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A mathematical model for continuous crystallization

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We discuss a mixed suspension mixed product removal crystallizer operated at thermodynamic equilibrium. We derive and discuss the mathematical model based on population and mass balance equations, and prove local existence and uniqueness of solutions using the method of characteristics. We also discuss the global existence of solutions for continuous and batch mode. Finally, a numerical simulation of a continuous crystallizer in steady state is presented. Copyright © 2014 John Wiley & Sons, Ltd.

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1. Introduction

Crystallization is a liquid-solid separation process, where solids are formed from a solution. The principal processes in crystallization include nucleation, crystal growth, breakage, attrition, and possibly agglomeration and secondary nucleation.

Nucleation is the phase where solute molecules dispersed in the surrounding solvent start to form clusters, which according to the operating conditions are arranged in a defined periodic manner. Crystal growth is the subsequent accretion process of nuclei, driven by supersaturation. Crystal birth and growth cease when the solid-liquid system reaches equilibrium due to the exhaustion of supersaturation.

Figure 1 shows a drawing of a continuous crystallizer with four possible external commands. This includes solute feed to maintain a satisfactory level of supersaturation, fines removal, used in industrial crystallizers to avoid a large quantity of extremely fine crystals hindering sustainable growth, product removal, continuous flow out of the container, and heating or cooling. Internal processes include crystal birth and growth, attrition and breakage, and possibly particle agglomeration. The process operates as follows: liquid solution is fed to the crystallizer. The supersaturation is generated by cooling. Due to supersaturation, crystals are formed from the solution and grow. Solution and crystals are continuously removed from the crystallizer by the product outlet (see several applications [6, 11, 12]). Continuous crystallization processes in the pharmaceutical industry are usually designed to obtain crystals with sizes in specific range, shape, purity, and polymorphic form like the active pharmaceutical ingredients (API)[11, 12].

Crystallization is modeled by a population balance equation, in combination with a molar balance and possibly a thermodynamic or energy balance. Mathematical models of crystallization are known for a variety of processes, but continuous crystallization with fines dissolution and classified product removal including breakage has not been discussed in the literature within a complete model including population, molar and energy balances. The model we derive here includes breakage, but not agglomeration, as the latter is known to be negligible.

Related models featuring structured population balances are used in the understanding of biological population dynamics. For instance, physiologically structured populations are investigated by Farkas [16] and Farkas and Hagen [18], where the authors study stability of such processes using the semigroup approach [16, 18]. Models including coagulation-fragmentation have been considered e.g. by Amann and Walker [20], where the authors discuss the existence of solutions of continuous diffusive coagulation-fragmentation models. Similar models without diffusion are discussed by Giri [14], Rudnicki [19] and Morale [21]. Models describing the spread of infection of transmitted diseases are investigated by Calsina and Farkas [17], where the authors discuss existence of solutions using a fixed-point approach. A similar line is chosen in an epidemic model treated by Lanelli

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[22, 23], and for tumor growth by Perthame [24, 25]. Laurençot and Walker [13, 15, 26] use an approach by weak solutions for a model of infectious diseases.

Our present work applies structured population balance models in tandem with mass and energy balances to the study of continuous crystallization of Potassium Chloride (KCl) with fines dissolution and product removal including breakage. Potassium Chloride is used in medicine to prevent or to treat low blood levels of potassium (hypokalemia). It is also used in food processing as a sodium-free substitute for table salt [27].

The mathematical model presented here is based on the following assumptions. Crystals are characterized by their size L, a one-dimensional quantity, and we introduce a volume shape factor k_v such that a particle of size L has volume $v_p = k_v L^3$. The crystallizer is operated at isothermal conditions under ideal mixing due to stirring. We assume that nucleation takes place at negligible size $L = 0$, that crystal growth rate is size independent, and that agglomeration is negligible.

Under these assumptions we derive the model equations, and then validate the model mathematically by proving first local, and then global, existence and uniqueness of the solution. We also indicate how more general situations (size dependent growth, temperature dependence of saturation constant, agglomeration) can be integrated in the setting. Our proof expands on Gurtin and MacCamy (see [3]) and Calsina (see [4]), see also [2, 7]. The full model for which existence of solutions is proved comprises equations (11)-(16), which we derive in the following sections.

The structure of the paper is as follows. In sections 2.1 and 2.2 the model of the mixed suspension mixed product removal (MSMPR) crystallizer is derived from population and mass balances. Local existence and uniqueness are proved in sections 3. Global existence is discussed in section 4. Finally, a numerical simulation of a continuous crystallizer in steady state is presented in section 5.

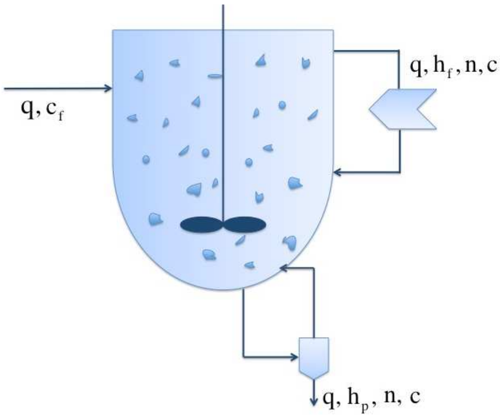

Figure 1. Continuous KCl-crystallizer [5] filled with liquid (solute and solvent) and solids (crystals). Internal processes are nucleation, growth, attrition and breakage, and possibly agglomeration. External phenomena include solute feed c_f at rate q (upper left) recycling of fines h_f (upper right), product removal h_p (lower right), stirring, and possibly heating or cooling to act on the saturation concentration c_s.

2. Modeling and process dynamics

2.1. Population balance equation

The population balance equation describes a first interaction between the population of solid crystals, classified by their size L, and a second ageless population of solute molecules of the constituent in liquid phase. The equation models birth, growth and death of crystals, as well as breakage and attrition.

We denote by $n(L,t)$ the number of crystals of the constituent of size L in one cubic meter of the suspension at time t, or crystal size distribution (CSD), whose unit is $[1/\text{mm} \cdot \ell]$. By $c(t)$ we denote the solute concentration of the constituent in the liquid phase, or in other words, the amount of solute per volume of the liquid part of the suspension, whose unit is $[\text{mol}/\ell]$. This second population is unstructured. Now the population balance equation has the form

$$\begin{cases} \dfrac{\partial n(L,t)}{\partial t} + G(c(t))\dfrac{\partial n(L,t)}{\partial L} = -\dfrac{q}{V}(1 + h_f(L) + h_p(L))\, n(L,t) \\ \qquad\qquad\qquad\qquad\qquad\qquad -a(L)\, n(L,t) + \displaystyle\int_L^\infty a(\hat{L}) b(\hat{L}, L)\, n(\hat{L}, t)\, d\hat{L} \\ n(L,0) = n_0(L), \qquad n(0,t) = \dfrac{B(c(t))}{G(c(t))} \end{cases} \tag{1}$$

Here the differential operator on the left describes the growth of the population of crystals of size L, while the terms on the right describe external effects like fines dissolution, product removal, flow into and out of the crystallizer, breakage and attrition. Extended modeling could also account for agglomeration of crystals [8, 9].

The function $h_p(L)$ describes the profile of the product removal filter, which removes large particles with a certain probability according to size. In the ideal case, assumed e.g. in [5], one has

$$h_p(L) = \begin{cases} R_p & \text{if } L \geq L_p \\ 0 & \text{if } L < L_p \end{cases} \tag{2}$$

where L_p is the product removal size and R_p the product removal rate. This corresponds to an ideal high-pass filter. Fines removal is characterized by the classification function h_f, which ideally is a low-pass filter of the form

$$h_f(L) = \begin{cases} 0 & \text{if } L > L_f \\ R_f & \text{if } L \leq L_f \end{cases} \tag{3}$$

where R_f is the fines removal rate, and L_f is the fine size. Notice that when $R_p = R_f = 0$, particles are removed indifferently of size due to flow out of the crystallizer with rate q/V. The case $q = 0$ corresponds to batch mode, where the total mass in the suspension is preserved.

