedure of Eq. (1.a), in order to remove the independent variable v, for the test-case 1 is now established as follows:

$$\mathcal{L}\left[\frac{\partial n(v,t)}{\partial t}\right]=\frac{\partial \bar{n}(s,t)}{\partial t};\ \mathcal{L}\left[\frac{\beta_0}{2}\int_0^v n(v-\tilde{v},t)n(\tilde{v},t)d\tilde{v}\right]=\frac{\beta_0}{2}\bar{n}^2(s,t);\ \mathcal{L}\left[\beta_0 n(v,t)\int_0^\infty n(\tilde{v},t)d\tilde{v}\right]=\beta_0 M_0(t)\bar{n}(s,t) \tag{2.a-c}$$

where, $\bar{n}(s,t)=\mathcal{L}[n(v,t)]$.

Therefore, the transformed differential equation for this case, from the results of Eqs. (2), together with the transformed initial condition, are written as

$$\frac{\partial \bar{n}(s,t)}{\partial t}+\beta_0 M_0(t)\bar{n}(s,t)=\frac{\beta_0}{2}\bar{n}^2(s,t) \tag{3.a}$$

$$\bar{n}(s,0)=\frac{(N_0/v_0)}{s+(1/v_0)} \tag{3.b}$$

The zeroth order moment $M_0(t)$ that appears in Eq. (3.a) is obtained from the its usual definition, and for this case is given by (Ramabhadran et al., 1976)

$$M_0(t)=\int_0^\infty n(v,t)dv=\frac{2N_0}{2+\beta_0 N_0 t} \tag{3.c}$$

Equations (3) are readily analytically solved, resulting

$$\bar{n}(s,t)=\frac{4(N_0/v_0)}{(2+\beta_0 N_0 t)^2\{s+2/[(2+\beta_0 N_0 t)v_0]\}} \tag{4}$$

Applying the inverse Laplace transform in Eq. (4), one obtains

$$n(v,t)=\mathcal{L}^{-1}[\bar{n}(s,t)]=\frac{4(N_0/v_0)}{(2+\beta_0 N_0 t)^2}\mathcal{L}^{-1}\left[\frac{1}{\{s+2/[(2+\beta_0 N_0 t)v_0]\}}\right]=\frac{4(N_0/v_0)}{(2+\beta_0 N_0 t)^2}\exp\left[-\frac{2}{(2+\beta_0 N_0 t)}\frac{v}{v_0}\right] \tag{5}$$

Assuming spherical particles, the volume distribution density is now related to its diameter distribution density through

$$n_D(D,t) = n(v,t)\frac{dv}{dD} = \frac{\pi D^2}{2} n(v,t) \tag{6}$$

Therefore, introducing Eq. (5) into Eq. (6), it results

$$n_D(D,t) = \frac{12(N_0/D_0)(D/D_0)^2}{(2+\beta_0 N_0 t)^2}\exp\left[-\frac{2}{(2+\beta_0 N_0 t)}\left(\frac{D}{D_0}\right)^3\right] \tag{7}$$

2.1.2. Test-case 2

Similarly, the Laplace transform procedure of Eq. (1.a) for the test-case 2 is done, to obtain:

$$\mathcal{L}\left[\frac{\partial n(v,t)}{\partial t}\right] = \frac{\partial \bar{n}(s,t)}{\partial t};\quad \mathcal{L}\left[\frac{\beta_1}{2}\int_0^v vn(v-\tilde{v},t)n(\tilde{v},t)d\tilde{v}\right] = -\beta_1 \bar{n}(s,t)\frac{\partial \bar{n}(s,t)}{\partial s};$$
$$\mathcal{L}\left[\beta_1 n(v,t)\int_0^\infty (v+\tilde{v})n(\tilde{v},t)d\tilde{v}\right] = -\beta_1 M_0(t)\frac{\partial \bar{n}(s,t)}{\partial s} + \beta_1 M_1(t)\bar{n}(s,t) \tag{8.a-c}$$

