Analytical Solutions to Fragmentation Equations with Flow

E. D. McGrady and Robert M. Ziff
Department of Chemical Engineering
University of Michigan
Ann Arbor, MI 48109

The kinetics of fragmentation processes for batch systems have been studied in depth. In this note, we develop the formal solution of the breakup population balance equation for flow systems with inflow and removal. To allow explicit solutions, we approximate physical breakup mechanisms by simple breakup rates. We consider both continuous and discrete systems.

The population balance equation PBE for continuous breakup processes, such as droplet breakup, have been considered often in connection with coagulation (Coulaloglou and Tavlarides, 1977). Complex interaction kernels for both breakup and coalescence have been assumed generally, and thus most solutions were found numerically (Ramkrishna, 1985). For the special case where there is an immediate breakup event for every coalescence (detailed balance condition) and particles only coalesce with, or breakup into, like size particles, Bajpai et al. (1976) have found an explicit solution. Curl (1963, 1967) has solved a similar model explicitly for mixing of reacting solutes dispersed in droplets. Ramkrishna (1985) has done extensive work on the continuous breakup equation; for example, Singh and Ramkrishna (1977) and Randolf (1969) have considered the case of crystallization with flow and binary breakup. In considering fragmentation in general, Fillopov (1961) has given a mathematical analysis for the case of multiple breakup while Goren (1968) and more recently Peterson (Peterson et al., 1985; Peterson, 1986) have discussed similarity (scaling) solutions.

In the following sections, we describe how batch transient and steady-state solutions may be used to find transient solutions for flow systems. The transient solutions to the batch breakup PBE have been presented in detail elsewhere (Mark and Simha, 1940; Fillipov, 1961; Kapur, 1970; McGrady and Ziff, 1987; Ziff and McGrady, 1985, 1986). To our knowledge, the steady-state problem has not been examined previously. Here we give the explicit solution to one physically important breakup kernel.

Present address of E. D. McGrady: Center for Naval Analyses, 4401 Ford Ave., P.O. Box 16268, Alexandria VA 22302.

Continuous Breakup

The PBE for breakup with inflow and removal is given by:

$$\frac{\partial c(x,t)}{\partial t} = -c(x,t)a(x) + \int_x^\infty c(y,t)b(x|y)a(y)\,dy - Bc(x,t) + c^{\text{feed}}(x) \quad (1)$$

Below we show that if a steady-state solution to Eq. 1 can be found and if the corresponding batch equations can be solved, the transient solution to Eq. 1 can also be found. In the following, we assume that Eq. 1 has been put in dimensionless form.

First, we write the general solution to Eq. 1 as:

$$c(x,t) = c^{ss}(x) + e^{-Bt}n(x,t) \quad (2)$$

Inserting Eq. 2 into Eq. 1, we find that the steady state component c^{ss} satisfies:

$$0 = -c^{ss}(x)a(x) + \int_x^\infty c^{ss}(y)b(x|y)a(y)\,dy - Bc^{ss} + c^{\text{feed}} = \Phi(c^{ss}) - Bc^{ss} + c^{\text{feed}} \quad (3)$$

and $n(x,t)$ satisfies

$$\frac{\partial n}{\partial t} = \Phi(n) \quad (4)$$

with $\Phi(n)$ defined as the linear break up operator:

$$\Phi(n) = -n(x,t)a(x) + \int_x^\infty n(y,t)b(x|y)a(y)\,dy \quad (5)$$

Equation 4 is simply the batch fragmentation equation. Thus we have divided the solution into time-dependent (batch startup) and -independent (steady state) parts. To find $c(x,t)$, we need the solution to Eq. 4 and the steady-state solution to Eq. 3 subject to the arbitrary feed $c^{\text{feed}}(x)$ and the initial condition $c(x, 0)$.