The growth rate $G(c(t))$ in (1) is assumed independent of crystal size L and depends on the concentration of solute $c(t)$ in the liquid phase. One often assumes a phenomenological formula

$$G(c(t)) = k_g \left(c(t) - c_s\right)^g, \tag{4}$$

where growth coefficient k_g and growth exponent g depend on the constituent, and where c_s is the saturation concentration, [5, 6, 10]. For theory it suffices to assume that G is locally Lipschitz with $G(c) > 0$ for supersaturation $c > c_s$, and $G(c) < 0$ for $c < c_s$, in which case crystals shrink.

The breakage integral on the right of (1) can be explained as follows. The term $a(L)$ represents the breakage rate, i.e., the probability that a particle of size L is broken into two particles of smaller sizes $\hat{L}, \hat{\hat{L}}$. The term $b(L, \hat{L})$ is the conditional probability that a particle of size L is broken onto two pieces of size $\hat{L}$ and $\hat{\hat{L}}$ with $\hat{L} \geq \hat{\hat{L}}$, where

$$L^3 = \hat{L}^3 + \hat{\hat{L}}^3, \tag{5}$$

assures that breakage does not change the overall crystal volume or mass, given that crystals are characterized by size L and have volume $k_v L^3$. This means we have a sink term and source term. The sink term gathers particles leaving size L by being broken down to smaller sizes $\hat{L} < L$. This leads to

$$\mathcal{Q}^-_{\text{break}}(L,t) = \int_{2^{-1/3}L}^{L} a(L) b(L,\hat{L}) n(L,t) d\hat{L} = a(L) n(L,t) \int_{2^{-1/3}L}^{L} b(L,\hat{L}) d\hat{L} = a(L) n(L,t),$$

as $b(L, \hat{L})$ is a probability density. The source term at size L has the form

$$\mathcal{Q}^+_{\text{break}}(L,t) = \int_{L}^{2^{1/3}L} a(\hat{L}) b(\hat{L}, L) n(\hat{L},t) d\hat{L} + \int_{2^{1/3}L}^{\infty} a(\hat{L}) b\left(\hat{L} \to \left(\hat{L}^3 - L^3\right)^{1/3}\right) n(\hat{L},t) d\hat{L},$$

representing particles broken down from all possible larger sizes $\hat{L} \geq L$ to size L. The left hand term counts those events where the larger particle has size L, the right hand term those where the particle of size L is the smaller one. In the event $b(L, \hat{L}) = b(L, (L^3 - \hat{L}^3)^{1/3})$, b will be symmetrized, so that the first integral has the form given on the right of (1).

Let us examine the mass balance of breakage. The total mass of crystals being broken is

$$m^-_{\text{break}}(t) = \int_0^\infty \mathcal{Q}^-_{\text{break}}(L,t) L^3 dL = \int_0^\infty a(L) n(L,t) L^3 \, dL.$$

On the other hand, the total mass of new crystals born due to breakage is

$$\begin{aligned} m^+_{\text{break}}(t) = \int_0^\infty \mathcal{Q}^+_{\text{break}}(L,t) L^3 dL &= \int_0^\infty \int_L^{2^{1/3}L} a(\hat{L}) b(\hat{L}, L) n(\hat{L},t) d\hat{L} L^3 dL \\ &\quad + \int_0^\infty \int_{2^{1/3}L}^{\infty} a(\hat{L}) b\left(\hat{L}, \left(\hat{L}^3 - L^3\right)^{1/3}\right) n(\hat{L},t) d\hat{L} L^3 dL \\ = \int_0^\infty a(\hat{L}) n(\hat{L},t) \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L}, L) L^3 dL d\hat{L} & \\ &\quad + \int_0^\infty a(\hat{L}) n(\hat{L},t) \int_0^{2^{-1/3}\hat{L}} b(\hat{L}, (\hat{L}^3 - L^3)^{1/3}) L^3 dL d\hat{L} \\ = \int_0^\infty a(\hat{L}) n(\hat{L},t) \left\{ \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L}, L) L^3 dL + \int_0^{2^{-1/3}\hat{L}} b(\hat{L}, (\hat{L}^3 - L^3)^{1/3}) L^3 dL \right\} d\hat{L} & \\ = \int_0^\infty a(\hat{L}) n(\hat{L},t) \hat{L}^3 dL = m^-_{\text{break}}(t), & \end{aligned}$$

at all times t, because the term $\{\dots\}$ equals

$$\begin{aligned}\{\dots\} &= \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)L^3dL + \int_0^{2^{-1/3}\hat{L}} b(\hat{L},(\hat{L}^3-L^3)^{1/3})L^3dL \\ &= \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)L^3dL + \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)\hat{\hat{L}}^3dL \\ &= \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)(L^3+\hat{\hat{L}}^3)dL = \hat{L}^3\int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)dL = \hat{L}^3.\end{aligned}$$

This confirms that breakage leaves the total crystal mass invariant. In contrast, if we compute the balance of number of individuals being broken, we obtain

$$\begin{aligned}\mathcal{Q}^+_{\text{break}}(t) &= \int_0^\infty \mathcal{Q}^+_{\text{break}}(L,t)dL = \int_0^\infty \int_L^{2^{1/3}L} a(\hat{L})b(\hat{L},L)n(\hat{L},t)d\hat{L}dL \\ &\qquad + \int_0^\infty \int_{2^{1/3}L}^\infty a(\hat{L})b\left(\hat{L},(\hat{L}^3-L^3)^{1/3}\right)n(\hat{L},t)d\hat{L}dL \\ &= \int_0^\infty a(\hat{L})n(\hat{L},t)\int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)dLd\hat{L} \\ &\qquad + \int_0^\infty a(\hat{L})n(\hat{L},t)\int_0^{2^{-1/3}\hat{L}} b(\hat{L},(\hat{L}^3-L^3)^{1/3})dLd\hat{L} \\ &= \int_0^\infty a(\hat{L})n(\hat{L},t)\left\{\int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)dL + \int_0^{2^{-1/3}\hat{L}} b(\hat{L},(\hat{L}^3-L^3)^{1/3})dL\right\}d\hat{L} \\ &= \int_0^\infty a(\hat{L})n(\hat{L},t)2dL = 2\mathcal{Q}^-_{\text{break}}(t),\end{aligned}$$

because the term $\{\dots\}$ equals

$$\{\dots\} = \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L}\to L)dL + \int_0^{2^{-1/3}\hat{L}} b(\hat{L},(\hat{L}^3-L^3)^{1/3})dL$$

$$= \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L}\to L)dL + \int_{2^{-1/3}\hat{L}}^{\hat{L}} b(\hat{L},L)dL = 2.$$

Not surprisingly, breakage doubles the total number of individuals of that part of the population which undergoes breakage.

Remark 1 *It is possible to classify particles by volume, as discussed in Hu, Rohani and Jutan ([28]). This requires organizing the balance equations accordingly, and simplifies the presentation of breakage, as for instance (5) becomes* $V = \hat{V} + \hat{\hat{V}}$. *Nonetheless we prefer to characterize particles by size* L, *as this allows to discuss first and second moments which describe the total crystal length and surface. In control applications [29] this allows for instance to address quantities like the weighted mean diameter* $d_{43} = \int_0^\infty n(L,t)L^4dL/\int_0^\infty n(L,t)L^3dL$, *the Sauter mean diameter* $d_{32} = \int_0^\infty n(L,t)L^3dL/\int_0^\infty n(L,t)L^2dL$ *[6, 8], and other quantities, which are not accessible in a model based on the unit* V.

Equation (1) goes along with initial and boundary conditions. The initial crystal distribution $n_0(L)$ is called the *seed*. The boundary condition $n(0,t) = B(c(t))/G(c(t))$ models birth of crystals at size $L = 0$ and is governed by the ratio B/G of birth rate $B(c)$ over growth rate $G(c)$. Again it is customary to assume a phenomenological law of the form

$$B(c(t)) = k_b\left((c(t)-c_s)_+\right)^b \tag{6}$$

for the birth rate, where k_b is the nucleation or birth coefficient, b the birth exponent, and $q_+ = \max\{0,q\}$. For theory it is enough to assume that B is locally Lipschitz with $B > 0$ for $c > c_s$ and $B = 0$ for $c \le c_s$, meaning that nucleation only takes place in a supersaturated suspension.