The transformed differential equation for this case is given from Eqs. (8), in the form

$$\frac{\partial \bar{n}(s,t)}{\partial t} + \beta_1[\bar{n}(s,t) - M_0(t)]\frac{\partial \bar{n}(s,t)}{\partial s} = -\beta_1 M_1(t)\bar{n}(s,t) \tag{9.a}$$

$$\bar{n}(s,0) = \frac{(N_0/v_0)}{s+(1/v_0)} \tag{9.b}$$

Where, the zeroth and first order moments, $M_0(t)$ and $M_1(t)$, respectively, in Eq. (9.a) are obtained as (Ramabhadran et al., 1976)

$$M_0(t) = \int_0^\infty n(v,t)dv = N_0\exp(-\beta_1 v_0 N_0 t);\quad M_1(t) = \int_0^\infty vn(v,t)dv = v_0 N_0 \tag{9.c,d}$$

Equation (9.a) is a type-hyperbolic differential equation and is analytically solved through the method of characteristics, to yield

$$\bar{n}(s,t) = -\frac{v_0 N_0 \exp(-\beta_1 v_0 N_0 t)}{2[1-\exp(-\beta_1 v_0 N_0 t)]}\left\{\sqrt{\left[s+\frac{2-\exp(-\beta_1 v_0 N_0 t)}{v_0}\right]^2 - \left[\frac{2}{v_0}\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}\right]^2} - \left[s+\frac{2-\exp(-\beta_1 v_0 N_0 t)}{v_0}\right]\right\} \tag{10}$$

The inverse Laplace transform of Eq. (10) is given by (Abramowitz and Stegun, 1972)

$$n(v,t) = \mathcal{L}^{-1}[\bar{n}(s,t)] = -\frac{v_0 N_0 \exp(-\beta_1 v_0 N_0 t)}{2[1-\exp(-\beta_1 v_0 N_0 t)]}\mathcal{L}^{-1}\left[\sqrt{\left[s+\frac{2-\exp(-\beta_1 v_0 N_0 t)}{v_0}\right]^2 - \left[\frac{2}{v_0}\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}\right]^2} - \left[s+\frac{2-\exp(-\beta_1 v_0 N_0 t)}{v_0}\right]\right] = \frac{N_0\exp(-\beta_1 v_0 N_0 t)}{v\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}}\exp\left\{-[2-\exp(-\beta_1 v_0 N_0 t)]\frac{v}{v_0}\right\} I_1\left[2\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}\,\frac{v}{v_0}\right] \tag{11}$$

Applying Eq. (6) into Eq. (11), the diameter distribution density is obtained as

$$n_D(D,t) = \frac{3(N_0/D_0)\exp(-\beta_1 v_0 N_0 t)}{(D/D_0)\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}}\exp\left\{-[2-\exp(-\beta_1 v_0 N_0 t)]\left(\frac{D}{D_0}\right)^3\right\} I_1\left[2\sqrt{[1-\exp(-\beta_1 v_0 N_0 t)]}\left(\frac{D}{D_0}\right)^3\right] \tag{12}$$

2.1.3. Test-case 3

For this case, the only difference, in relation to the test-case 1, is the Laplace transform of the first term on the right side of Eq. (1.a), which is given as follows:

$$\mathcal{L}\left[\frac{\partial[\sigma v n(v,t)]}{\partial v}\right] = -\sigma s \frac{\partial \bar{n}(s,t)}{\partial s} \tag{13.a}$$

Therefore, the transformed differential equation for this test-case 3 is obtained from Eqs. (2) in conjunction with Eq. (13.a), to yield

$$\frac{\partial \bar{n}(s,t)}{\partial t} - \sigma s \frac{\partial \bar{n}(s,t)}{\partial s} = \frac{\beta_0}{2}\bar{n}^2(s,t) - \beta_0 M_0(t)\bar{n}(s,t) \tag{14.a}$$

$$\bar{n}(s,0) = \frac{(N_0 / v_0)}{s + (1/v_0)} \tag{14.b}$$

where, the zeroth order moment $M_0(t)$ is that same as given by Eq. (3.c).