Here we present a general development of the analytic steady-state theory and give one special solution as an example. If we consider a monodisperse feed, $c^{\text{feed}}(x) = \delta(x - \phi)$, the steady-state distribution c_ϕ^{ss} is the solution to

$$0 = \Phi(c_\phi^{ss}) - Bc_\phi^{ss}(x) + \delta(x - \phi) \tag{6}$$

Then, because Eq. 3 is linear, the solution $c^{ss}(x)$ for an arbitrary feed $c^{\text{feed}}(x)$ can be found from:

$$c^{ss}(x) = \int_x^\infty c_\phi^{ss}(x)c^{\text{feed}}(\phi)\,d\phi \tag{7}$$

To develop the solution to Eq. 6, we note that at $x = \phi$, c_ϕ^{ss} must have a contribution from the delta-function feed term. Thus we write:

$$c_\phi^{ss} = p(\phi)\delta(x - \phi) + q_\phi^{ss}(x) \tag{8}$$

Inserting Eq. 8 into Eq. 6 we find $p(\phi) = 1/(a(\phi) + B)$ and the following equation for q_ϕ^{ss}:

$$0 = \Phi(q_\phi^{ss}) + Bq_\phi^{ss}(x) + \frac{a(\phi)b(x|\phi)}{a(\phi) + B} \tag{9}$$

for $x < \phi$. The last term in Eq. 9 results from integrating the delta function in Eq. 8. In terms of q_ϕ^{ss}, Eq. 7 becomes:

$$c^{ss}(x) = \frac{c^{\text{feed}}(x)}{a(x) + B} + \int_x^\infty q_\phi^{ss}c^{\text{feed}}(\phi)\,d\phi \tag{10}$$

We now show how the general solution to the batch equation (Eq. 4) can be used to find $n(x, t)$ in Eq. 2. To relate the initial condition of the system, $c(x, 0)$ to the initial condition for $n(x, t)$, we let $t = 0$ in Eq. 2 and find:

$$n(x, 0) = c(x, 0) - c^{ss}(x) \tag{11}$$

Let $n_\lambda(x, t)$ be the solution to Eq. 4 for a monodisperse initial condition, $n(x, 0) = \delta(x - \lambda)$. Thus because of the linearity of Φ, $n(x, t)$ follows from:

$$n(x, t) = \int_x^\infty n_\lambda(x, t)\,[c(\lambda, 0) - c^{ss}(\lambda)]\,d\lambda \tag{12}$$

We can write n_λ as

$$n_\lambda(x, t) = r(x, t)\delta(x - \lambda) + q_\lambda(x, t) \tag{13}$$

It follows from Eq. 4 that $r(x, t) = e^{-a(x)t}$, and $q_\lambda(x, t)$ satisfies:

$$\frac{\partial q_\lambda}{\partial t} = \Phi(q_\lambda) + a(\lambda)b(x|\lambda)e^{-a(\lambda)t} \quad x < \lambda \tag{14}$$

where the last term results from the integration of the gain term in Eq. 4 across the delta function in Eq. 13. Thus, putting Eqs. 12, 13 and 14 together, we find that $n(x, t)$ is formally determined by

$$n(x, t) = e^{-a(x)t}[c^0(x) - c^{ss}(x)] + \int_x^\infty q_\lambda(x, t)\,[c^0(\lambda) - c^{ss}(\lambda)]\,d\lambda \tag{15}$$

To write the general form of the solution, we substitute Eqs. 2 and 10 into Eq. 15 and obtain:

$$\begin{aligned} c(x,t) = {} & \frac{c^{\text{feed}}(x)}{a(x) + B} + 2\int_x^\infty q_z^{ss}(x)c^{\text{feed}}(z)\,dz + e^{-(a(x)+B)t} \\ & \cdot \left[c(x,0) - \frac{c^{\text{feed}}(x)}{a(x) + B} - 2\int_x^\infty q_z^{ss}(x)c^{\text{feed}}(z)\,dz\right] + e^{-Bt}\int_x^\infty \\ & \cdot\, dz q_z(x,t)\left[c(z,0) - \frac{c^{\text{feed}}(z)}{a(z) + B} - 2\int_z^\infty dy q_y^{ss}(z)c^{\text{feed}}(y)\right] \end{aligned} \tag{16}$$