2.2. Mole balance equation

We now derive a second equation, which models the influence of various internal and external effects on the second population, the concentration $c(t)$ of solute molecules in the liquid. The so-called mole balance equation is obtained by investigating the mass balance within the crystallizer.

Table 1. Parameters.

Feed rate	q	0.05	l/min
Total volume	V	10.5	l
Fines removal cut size	L_f	0.2	mm
Product cut size	L_p	1	mm
Fines removal constant	R_f	5	–
Product removal constant	R_p	2	–
Growth rate constant	k_g	0.0305	$mm\,l/min\,mol$
Growth rate exponant	g	1	–
Nucleation rate constant	k_b	8.36×10^9	$l^3/min\,mol^4$
Nucleation rate exponant	b	4	–
KCl crystal density	ρ	1989	g/l
Mole mass KCl	M	74.551	g/mol
Volumetric shape factor	k_v	0.112	–
Saturation concentration	c_s	4.038	mol/l

Table 2. Size- and time-dependent quantities

quantity	symbol	unit
Crystal size distribution	$n(L,t)$	$\#/mm\,l$
Crystal seed	$n_0(L)$	$\sharp/m.m^3$
Crystal breakage rate	$a(L)$	$1/min$
Breakage type	$b(\hat{L},L)$	$1/mm$
Molar concentration of solute in liquid	$c(t)$	mol/l
Solute feed concentration	$c_f(t)$	mol/l

In this study we will consider the total volume V of the suspension as constant, which leads to the formula

$$\begin{aligned}\frac{dm_{\text{solute}}(t)}{dt} &= \frac{d(V\varepsilon c(t)M)}{dt} = V\frac{d\varepsilon(t)}{dt}c(t)M + V\varepsilon(t)\frac{dc(t)}{dt}M \\ &= qc_fM - qc(t)\varepsilon M - 3Vk_v\rho G(c(t))\int_0^\infty n(L,t)L^2dL + qk_v\rho\int_0^\infty h_f(L)n(L,t)L^3dL,\end{aligned} \tag{7}$$

where $c_f(t)$ is the feed concentration and $\varepsilon(t)$ is the void fraction (see also (51) in the appendix), which takes the form

$$\varepsilon(t) = 1 - k_v\int_0^\infty n(L,t)L^3dL. \tag{8}$$

Typical parameter values for KCl-crystallization are given in Table 1. By substituting (8) and its derivative into (7), the mass balance of solute in the liquid phase is finally obtained as

$$M\frac{dc(t)}{dt} = \frac{q(\rho - Mc(t))}{V} + \frac{\rho - Mc(t)}{\varepsilon(t)}\frac{d\varepsilon(t)}{dt} + \frac{qc_f(t)M}{V\varepsilon(t)} - \frac{q\rho}{V\varepsilon(t)}(1 + k_v\nu(t)) \tag{9}$$

with

$$\nu(t) = R_p\int_{L_p}^\infty n(L,t)L^3dL. \tag{10}$$

More details on how (9) is derived from the mass balance are given in the appendix.

3. Existence and uniqueness

In this chapter we develop our proof of local existence and uniqueness. This requires several preparatory steps. For convenience, let us recall the complete model described by the population and molar balances. The population balance equation is given by

$$\frac{\partial n(L,t)}{\partial t} = -G(c(t))\frac{\partial n(L,t)}{\partial L} - h(L)n(L,t) + w(L,t) \tag{11}$$

where

$$w(L,t) := \int_L^\infty a(\hat{L})b(\hat{L},L)n(\hat{L},t)d\hat{L} \tag{12}$$

is the source term due to breakage and attrition, and

$$h(L) := (q/V)(1+h_f(L)+h_p(L)) + a(L) > 0 \tag{13}$$

is the sink gathering all attenuating terms. The boundary value is given by

$$n(0,t) = \frac{B(c(t))}{G(c(t))}, \qquad t \geq 0 \tag{14}$$

and the initial condition is given by

$$n(L,0) = n_0(L), \qquad L \in [0,\infty) \tag{15}$$

coupled with the mole balance equation

$$\frac{dc(t)}{dt} = -\left(\frac{q}{V} + \frac{\epsilon'(t)}{\epsilon(t)}\right)c(t) + \frac{1}{\epsilon(t)}\left[\frac{qc_f(t)}{V} + \frac{\rho}{M}\epsilon'(t) - \frac{q\rho}{VM}(1+k_vR_p\nu(t))\right] + \frac{q\rho}{VM} \tag{16}$$
$$c(0) = c_0$$

where

$$\epsilon(t) = 1 - k_v\int_0^\infty n(L,t)L^3\,dL \qquad \text{and} \qquad \nu(t) = \int_0^\infty h_p(L)n(L,t)L^3 dL.$$

3.1. Moments and characteristics

In this part we present a transformation of the model using moments and characteristic curves. Introducing the third moment $\mu(t) = \int_0^\infty n(L,t)L^3\,dL$, we have $\epsilon(t) = 1 - k_v\mu(t)$ and $\epsilon'(t) = -k_v\mu'(t)$.

Lemma 3.1 *Suppose the function $c(t)$ with $c(0) = c_0 > 0$ satisfies the mole balance equation (16). Then*

$$c(t) = e^{\int_0^t -\left(\frac{q}{V}+\frac{-k_v\mu'(\tau)}{1-k_v\mu(\tau)}\right)d\tau}\left(c_0 + \int_0^t\left[\frac{1}{1-k_v\mu(\tau)}\left[\frac{qc_f(\tau)}{V} + \frac{\rho}{M} - k_v\mu'(\tau) - \frac{q\rho}{VM}(1+k_v\nu(\tau))\right] + \frac{q\rho}{VM}\right]\right.$$
$$\left.\times e^{\int_0^\tau\left(\frac{q}{V}+\frac{-k_v\mu'(s)}{1-k_v\mu(s)}\right)ds}d\tau\right). \tag{17}$$

Proof By solving (16) with respect to $c(t)$ using variation of the constant, we obtain

$$c(t) = e^{\int_0^t -\left(\frac{q}{V}+\frac{\epsilon'(\tau)}{\epsilon(\tau)}\right)d\tau}\left(c_0 + \int_0^t\left[\frac{1}{\epsilon(\tau)}\left[\frac{qc_f(\tau)}{V} + \frac{\rho}{M}\epsilon'(\tau) - \frac{q\rho}{VM}(1+k_vR_p\nu(\tau))\right] + \frac{q\rho}{VM}\right]e^{\int_0^\tau\left(\frac{q}{V}+\frac{\epsilon'(s)}{\epsilon(s)}\right)ds}d\tau\right). \tag{18}$$

Now using $\epsilon(t) = 1 - k_v\mu(t)$ and $\epsilon'(t) = -k_v\mu'(t)$, (18) transforms into

$$c(t) = e^{\int_0^t -\left(\frac{q}{V}+\frac{-k_v\mu'(\tau)}{1-k_v\mu(\tau)}\right)d\tau}\left(c_0 + \int_0^t\left[\frac{1}{1-k_v\mu(\tau)}\left[\frac{qc_f(\tau)}{V} + \frac{\rho}{M} - k_v\mu'(\tau) - \frac{q\rho}{VM}(1+k_v\nu(\tau))\right] + \frac{q\rho}{VM}\right]\right.$$
$$\left.\times e^{\int_0^\tau\left(\frac{q}{V}+\frac{-k_v\mu'(s)}{1-k_v\mu(s)}\right)ds}d\tau\right). \tag{19}$$