Equation (14.a) is also analytically solved through the method of characteristics, to yield

$$\bar{n}(s,t) = \frac{4(N_0 / v_0)e^{-\sigma t}}{(2+\beta_0 N_0 t)^2\{s + 2e^{-\sigma t}/[(2+\beta_0 N_0 t)v_0]\}} \tag{15}$$

The inverse Laplace transform of Eq. (15) is

$$n(v,t) = \mathcal{L}^{-1}[\bar{n}(s,t)] = \frac{4(N_0 / v_0)e^{-\sigma t}}{(2+\beta_0 N_0 t)^2}\mathcal{L}^{-1}\left[\frac{1}{\{s + 2e^{-\sigma t}/[(2+\beta_0 N_0 t)v_0]\}}\right] = \frac{4(N_0 / v_0)}{(2+\beta_0 N_0 t)^2}\exp\left[-\frac{2e^{-\sigma t}}{(2+\beta_0 N_0 t)}\frac{v}{v_0} - \sigma t\right] \tag{16}$$

In terms of diameter distribution density by applying Eq. (6) into Eq. (16), one has

$$n_D(D,t) = \frac{12(N_0 / D_0)(D/D_0)^2}{(2+\beta_0 N_0 t)^2}\exp\left[-\frac{2e^{-\sigma t}}{(2+\beta_0 N_0 t)}\left(\frac{D}{D_0}\right)^3 - \sigma t\right] \tag{17}$$

2.2. The numerical inverse Laplace transform

The Laplace transform of f(t) given by $F(s) = \int_0^\infty f(t)e^{-st}dt$ is a highly stable operation, in the sense that small fluctuations in f(t) are averaged out in determination of the area under a curve. Also, the weighting factor, e^{-st}, means that the behavior of f(t) at large t variable is effectively ignored, unless s is small. As a consequence of these effects, a large change in f(t) at large t indicates a very small in F(s). In contrast, the inverse Laplace transform of a function, going from F(s) to f(t), is highly unstable. A tiny variation in F(s) may result in a large variation of f(t). However, there is no general, completely satisfactory numerical method for inverting Laplace transform. But, to relatively smooth functions, various methods are presented in the literature. For example, Bellman et al. (1966) convert the Laplace transform to a Mellin transform ($x=e^{-t}$) and use numerical quadrature based on shifted Legendre polynomials. Krylov and Skoblya (1969) focus on evaluation of the Bromwich integral, as one technique to replace the integrand with an interpolating polynomial of negative powers and integrate analytically.

Equations (4), (10) and (15) above are non-linear in the s variable. Particularly, for these test-cases, it was possible to perform the inverse Laplace transform. However, there will be situations where the inverse transform of such complex functions cannot be determined exactly or explicitly.

Alternatively, aiming at more general cases, in the present work, Eqs. (4), (10) and (15) are numerically inverted through of the subroutine DINLAP from the IMSL Library (1991) at each time and spatial position of interest with a required tolerance of 10^{-4}. The subroutine DINLAP computes the inverse Laplace transform of a complex-valued function. The computation of the inverse Laplace transform is based on applying the epsilon algorithm to the complex Fourier series obtained as a discrete approximation to the inversion integral. Given a complex-valued transform F(s), the trapezoidal rule gives the approximation to the inverse transform.

3. RESULTS AND DISCUSSION

Numerical results for the diameter distribution density were obtained for the three test-cases considered. For this purpose, were developed computer codes in Fortran 90/95 programming language, which were run on a Pentium IV 1.70 GHz computer. For the numerical inversion of Eqs. (4), (10) and (15), the subroutine DINLAP from the IMSL (1991) Library was used with a tolerance of 10^{-4}.

Figures 1 to 5 bring the comparison among the present results (analytical and numerical inversion) against those obtained by Gelbard and Seinfeld (1978) also using solutions via Laplace transform for the cases of pure coagulation with constant kernel, condensation and coagulation with constant kernel pure coagulation with varying kernel, here referred to as test-case 1, 2 and 3, respectively.

Figure 1 shows the numerical inversion and exact results for the diameter density for the test-case 1 (pure coagulation with constant kernel), using equally space elements in the range [0,3] of the ratio (D/D_0), for the dimensionless times ($\tau=\beta_0 N_0 t$), τ=0, 0.25, 1.0 and 2.0. An excellent agreement between the numerical inversion and exact solution was obtained, as well as with those results of Gelbard and Seinfeld (1978). Here, the is related to the diameter distribution in the form: $N(D,\tau)=D_0 n_D(D,\tau)/N_0$.