As an example, we consider the multiple breakup model with $a(x) = x^{\beta+1}$, $b(x|y) = (\beta + 2)/y^{\nu+1}x^\nu$ which has been discussed previously (Fillipov, 1961; Peterson, 1986; McGrady and Ziff, 1987). For q_λ^{ss}, we need to solve the steady-state equation (Eq. 9) which becomes:

$$0 = -q_\lambda^{ss}x^{\beta+1} + 2\int_x^\lambda w^{\beta-\nu}x^\nu q_\lambda^{ss}(w)\,dw - Bq_\lambda^{ss}(x) + \frac{(\nu + 2)\lambda^{\beta-\nu}x^\nu}{\lambda^{\beta+1} + B} \tag{17}$$

We find:

$$q_\lambda^{ss} = 2\lambda^{(2\beta-\nu)/2}\frac{(\lambda^{\beta+1} + B)^{(\nu-\beta+1)/(\beta+1)}}{(x^{\beta+1} + B)^{(\beta+\nu+3)/(\beta+1)}} \tag{18}$$

and the solution to Eq. 14 gives:

$$q_\lambda(x, t) = e^{-t\lambda^{\beta+1}}\left(\delta(x - \lambda) + (\nu + 2)t\lambda^{\beta-\nu}x^\nu M \cdot \left[\frac{\beta + \nu + 3}{\beta + 1}, 2, (\lambda^{\beta+1} - x^{\beta+1})t\right]\right) \tag{19}$$

where $M[a, b, \lambda]$ is Kummer's confluent hypergeometric function (Abramowitz and Stegun, 1964) and $c(x, 0)$ is the initial condition. If we assume that $\beta = \nu = 0$ (equal reactivity model), no disperse phase is present initially [$c(x, 0) = 0$ for all x], and a monodisperse feed [$c^{\text{feed}}(x) = \delta(x - \phi)$], Eq. 19 becomes:

$$q_z(x, t) = e^{-xt}\{\delta(x - z) + [2t + t^2(z - x)]\} \tag{20}$$

and Eq. 16 yields:

$$c(x, t) = (1 - e^{-(x+B)t})\left[\frac{\delta(x - \phi)}{\phi + B} + \frac{2\phi^2(B + \phi)}{(x + B)^3}\right] - \frac{e^{-(x+B)t}(\phi + B)}{(x + B)^2}[2t - t^2(x + B)(\phi - x)] \tag{21}$$

In this case we have found a completely explicit solution to the flow problem with fragmentation.

Discrete Breakup

An analysis similar to that given for continuous breakup also holds for the discrete process. We can rewrite Eq. 1 in its discrete analog as:

$$\frac{dc_k}{dt} = -c_k a_k + \sum_{j>k} c_j a_j b_k^j - Bc_k + c_k^{\text{feed}} \tag{22}$$

We write:

$$c_k(t) = c_k^{ss} + e^{-Bt} n_k(t) \tag{23}$$

where c_k^{ss} is the concentration of particles size k in the steady-state size distribution and $n_k(t)$ is the concentration of particles that size in the transient distribution. In principle, Eq. 22 can be solved for any $a_j b_k^j$ as the set of ordinary differential equations it describes are triangular as in the case with no feed or removal (Basedow et al., 1978). Compact solutions, expressed as series of simple functions are not as easily obtained, however. The steady-state size distribution, c_k^{ss}, satisfies:

$$0 = -a_k c_k^{ss} + \sum_{j>k} a_j b_k^j c_j^{ss} - Bc_k^{ss} + \delta_{k-f} \tag{24}$$

where the general feed c_k^{feed} has been replaced with a monodisperse feed, $c_k^{\text{feed}} = \delta_{k-f}$. To recover the general case we simply write:

$$c_k^{ss} = \sum_{f=k}^{\infty} c_k^{ss}(f) c_f^{\text{feed}} \tag{25}$$

as a result of the linearity of Eq. 22. As in the case of continuous breakup, if we assume a steady-state solution of the form:

$$c_k^{ss} = p_k \delta_{k-f} + q_k^{ss} \tag{25}$$

we find $p_k = 1/(a_k + B)$ from Eq. 23. Substituting Eq. 25 into Eq. 22 also gives:

$$0 = -a_k q_k^{ss} + \sum_{j=k+1}^{f} a_j b_k^j q_j^{ss} - Bq_k^{ss} + \delta_{k-f} \tag{26}$$

for the function q_k^{ss}.