□

Next we introduce characteristic curves. For t_0 and L_0 fixed we let ϕ_{t_0,L_0} be the solution of the initial value problem

$$\frac{d\phi(t)}{dt} = G(c(t)), \quad \phi(t_0) = L_0.$$

Since the right hand side does not depend on L, we have explicitly

$$\phi_{t_0,L_0}(t) = L_0 + \int_{t_0}^t G(c(\tau))d\tau. \tag{20}$$

We write specifically $z(t) := \phi_{0,0}(t)$. Now we introduce a family of functions $N_{t,L}$ which we use later to define $n(L,t)$ via $N_{t_0,L_0}(t) := n(\phi_{t_0,L_0}(t),t)$. If we let $L = \phi_{t_0,L_0}(t)$, then N_{t_0,L_0} satisfies

$$\frac{dN_{t_0,L_0}(t)}{dt} = \frac{\partial n(L,t)}{\partial L}\phi'_{t_0,L_0}(t) + \frac{\partial n(L,t)}{\partial t} = \frac{\partial n(L,t)}{\partial L}G(c(t)) + \frac{\partial n(L,t)}{\partial t}.$$

Therefore (11) transforms into

$$\frac{dN_{t_0,L_0}(t)}{dt} = -h(\phi_{t_0,L_0}(t))N_{t_0,L_0}(t) + w(\phi_{t_0,L_0}(t),t), \tag{21}$$

and we consequently use these ODEs to define the functions $N_{t,L}$. Integration of (21) gives

$$N_{t_0,L_0}(t) = \left(N_{t_0,L_0}(t_0) + \int_{t_0}^{t} w(\phi_{t_0,L_0}(\tau),\tau)\exp\left\{\int_{t_0}^{\tau} h(\phi_{t_0,L_0}(\sigma))d\sigma\right\}d\tau\right)\exp\left\{-\int_{t_0}^{t} h(\phi_{t_0,L_0}(\tau)d\tau\right\}. \tag{22}$$

We can exploit this for two possible situations, where $N_{t_0,L_0}(t_0)$ can be given an appropriate value.

Before putting this to work, we will need two auxiliary functions τ and ξ, which are easily defined using the characteristics. First we define $\tau = \tau(t,L)$ implicitly by

$$\phi_{\tau,0}(t) = L, \text{ or equivalently, } \phi_{t,L}(\tau) = 0, \tag{23}$$

or again,

$$\int_{\tau(t,L)}^{t} G(c(\sigma))\,d\sigma = L. \tag{24}$$

Then we define $\xi = \xi(t,L) = \phi_{t,L}(0)$, which gives $\xi = L + \displaystyle\int_t^0 G(c(\tau))\,d\tau$.

Definition 3.1 *We define a candidate solution $n(L_0,t_0)$ of the population balance equation (1) by the formula*

$$n(L_0,t_0) = \begin{cases} \left(\dfrac{B(c(\tau_0))}{G(c(\tau_0))} + \displaystyle\int_{\tau_0}^{t_0} w(\phi_{\tau_0,0}(s),s)\exp\left\{\int_{\tau_0}^{s} h(\phi_{\tau_0,0}(\sigma))d\sigma\right\}ds\right) \\ \qquad\qquad \times\exp\left(-\displaystyle\int_{\tau_0}^{t_0} h(\phi_{\tau_0,0}(s))\,ds\right), \text{ if } L_0 < z(t_0) \\ \\ \left(n_0(\phi_{t_0,L_0}(0)) + \displaystyle\int_{0}^{t_0} w(\phi_{t_0,L_0}(s),s)\exp\left\{\int_{0}^{s} h(\phi_{t_0,L_0}(\sigma))d\sigma\right\}ds\right) \\ \qquad\qquad \times\exp\left(-\displaystyle\int_{0}^{t_0} h(\phi_{t_0,L_0}(s))\,ds\right), \text{ if } L_0 \geq z(t_0) \end{cases} \tag{25}$$

where on the right hand side the construction uses $N_{t,L}$, respectively, (22), and where $\tau_0 = \tau(t_0,L_0)$, $L_0 = \phi(t_0)$, $z(t_0) = \phi_{0,0}(t_0)$ and $t_0 \in [0,T]$.

The formula is justified as follows. Let t_0, L_0 be such that $L_0 < z(t_0) = \phi_{0,0}(t_0)$. This is the case where $\tau_0 = \tau(t_0,L_0) > 0$. Here we consider equation (21) for $N_{\tau_0,0}$ with initial value $N_{\tau_0,0}(\tau_0) = n(\phi_{\tau_0,0}(\tau_0),\tau_0) = n(0,\tau_0) = B(c(\tau_0))/G(c(\tau_0))$. This uses the fact that $\phi_{\tau_0,0}(\tau_0) = 0$ according to the definition of $\phi_{\tau,0}$. Integration clearly gives the upper branch of (25).

Next consider t_0, L_0 such that $L_0 \geq z(t_0)$. Then $\tau_0 < 0$, so that we do not want to use it as initial value. We therefore apply (21), (22) to N_{t_0,L_0}, now with initial time 0. Then we get

$$N_{t_0,L_0}(t_0) = \left(N_{t_0,L_0}(0) + \int_0^{t_0} w(\phi_{t_0,L_0}(s),s)\exp\left\{\int_0^s h(\phi_{t_0,L_0}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_0^{t_0} h(\phi_{t_0,L_0}(s))\,ds\right).$$

Here $N_{t_0,L_0}(0) = n(\phi_{t_0,L_0}(0),0) = n_0(\phi_{t_0,L_0}(0))$, so we get the lower branch of (25) all right. This justifies formula (25).

By using moments, c, $\phi_{t,L}$ and $n(L,t)$ defined by (19), (20), (25), we will represent the problem of existence and uniqueness of the solution of (11), (16) as a fixed-point problem for an operator $\mathcal{Q}$, which we define subsequently. The operator acts on triplets (w,μ,ν) and shall be defined successively according to the following diagram:

$$\mathcal{Q} : (w,\mu,\nu) \xrightarrow{(19)} c \xrightarrow{(20)} \phi_{t,L} \xrightarrow{(25)} n(L,t) \xrightarrow{(32),(28),(29),(31)} (\widetilde{w},\widetilde{\mu},\widetilde{\nu}). \tag{26}$$

We will then prove that $\mathcal{Q}$ is a self-map and a contraction with respect to specific metric on a suitably defined space X_T, so that it has a fixed point, which will provide the solution of the model (11)-(16).

3.2. Closing the cycle

In this section we prove several facts which we will need later. Our first step is to close the cycle and complete the definition of $\mathcal{Q}$, which requires passing from $n(L,t)$, defined via (25), back to the moment functions μ, ν. At least we hope to get back to μ, ν via a fixed-point argument. Since this is the objective of the proof, we need to give new names to the moment functions defined via $n(L,t)$, and we will call them $\widetilde{\mu}$, $\widetilde{\nu}$, and $\widetilde{w}$, and this will complete the definition of $\mathcal{Q}$.

Integration of (25) with respect to $L^3 dL$ is cut into two steps. Fixing $t \geq 0$, we first integrate $n(L,t)L^3$ form $L=0$ to $L = z(t) = \phi_{0,0}(t) > 0$, and then from $L = z(t)$ to infinity. In order to be allowed to do this, we need integrability hypotheses on $w(L,t)$. We assume that $w \in E$, where E is the Banach space

$$E = W_u^{1,\infty}(\mathbb{R}^+ \times [0,T]) \cap \mathcal{L}_u^1(\mathbb{R}^+ \times [0,T], L^2 dL) \cap \mathcal{L}_u^1(\mathbb{R}^+ \times [0,T], L^3 dL).$$

Specifically, as indicated by the subscript u, every $w \in E$ is uniformly Lipschitz continuous in the first coordinate by use of the norm $\|w\|_\infty + \|w\|_L$ with

$$\|w\|_L = \sup_{0 \leq t \leq T} \sup_{L \neq \hat{L}} \frac{|w(L,t) - w(\hat{L},t)|}{|L - \hat{L}|}.$$

Similarly, the $\mathcal{L}_u^1$-norms are understood in the sense

$$\|w\|_1 = \sup_{0 \leq t \leq T} \int_0^\infty |w(L,t)| L^2 dL < \infty, \quad |||w|||_1 = \sup_{0 \leq t \leq T} \int_0^\infty |w(L,t)| L^3 dL < \infty.$$