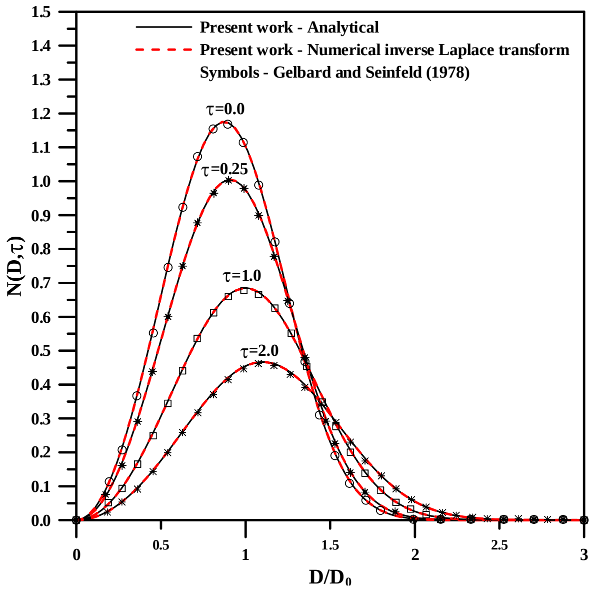

Figure 1. Comparison of the $N(D,\tau)$distribution for pure coagulation with constant kernel (test-case 1).

Figure 2 shows similar analysis as for Fig. 1, however, in order to emphasize the long-time behavior of the solution (τ=0, 10.0 and 20.0), the distribution $N(D,\tau)$ is multiplied by the term $(\tau+2)^2/4$, so that, $N_2(D,\tau)=(\tau+2)^2 N(D,\tau)/4$. Also, an excellent agreement among the results is verified.

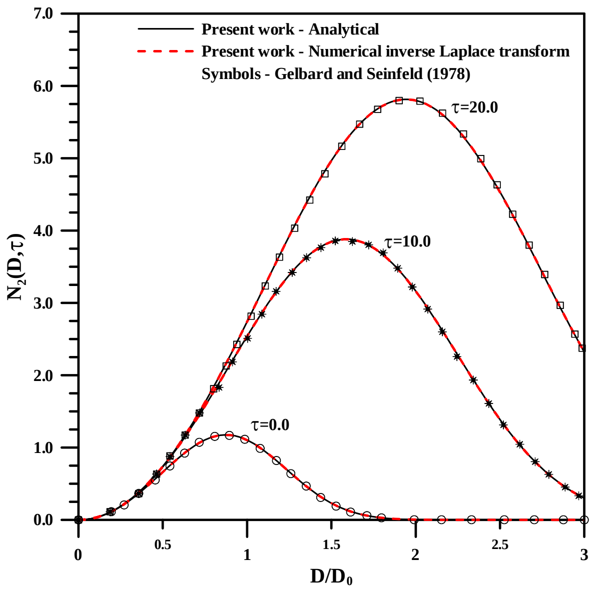

Figure 2. Comparison of the $N_2(D,\tau)$distribution for pure coagulation with constant kernel (test-case 1).

Figure 3 brings an analysis similar to that shown in Fig. 2, but using equally space elements in the range [0,4] of the ratio (D/D_0). The excellent agreement of the results is again evidenced.

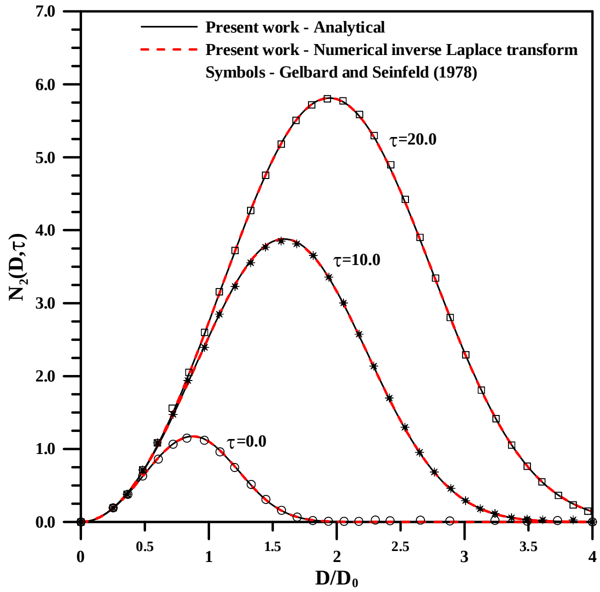

Figure 3. Comparison of the $N_2(D,\tau)$distribution for pure coagulation with constant kernel and ratio (D/D_0) in the range [0,4] (test-case 1).