As was the case for the continuous model, the solution for $n_k(t)$ in Eq. 23 is given by the transient, batch equation for discrete fragmentation. Again, if we have the solution n_k^ℓ, then the solution for a general initial condition can be found by convoluting the monodisperse solution with the general initial condition and steady-state solutions. In this case the initial distribution for n_k is given by $c_\ell(0) - c_\ell^{ss}$, so we have:

$$n_k(t) = \sum_{\ell=k}^{\infty} n_k^\ell [c_\ell(0) - c_\ell^{ss}] \tag{27}$$

Thus we use solutions to the batch equation for monodisperse initial conditions $n_k^\ell(0) = \delta_{k-\ell}$ to find the transient behavior. The development is analogous to the continuous problem as given in the previous section.

Here we give an example of flow solution for a discrete problem analogous to the continuous model whose solution is given by Eq. 24. In this model, the product formation rate does not depend on particle size since $a_k b_j^k = 2$. Equation 24 becomes:

$$0 = -c_k^{ss}(k-1) + 2\sum_{j>k} c_j^{ss} - Bc_k^{ss} + \delta_{k-f} \tag{28}$$

Simple successive substitution with $k = f, f-1, f-2, \ldots$ gives:

$$c_k^{ss} = \begin{cases} \dfrac{1}{B+f-1} & \text{if } k = f; \\[2ex] \dfrac{2(f+B)}{(k-1+B)(k+B)(k+1+B)} & \text{if } k < f \end{cases} \tag{29}$$

for the steady-state solution. The solution to the batch equation is given by (Montroll and Simha, 1940):

$$n_k^\ell = \begin{cases} e^{-(l-1)t} & \text{if } k = \ell; \\ e^{-(k-1)t}[2p + p^2(\ell - k - 1)] & \text{if } k < \ell; \end{cases} \tag{30}$$

Combining Eqs. 23, 27, 29 and 30 and assuming a zero initial condition $c_k(0) = 0$, as we did for the continuous example, we find:

$$c_k(t) = \begin{cases} (1 - e^{-(f+B-1)t}) \dfrac{1}{(f+B-1)} & \text{if } k = f; \\[2ex] (1 - e^{-(f+B-1)t}) \\ \quad \cdot \dfrac{(k+B-1)2(f+B)}{(k+B)(k+B+1)} \\ \quad - \dfrac{e^{-(k+B-1)t}(f+B)}{(k+B)(k+B+1)}(2p + p^2(k+B)) \\ \quad + e^{-(k+B-1)t} p^2 \quad \text{if } k < f. \end{cases} \tag{31}$$

This is the explicit solution of the model with a monodisperse feed at size f, in a system with no particles present initially.

Conclusions

We have thus given a recipe to find the solution to the PBE with inflow and outflow, and found explicit solutions for an important class of rate kernel. This rate expression applies to such widely diverse areas as aerosol breakup (Peterson, 1985, 1986), depolymerization kinetics, and solids grinding (Austin, 1976).

Notation

$a(x), a_k$ = rate at which a particle size k or x breaks up
B = dimensionless particle removal strength (constant)
$b(x|y), b_k^j$ = probability of producing a particle size x or k when one of j or y breaks up
c^{feed} = particle size distribution of inflow stream
$c(x, t), c_k(t)$ = continuous or discrete particle size distributions
f = discrete monodisperse feed size
j, k = discrete particle size
ℓ = size of discrete, monodisperse initial condition
$n(x, t)$ = batch startup portion of the flow equation solution
$p(\phi)$ = feed contribution term to the steady-state solution
q_ϕ^{ss} = transient, startup term for the steady-state solution

t = time
x, y = continuous particle size

Greek letters

δ = Dirac delta function (continuous), Krondecker delta function
λ = continuous size of monodisperse initial condition
Φ = fragmentation equation operator, Eq. 5
ϕ = particle size of monodisperse feed stream (discrete)

Subscripts/superscripts

ϕ, f = particle size distribution given for monodisperse feed size ϕ or f
λ, ℓ = particle size distribution given for monodisperse initial condition λ or ℓ
ss = steady-state particle size distribution

Literature Cited

Abramowitz, M., and I. Stegun, *Handbook of Mathematical Functions*, Dover, New York (1964).