Integration of (25) with respect to $L^3 dL$ then gives

$$\begin{aligned}
\widetilde{\mu}(t) &= \int_0^\infty n(L,t) L^3 dL \\
&= \int_0^{z(t)} \left(\frac{B(c(\tau))}{G(c(\tau))} + \int_\tau^t w(\phi_{\tau,0}(s),s) \exp\left\{ \int_\tau^s h(\phi_{\tau,0}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_\tau^t h(\phi_{\tau,0}(s))\, ds \right) L^3 dL \\
&+ \int_{z(t)}^\infty \left(n_0(\phi_{t,L}(0)) + \int_0^t w(\phi_{t,L}(s),s) \exp\left\{ \int_0^s h(\phi_{t,L}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_0^t h(\phi_{t,L}(s))\, ds \right) L^3 dL.
\end{aligned} \tag{27}$$

In the first integral $\int_0^{z(t)}$ we use the change of variables $L \to \tau = \tau(t,L)$. That means

$$[0, z(t)] \ni L \mapsto \tau(t,L) \in [0,t], \qquad dL = G(c(t)) d\tau.$$

In the second integral $\int_{z(t)}^\infty$ we use the change of variables $L \to \xi(t,L) := \phi_{t,L}(0)$. Then

$$[z(t), \infty) \ni L \mapsto \xi \in [0, \infty), \qquad dL = d\xi.$$

The inverse relation is $L = \xi + \displaystyle\int_0^t G(c(\sigma)) d\sigma = \xi + z(t)$. From (27) we obtain

$$\begin{aligned}
\widetilde{\mu}(t) = &\int_0^t \left(B(c(\tau)) + \int_\tau^t w(\phi_{\tau,0}(s),s) \exp\left\{ \int_\tau^s h(\phi_{\tau,0}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_\tau^t h(\phi_{\tau,0}(s))\, ds \right) L(\tau)^3\, d\tau \\
&+ \int_0^\infty \left(n_0(\xi) + \int_0^t w(\phi_{0,\xi}(s),s) \exp\left\{ \int_0^s h(\phi_{0,\xi}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_0^t h(\phi_{0,\xi}(s))\, ds \right) L(\xi)^3 d\xi,
\end{aligned} \tag{28}$$

where $L(\tau) = \int_\tau^t G(c(\sigma))\, d\sigma$ and $L(\xi) = \xi + \int_0^t G(c(\sigma))\, d\sigma$, and where we use $\phi_{t,L}(s) = \phi_{0,\xi}(s)$ in the second integral. For fixed t the functions $L \mapsto \tau(t,L)$ and $\tau \mapsto L(\tau)$ are inverses of each other, and similarly, $L \leftrightarrow \xi$ are in one-to-one correspondence via the formula $L = \xi - \int_0^t G(c(\sigma))\, d\sigma$. A formula similar to (28) is obtained for $\widetilde{\nu}$:

$$\begin{aligned}
\widetilde{\nu}(t) = &\int_0^t \left(B(c(\tau)) + \int_\tau^t w(\phi_{\tau,0}(s),s) \exp\left\{ \int_\tau^s h(\phi_{\tau,0}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_\tau^t h(\phi_{\tau,0}(s))\, ds \right) h_p(\tau) L(\tau)^3\, d\tau \\
&+ \int_0^\infty \left(n_0(\xi) + \int_0^t w(\phi_{0,\xi}(s),s) \exp\left\{ \int_0^s h(\phi_{0,\xi}(\sigma)) d\sigma \right\} ds \right) \exp\left(- \int_0^t h(\phi_{0,\xi}(s))\, ds \right) h_p(\xi) L(\xi)^3 d\xi.
\end{aligned} \tag{29}$$

Concerning $\widetilde{\mu}$, we need to represent its derivative. Starting with

$$\widetilde{\mu}'(t) = \int_0^\infty \frac{\partial n(L,t)}{\partial t} L^3\, dL$$

and substituting the population balance equation (11) gives

$$\begin{aligned}\widetilde{\mu}'(t) &= -\int_0^\infty \left[G(c(t))\frac{\partial n(L,t)}{\partial L} + h(L)n(L,t) - w(L,t)\right] L^3 dL \\ &= 3G(c(t))\int_0^\infty n(L,t)L^2\,dL - \int_0^\infty h(L)n(L,t)L^3\,dL + \int_0^\infty w(L,t)L^3\,dL \end{aligned} \tag{30}$$

via integration by parts. Now we can substitute the expression (25) for $n(L,t)$. Using the same strategy of integration $[0, z(t)]$ followed by $[z(t), \infty)$ as in (28) we obtain

$$\begin{aligned}\widetilde{\mu}'(t) = {} & 3G(c(t))\int_0^t \left(B(c(\tau)) + \int_\tau^t w(\phi_{\tau,0}(s),s)\exp\left\{\int_\tau^s h(\phi_{\tau,0}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_\tau^t h(\phi_{\tau,0}(s))\,ds\right)L(\tau)^2\,d\tau \\ & + 3G(c(t))\int_0^\infty \left(n_0(\xi) + \int_0^t w(\phi_{0,\xi}(s),s)\exp\left\{\int_0^s h(\phi_{0,\xi}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_0^t h(\phi_{0,\xi}(s))\,ds\right)L(\xi)^2 d\xi \\ & - \int_0^t h(\phi_{\tau,0}(t))\left(B(c(\tau)) + \int_\tau^t w(\phi_{\tau,0}(s),s)\exp\left\{\int_\tau^s h(\phi_{\tau,0}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_\tau^t h(\phi_{\tau,0}(s))\,ds\right)L(\tau)^3 d\tau \\ & - \int_0^\infty h(\phi_{t,L}(t))\left(n_0(\xi) + \int_0^t w(\phi_{0,\xi}(s),s)\exp\left\{\int_0^s h(\phi_{0,\xi}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_0^t h(\phi_{0,\xi}(s))\,ds\right)L(\xi)^3 d\xi \\ & + G(c(t))\int_0^t w(\phi_{\tau,0}(t),t)L(\tau)^3 d\tau + \int_0^\infty w(\phi_{0,\xi}(t),t)L(\xi)^3 d\xi. \end{aligned} \tag{31}$$

This representation, which can also be obtained by a direct differentiation of (28), shows that $\widetilde{\mu}'$ is a continuous function if we assume that $n_0(L)$ and h are continuous, and by continuity of $w \in E$.

In (28), (29) and (31) we can now substitute the expression $B(c(t)) = k_b(c(t) - c_s)_+^b$ using (18). This means the moments $\widetilde{\mu}, \widetilde{\nu}$, and also $\widetilde{\mu}'$, are defined by way of the characteristics, which are by themselves defined via the original moments μ, ν, closing the cycle.