Figure 4 shows a comparison of the N(D,t)distribution for pure coagulation with varying kernel (test-case 2), also using equally space elements in the range [0,3] of the ratio (D/D_0), for the dimensionless times using equally space elements in the range [0,3] of the ratio (D/D_0), for the dimensionless times ($\tau=\beta_1 v_0 N_0 t$), t=0, 0.25, 1.0 and 2.0. Once again, the three sets of results are in perfect agreement, which emphasizes the ability of the Laplace transform with numerical inversion in handling such types of problems.

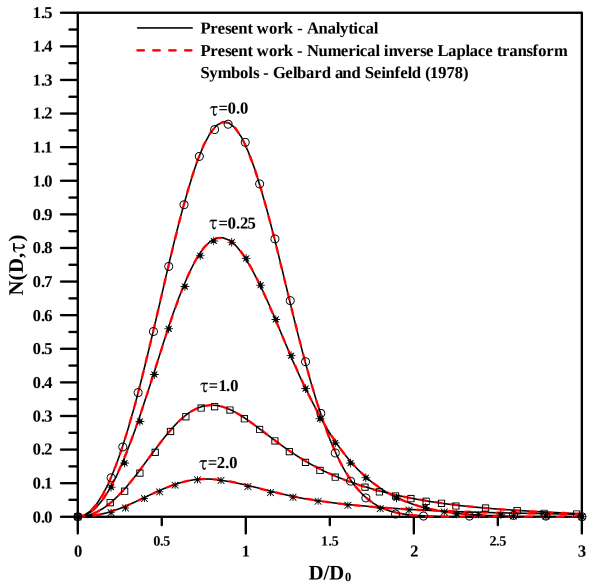

Figure 4. Comparison of the $N(D,\tau)$distribution for pure coagulation with varying kernel (test-case 2).

Finally, Fig. 5 shows the numerical inversion and exact results for the comparison of the $N(D,\tau)$distribution for both coagulation and particle growth with constant kernel (test-case 3), using equally space elements in the range [0,3] of the ratio (D/D_0), for the dimensionless times ($\tau=\beta_0 N_0 t$), τ=0, 0.25, 1.0 and 2.0 and the parameter $\Lambda=\sigma/(\beta_0 N_0)=1$. As observed throughout the analysis an excellent agreement is verified among the sets of results.

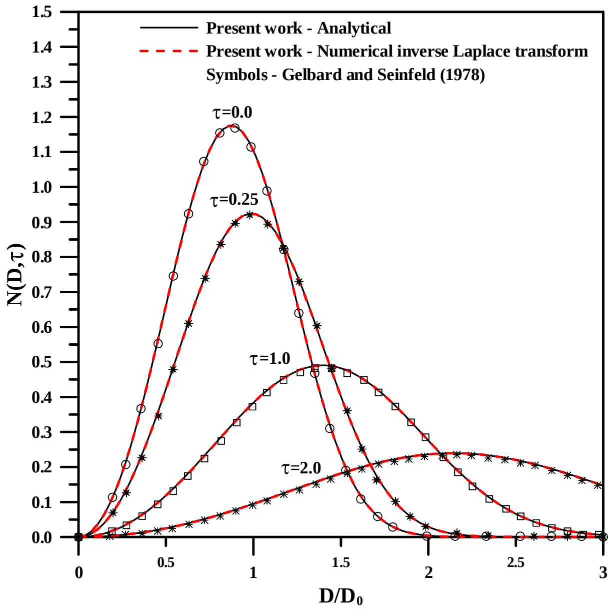

Figure 5. Comparison of the N(D,τ)distribution for both coagulation and particle growth with constant kernel (test-case 3).

3. CONCLUSIONS

The solution of population balance equation via Laplace transform with numerical inversion, for all the test-cases studied, shown excellent agreement among the results. Therefore, this technique can be used as an important alternative tool in handling problems mathematically described by integro-differential equations, as those of the analysis of particulate systems, coagulation and condensation of aerosols and crystallization processes.

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