Austin, L., K. Shoji, V. Bhatia, V. Jindal, and K. Savage, "Some Results on the Description of Size Reduction as a Rate Process in Various Mills," *Ind. Eng. Chem. Proc. Des. Dev.*, **15**, 187 (1976).

Bajpai, R. K., D. Ramkrishna, and A. Prokop, "A Coalescence Redispersion Model for Drop-Size Distributions in an Agitated Vessel," *Chem. Eng. Sci.*, **31**, 913 (1976).

Basedow, A. M., K. H. Ebert, and H. J. Ederer, "Kinetic Studies on the Acid Hydrolysis of Dextran," *Macromol.*, **11**, 774 (1978).

Coulaloglou, C. A., and L. L. Tavlarides, "Description of Interaction Processes in Agitated Liquid-Liquid Dispersions," *Chem. Eng. Sci.*, **32**, 1289 (1977).

Curl, R. L., "Dispersed Phase Mixing: I. Theory and Effects in Simple Reactors," *AIChE J.*, **9**, 175 (1963).

Curl, R. L., "Dispersed Phase Mixing Effects on Second Moments in Dominantly First Order, Backmix Reactors," *Chem. Eng. Sci.*, **22**, 353 (1967).

Fillipov, A. F., "On the Distribution of the Sizes of Particles which Undergo Splitting," *Theory Probab. Its Appl.*, Engl. Transl., **4**, 275 (1961).

Goren, S. L., "Distribution of Lengths in the Breakage of Fibres or Linear Polymers," *Can. J. of Chem. Eng.*, **46**, 185 (1968).

Kapur, P. C., "A Similarity Solution to an Integro-Differential Equation Describing Batch Grinding," *Chem. Eng. Sci.*, **25**, 899 (1970).

Mark, H., and R. Simha, "Degradation of Long Chain Molecules," *Trans. Faraday Soc.*, **35**, 611 (1940).

McGrady, E. D., and R. M. Ziff, " 'Shattering' Transition in Fragmentation," *Phys. Rev. Lett.*, **58**, 892 (Mar. 2, 1987).

Peterson, T. W., M. V. Scotto, and A. F. Sarofim, "Comparison of Comminution Data with Analytical Solutions of the Fragmentation Equation," *Powder Technol.*, **45**, 87 (1985).

Peterson, T. W., "Similarity Solutions for the Population Balance Equation Describing Particle Fragmentation," *Aero. Sci. Tech.*, **5**, 93 (1986).

Randolf, A. D., "Effect of Crystal Breakage on Crystal Size Distribution in a Mixed Suspension Crystallizer," *Ind. Eng. Chem. Fund.*, **8**, 53 (1969).

Ross, S. L., F. H. Verhoff, and R. L. Curl, "Droplet Breakage and Coalescence Processes in an Agitated Dispersion: 2. Measurement and Interpretation of Mixing Experiments," *Ind. Eng. Chem. Fund.*, **17**, 101 (1978).

Singh, P. N., and D. Ramkrishna, "Solution of Population Balance Equations by MWR," *Comp. and Chem. Eng.*, **1**, 23 (1977).

Verhoff, F. H., S. L. Ross, and R. L. Curl, "Breakage and Coalescence Processes in Agitated Dispersion. Experimental System and Data Reduction," *Ind. Eng. Fund.*, **16**, 371 (1977).

Ziff, R. M., and E. D. McGrady, "The Kinetics of Cluster Fragmentation and Depolymerisation," *J. Phys. A: Math. Gen.*, **18**, 3027 (1985).

Ziff, R. M., and E. D. McGrady, "The Kinetics of Polymer Degradation," *Macromol.*, **19**, 2513 (1986).

Manuscript received Aug. 21, 1987, and revision received June 27, 1988.