We also need to get back to the function $w(L,t)$. The element obtained by closing the cycle will be denoted $\widetilde{w}(L,t)$, and the fixed-point argument will have to show $w = \widetilde{w}$, just as for the moments. We introduce

$$\beta(L,\hat{L}) = \begin{cases} a(L)b(L \to \hat{L}), & \text{if } L \geq \hat{L} \\ 0, & \text{else} \end{cases}$$

then we can write

$$\widetilde{w}(\hat{L},t) = \int_{\hat{L}}^\infty a(L)b(L,\hat{L})n(L,t)dL = \int_0^\infty \beta(L,\hat{L})n(L,t)dL,$$

which is essentially like the moment integral (28), the function L^3 being replaced by $\beta(L,\hat{L})$. Applying the same technique as in the case of (28), we obtain

$$\begin{aligned}\widetilde{w}(\hat{L},t) = {} & \int_0^t \left(B(c(\tau)) + \int_\tau^t w(\phi_{\tau,0}(s),s)\exp\left\{\int_\tau^s h(\phi_{\tau,0}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_\tau^t h(\phi_{\tau,0}(s))\,ds\right)\beta(L(\tau),\hat{L})\,d\tau \\ & + \int_0^\infty \left(n_0(\xi) + \int_0^t w(\phi_{t,L}(s),s)\exp\left\{\int_0^s h(\phi_{t,L}(\sigma))d\sigma\right\}ds\right)\exp\left(-\int_0^t h(\phi_{t,L}(s))\,ds\right)\beta(L(\xi),\hat{L})d\xi, \end{aligned} \tag{32}$$

which expresses $\widetilde{w}$ in terms of (w,μ,ν). In particular, this shows continuity of $\widetilde{w}$. For convenience we again summarize the construction of $\mathcal{Q}$:

$$(w,\mu,\mu',\nu) \xrightarrow{(19)} c \xrightarrow{(20)} \phi_{t,L} \xrightarrow{(25)} n(L,t) \xrightarrow{(32),(28),(29),(31)} (\widetilde{w},\widetilde{\mu},\widetilde{\mu}',\widetilde{\nu}). \tag{33}$$

This means we can now represent the problem of existence and uniqueness of the solution of (11), (16) as a fixed-point problem for (33). Note that $\mathcal{Q}$ as self-mapping of the Banach space $E \times C^1[0,T] \times C[0,T]$, with arguments $w \in E$, $\mu \in C^1[0,T]$, $\nu \in C[0,T]$.

Let us make the following assumptions: n_0 is continuous, $n_0 \geq 0$, and

$$(H_1) \qquad \begin{aligned} &\mu_{0,\infty} := \max_{0\leq L<\infty} n_0(L) < +\infty, \quad \mu_{0,1} := \int_0^\infty n_0(L)dL < +\infty, \\ &\mu_{0,2} := \int_0^\infty n_0(L)L^2 dL < +\infty, \quad \mu_{0,3} := \int_0^\infty n_0(L)L^3 dL < +\infty. \end{aligned}$$

Lemma 3.2 *Under the above hypotheses, suppose $w \in E$, $\mu \in C^1[0,T]$, $\nu \in C[0,T]$ are given functions. Define $\widetilde{w}$, $\widetilde{\mu}$ and $\widetilde{\nu}$ via (32), (28), (29) using the construction in (33). Then $\widetilde{w} \in E$, $\widetilde{\mu} \in C^1[0,T]$ and $\widetilde{\nu} \in C[0,T]$.*

Proof As $n_0(L)$ is continuous by hypothesis, $\int_0^\infty n_0(L)L^2 dL < +\infty$, $\int_0^\infty n_0(L)L^3 dL < +\infty$ and $w \in E$, we have $\widetilde{w} \in E$.

The statement is clear for $\widetilde{\nu}$, which is represented explicitly using the right hand side of (29). The representation of $\widetilde{\mu}'$ given by (31) shows that $\widetilde{\mu}'$ is a continuous function. Then $\widetilde{\mu} \in C^1[0,T]$. □

3.3. Setting up the space

In this section we define a space X_T on which $\mathcal{Q}$, defined through (33), acts as a self map and a contraction. Let us start with some hypotheses, giving rise to suitable constants. We assume continuity of a and b and that

$$(H_2) \qquad \|a\|_\infty := \max_{0\le L<\infty} a(L) < +\infty, \qquad \|a\|_L = \sup_{0\le L<\hat{L}} \left| \frac{a(L)-a(\hat{L})}{L-\hat{L}} \right| < \infty,$$

and

$$(H_3) \qquad \|b\|_\infty := \max_{0\le L\le\hat{L}} b(\hat{L},L) < +\infty, \quad \|b\|_L := \sup_{L\ge 0} \sup_{L\le\hat{L}<\hat{\hat{L}}} \left| \frac{b(L,\hat{L})-b(L,\hat{\hat{L}})}{\hat{L}-\hat{\hat{L}}} \right| < \infty.$$

Let us now consider initial conditions. As we expect the function $w\in E$ to satisfy (32), it should satisfy this at $t=0$, which leads to the initial condition $w(L,0)=\int_L^\infty a(\hat{L})b(\hat{L},L)n_0(\hat{L})d\hat{L} =: w_{L,0} \le J$ for all L.

As $\mu\in C^1[0,T]$ is expected to be the moment function of $n(L,t)$ and to coincide with $\widetilde{\mu}$, it must satisfy the initial condition $\mu(0)=\int_0^\infty n_0(L)L^3\,dL=\mu_{0,3}$, and similarly $\nu(0)=\int_0^\infty n_0(L)h_p(L)L^3\,dL =: \nu_{0,3}$ for $\nu\in C[0,T]$. For μ' we have to put

$$\mu'(0)=3G(c_0)\int_0^\infty n_0(L)L^2dL - \int_0^\infty h(L)n_0(L)L^3dL + \int_0^\infty w(L,0)L^3dL =: \mu'_0,$$

We have

Lemma 3.3 *Suppose μ,μ',ν,w satisfy the above initial conditions. Let $\widetilde{w}$, $\widetilde{\mu}$, $\widetilde{\nu}$ be defined as the right hand sides of (32), (28) and (29), $\widetilde{\mu}'$ by the right hand side of (31). Then $\widetilde{w}(L,0)=w_{L,0}$, $\widetilde{\mu}(0)=\mu_{0,3}$, $\widetilde{\nu}(0)=\nu_{0,3}$, and $\widetilde{\mu}'(0)=\mu'_0$.*

Proof Passing to the limit $t\to 0^+$ in (28), (31), (29) and (32) shows that the initial conditions for $\widetilde{\mu}$, $\widetilde{\mu}'$ $\widetilde{\nu}$ and $\widetilde{w}$ are satisfied. □

In other words, the operator $\mathcal{Q}$ respects initial values. This suggest defining the following subset X_T of $E\times C^1[0,T]\times C[0,T]$.

$$\begin{aligned} X_T = &\{(w,\mu,\nu)\in E\times C^1[0,T]\times C[0,T] : 0\le w(L,t)\le J \text{ for all } L\ge 0 \quad \text{and} \quad 0\le t\le T, |\mu'(t)|\le K', 0\le \mu(t)\le K, \\ &0\le \nu(t)\le R_p\mu(t) \;\text{ for all } 0\le t\le T, w(L,0)=w_{L,0}, \mu(0)=\mu_{0,3}, \mu'(0)=\mu'_0, \nu(0)=\nu_{0,3}\}. \end{aligned}$$

The idea is now to adjust $T>0$, and $J>0$, $K>0$, $K'>0$ such that $\mathcal{Q}: X_T\to X_T$ becomes a contraction. This involves two steps. First we have to assure that $\mathcal{Q}(X_T)\subset X_T$, and then we have to prove contractibility.

To show $\mathcal{Q}(X_T)\subset X_T$, we clearly need $\mu_{0,3}<K$ and $\nu_{0,3}\le\mu_{0,3}$. Notice next that $0\le\mu\le K$ implies

$$\frac{1}{\epsilon(t)}=\frac{1}{1-k_v\mu(t)}\le\frac{1}{1-k_vK},$$

so we would like to choose K such that $k_vK<1$. This is possible as long as we have

$$(H_4) \qquad \mu_{0,3}k_v<1, \qquad c_f(t)\le c_{fT} := \max_{0\le t\le T}|c_f(t)|<\infty \quad \text{for} \quad t\in[0,T].$$

Notice that this is equivalent to $\epsilon(0)>0$, which is perfectly reasonable physically. We also make the assumption

$$(H_5) \qquad \|h_{f,p}\|_\infty := \max_{0\le L<\infty} h_{f,p}(L) < +\infty, \qquad \|h_{f,p}\|_L = \sup_{0\le L<\hat{L}} \frac{|h_{f,p}(L)-h_{f,p}(\hat{L})|}{|L-\hat{L}|} < \infty,$$

where $h_{f,p}$ stands for h_f or h_p. As a consequence of (H_5) and (H_2), the function $h=(q/V)(1+h_f+h_p)+a$ also satisfies $\|h\|_\infty<\infty$ and $\|h\|_L<\infty$.

Remark 2 *Note that (H_5) is a realistic hypothesis, but is not satisfied for the ideal high and low pass filters (2) and (3). It is easy to extend our result to piecewise Lipschitz functions $h_{f,p}$ in order to include the ideal filters formally, but in order to keep things simple, we use (H_5).*

3.4. The main result

In this section we present the main result of local existence and uniqueness. Let us start by defining the complete metric. Observe that a Banach space norm on $E\times C^1[0,T]\times C[0,T]$ is

$$\big|m\big| = \|w\|_L + ||w||_1 + |||w|||_1 + \|\mu'\|_\infty + |\mu(0)| + \|\nu\|_\infty,$$

where $m = (w, \mu, \nu) \in E \times C^1[0,T] \times C[0,T]$ and $\|w\|_L = \sup_{t\in[0,T], L\neq\hat{L}} |w(L,t) - w(\hat{L},t)|/|L-\hat{L}|$, $\|w\|_1 = \sup_{t\in[0,T]} \int_0^\infty w(L,t)L^2 dL$, $|||w|||_1 = \sup_{t\in[0,T]} \int_0^\infty w(L,t)L^3 dL$. Since $\mu(0)$ is fixed for elements $m = (w,\mu,\nu) \in X_T$, we obtain a metric $\mathrm{dist}(m_1, m_2)$ on X_T by

$$\mathrm{dist}(m_1, m_2) = \|w_1 - w_2\|_L + \|w_1 - w_2\|_1 + |||w_1 - w_2|||_1 + \|\mu_1' - \mu_2'\|_\infty + \|\nu_1 - \nu_2\|_\infty, \tag{34}$$

where $m_i = (w_i, \mu_i, \nu_i) \in X_T$, $i = 1, 2$. Completeness of the metric follows from closedness of X_T in $E \times C^1[0,T] \times C[0,T]$.

Theorem 3.1 *Suppose $(H_1) - (H_5)$ are satisfied. Then $T > 0$ can be chosen sufficiently small so that $\mathcal{Q} : X_T \to X_T$ is a self-map and a contraction with respect to the metric (34) on X_T. Consequently, $\mathcal{Q}$ has a fixed point (w^*, μ^*, ν^*) on X_T.*

Proof 1) Proving self-map
As a consequence of $m \in X_T$ we have $|\epsilon'(t)| = k_v|\mu'(t)| \le k_v K'$ on $[0,T]$. Using these bounds on ϵ, ϵ', we get

$$c(t) \le c_T := \left(c_0 + T\frac{1}{1-k_vK}\left(\frac{qc_{fT}}{V} + \frac{\rho}{M} + k_vK'\right) + T\frac{q\rho}{VM}\right) \exp\left\{\frac{2Tk_vK'}{1-k_vK}\right\} \tag{35}$$

for $t \in [0,T]$. Note that we have convergence $c_T \to c_0$ as $T \to 0^+$.

From (35) we obtain $B(c(t)) \le k_b(c_T - c_s)^b =: B_T$ on $[0,T]$, where $B_T \to k_b(c_0 - c_s)^b$ as $T \to 0^+$. This leads to the estimate

$$\begin{aligned}\widetilde{\mu}(t) \le &\int_0^t B_T L(\tau)^3 d\tau + \int_0^t \int_\tau^t w(\phi_{\tau,0}(s), s) ds L(\tau)^3 d\tau \\ &+ \int_0^\infty n_0(\xi) L(\xi)^3 d\xi + \int_0^\infty \int_0^t w(\phi_{t,L}(s), s) e^{\int_t^s h(\phi_{0,\xi}(\sigma))d\sigma} ds L(\xi)^3 d\xi \\ &\le T(B_T L(T)^3 + \|w\|_\infty L(T)^3) + \mu_{0,3} + e^{T\|h\|_\infty} T|||w|||_1 =: k_1(T)\end{aligned} \tag{36}$$

on $[0,T]$, with $|||w|||_1 = \sup_{0\le t\le T} \int_0^\infty w(L,t)L^3 dL < \infty$ because of $w \in E$. Therefore, $k_1(T) \to \mu_{0,3}$ as $T \to 0^+$. Since $\mu_{0,3} < K$, we can arrange $\widetilde{\mu}(t) < K$ for all $t \in [0,T]$ by choosing $T > 0$ sufficiently small.

Concerning $\widetilde{\nu}$, the expressions (28) and (29) clearly imply $\widetilde{\nu} \le R_p\widetilde{\mu}$. Now let us estimate $\widetilde{\mu}'$. Passing to the limit in (30) and using continuity of $\widetilde{\mu}'$, established through (31), yields

$$\widetilde{\mu}'(t) \to 3G(c_0)\mu_{0,2} - \int_0^\infty h(L)n_0(L)L^3 dL + \int_0^\infty w(L,0)L^3 dL = \mu_0'$$

as $t \to 0^+$. Therefore, as soon as $-K' < \mu_0' < K'$, we can choose $T > 0$ sufficiently small to guarantee $-K' \le \widetilde{\mu}'(t) < K'$ on $[0,T]$. Finally, for $\widetilde{w}$ we also have $\widetilde{w}(L,t) \to w(L,0) = w_{L,0}$ as $t \to 0^+$ uniformly over $L \in [0,\infty)$, so it suffices to choose $J > w_{L,0}$ for all L. We have to show that $\widetilde{w} \in E$, and in particular, $|\widetilde{w}(\hat{\hat{L}}, t) - \widetilde{w}(\hat{L}, t)| \le C|\hat{\hat{L}} - \hat{L}|$. This follows from the uniform Lipschitz property of β with respect to the second variable in hypothesis (H_3). Altogether we have proved the following fact:

Suppose $(H_1) - (H_5)$ are satisfied, then we can fix $K > \mu_0$ such that $k_vK < 1$. Suppose further that $-K' < \mu_0' < K'$. Then $\mathcal{Q}(X_T) \subset X_T$ for $T > 0$ sufficiently small.

2) Proving contractibility
We now proceed to the core of the proof, where we show that $\mathcal{Q}$ is a contraction with respect to the metric (34) on X_T. By applying the Banach fixed-point principle we will then ultimately conclude.

By using the metric (34), we intend to prove $\mathrm{dist}(\mathcal{Q}(m_1), \mathcal{Q}(m_2)) \le \gamma\, \mathrm{dist}(m_1, m_2)$ for some $0 < \gamma < 1$. Let $m_1 = (w_1, \mu_1, \nu_1)$, $m_2 = (w_2, \mu_2, \nu_2) \in X_T$. Let c_1, c_2 be the corresponding expressions (19). Obtain the characteristic curves $\phi^1_{t,L}$, $\phi^2_{t,L}$ from (20). Define $N^1_{t,L}(s) = n(\phi^1_{t,L}(s), s)$, and similarly $N^2_{t,L}$. Finally, define $\widetilde{w}_1$, $\widetilde{w}_2$ via formula (32), $\widetilde{\mu}_1$, $\widetilde{\mu}_2$ via formula (28), $\widetilde{\mu}_1{}'$, $\widetilde{\mu}_2'$ via (31), and $\widetilde{\nu}_1, \widetilde{\nu}_2$ via (29). We have to prove an estimate of the form $\|\widetilde{\mu}_1' - \widetilde{\mu}_2'\|_\infty \le T\, c\, \mathrm{dist}(m_1, m_2)$ and similar ones for each of the six norm expressions in (34). By choosing $T > 0$ sufficiently small, we will then get a space X_T such that $\mathcal{Q} : X_T \to X_T$ is a contraction with respect to the distance (34).

(i) Estimating $c_1 - c_2$
The key to prove our contractibility estimates is of course to prove this first for the building elements, that is $c_1 - c_2$, $\phi^1 - \phi^2$, etc., as those arise in the moment expressions. We start by estimating $c_1 - c_2$.

From (19) we get a decomposition $c(t) = a(t) + b(t)$ with

$$a(t) = c_0 \exp\left\{-\int_0^t \left(\frac{q}{V} - \frac{k_v\mu'(\tau)}{1-k_v\mu(\tau)}\right)\right\} \tag{37}$$

$$b(t) = \int_0^t \left[\frac{1}{1-k_v\mu(\tau)}\left[\frac{qc_f(\tau)}{V} + \frac{\rho}{M} - k_v\mu'(\tau) - \frac{q\rho}{VM}(1 + k_v\nu(\tau))\right] + \frac{q\rho}{VM}\right] \exp\left\{\int_t^\tau \left(\frac{q}{V} - \frac{k_v\mu'(s)}{1-k_v\mu(s)}\right) ds\right\} d\tau. \tag{38}$$

We estimate (37) and (38) separately. Let us examine (37). We have

$$a_1(t)-a_2(t)=c_0\exp\left(-\int_0^T \frac{q}{V}d\tau\right)\left[\exp\int_0^t \frac{k_v\mu_1'(\tau)}{1-k_v\mu_1(\tau)}d\tau-\exp\int_0^t \frac{k_v\mu_2'(\tau)}{1-k_v\mu_2(\tau)}d\tau\right]$$

hence $|a_1(t)-a_2(t)|\le c_0|e^{A_1(t)}-e^{A_2(t)}|$. Now we use the estimate $|e^a-e^b|\le\max\{e^a,e^b\}|a-b|$ to obtain

$$|a_1(t)-a_2(t)|\le c_0\max\left\{e^{A_1(t)},e^{A_2(t)}\right\}|A_1(t)-A_2(t)|.$$

We therefore have to estimate the maximum $\max\{e^{A_1},e^{A_2}\}$ and $|A_1-A_2|$. We find

$$\begin{aligned}|A_1(t)-A_2(t)| &\le \int_0^t\left(\frac{k_v}{1-k_v\mu_1(\tau)}|\mu_1'(\tau)-\mu_2'(\tau)|+k_v|\mu_2'(\tau)|\left|\frac{1}{1-k_v\mu_1(\tau)}-\frac{1}{1-k_v\mu_2(\tau)}\right|\right)d\tau\\ &\le \frac{k_v}{1-k_vK}\int_0^t|\mu_1'(\tau)-\mu_2'(\tau)|d\tau+k_vK'\frac{k_v}{(1-k_vK)^2}\int_0^t|\mu_1(\tau)-\mu_2(\tau)|d\tau.\end{aligned}$$

On $[0,T]$ we can estimate $\|A_1-A_2\|_\infty\le C_1T\|\mu_1'-\mu_2'\|_\infty$ for a certain $C_1>0$, using $\|\mu_1-\mu_2\|_\infty\le TC_2\|\mu_1'-\mu_2'\|_\infty$ for a certain C_2. Moreover, concerning the maximum, we have

$$|A_i(t)|\le t\frac{k_vK}{1-k_vK}.$$

Altogether, we have therefore proved an estimate of the form $\|a_1-a_2\|_\infty\le c_0e^{T\frac{k_vK'}{1-k_vK}}C_1T\|\mu_1'-\mu_2'\|_\infty$. For the second term (38) we have

$$\begin{aligned}b_1(t)-b_2(t)&=\int_0^t e^{-(q/V)(t-\tau)}\widetilde{b}_1(\tau)\left[\exp\int_t^\tau-\frac{k_v\mu_1'(s)}{1-k_v\mu_1(s)}ds-\exp\int_t^\tau-\frac{k_v\mu_2'(s)}{1-k_v\mu_2(s)}ds\right]d\tau\\&\quad+\int_0^t e^{-(q/V)(t-\tau)}\exp\int_t^\tau-\frac{k_v\mu_2'(s)}{1-k_v\mu_2(s)}ds\cdot(\widetilde{b}_1(\tau)-\widetilde{b}_2(\tau))d\tau,\end{aligned}$$

where $\widetilde{b}_i(t)=\frac{1}{1-k_v\mu_i(t)}\left[\frac{qc_f(t)}{V}+\frac{\rho}{M}-k_v\mu_i'(t)-\frac{q\rho}{VM}(1+k_v\nu_i(t))\right]+\frac{q\rho}{VM}$. Now for $t\in[0,T]$,

$$|\widetilde{b}_i(t)|\le\frac{1}{1-k_vK}\left[\frac{qc_{fT}}{V}+\frac{\rho}{M}+\frac{q\rho}{VM}k_vK\right]+\frac{q\rho}{VM}=:C_3.$$

Put $B_i(t,\tau)=\int_\tau^t\frac{k_v\mu_i'(s)}{1-k_v\mu_i(s)}ds$, then $|B_i(t,\tau)|\le\frac{k_vK}{1-k_vK}(t-\tau)$ for $0\le\tau\le t$. Then b_1-b_2 can be estimated as

$$\begin{aligned}|b_1(t)-b_2(t)|&=\left|\int_0^t e^{-(q/V)(t-\tau)}\widetilde{b}_1(\tau)\left(e^{B_1(t,\tau)}-e^{B_2(t,\tau)}\right)d\tau+\int_0^t e^{-(q/V)(t-\tau)}e^{B_2(t,\tau)}\left(\widetilde{b}_1(\tau)-\widetilde{b}_2(\tau)\right)d\tau\right|\\&\le C_3\int_0^t\left|e^{B_1(t,\tau)}-e^{B_2(t,\tau)}\right|d\tau+e^{t\frac{k_vK}{1-k_vK}}\int_0^t|\widetilde{b}_1(\tau)-\widetilde{b}_2(\tau)|d\tau\\&\le C_3e^{tk_vK/(1-k_vK)}\int_0^t|B_1(t,\tau)-B_2(t,\tau)|d\tau+e^{t\frac{k_vK}{1-k_vK}}\int_0^t\left|\widetilde{b}_1(\tau)-\widetilde{b}_2(\tau)\right|d\tau.\end{aligned}$$

Finally,

$$\begin{aligned}|B_1(t,\tau)-B_2(t,\tau)|&\le\int_\tau^t\frac{k_v}{1-k_vK}|\mu_1'(s)-\mu_2'(s)|ds+\int_\tau^t k_vK\left|\frac{1}{1-k_v\mu_1(s)}-\frac{1}{1-k_v\mu_2(s)}\right|ds\\&\le(t-\tau)\frac{k_v}{1-k_vK}\|\mu_1'-\mu_2'\|_\infty+(t-\tau)\frac{k_v^2K}{(1-k_vK)^2}\|\mu_1-\mu_2\|_\infty\\&\le(t-\tau)\left[\frac{k_v}{1-k_vK}+tC_2\frac{k_v^2K}{(1-k_vK)^2}\right]\|\mu_1'-\mu_2'\|_\infty.\end{aligned}$$

Integrating gives $\int_0^t|B_1(t,\tau)-B_2(t,\tau)|d\tau\le C_{3t}t^2\|\mu_1'-\mu_2'\|_\infty$, where $C_{3t}\to k_v/(1-k_vK)$ as $t\to0^+$. Now

$$\begin{aligned}|\widetilde{b}_1(t)-\widetilde{b}_2(t)|&=\frac{1}{1-k_v\mu_1(t)}\left[\frac{qc_f(t)}{V}+\frac{\rho}{M}-k_v\mu_1'(t)-\frac{q\rho}{VM}(1+k_v\nu_1(t))\right]\\&\quad-\frac{1}{1-k_v\mu_2(t)}\left[\frac{qc_f(t)}{V}+\frac{\rho}{M}-k_v\mu_2'(t)-\frac{q\rho}{VM}(1+k_v\nu_2(t))\right]\\&=\left[\frac{qc_f(t)}{V}+\frac{\rho}{M}-\frac{q\rho}{VM}\right]\left(\frac{1}{1-k_v\mu_1(t)}-\frac{1}{1-k_v\mu_2(t)}\right)\\&\quad+\left[\frac{k_v}{1-k_v\mu_1(t)}\mu_1'(t)-\frac{k_v}{1-k_v\mu_2(t)}\mu_2'(t)\right]-\frac{q\rho}{VM}\left[\frac{k_v}{1-k_v\mu_1(t)}\nu_1(t)-\frac{k_v}{1-k_v\mu_2(t)}\nu_2(t)\right]\\&=:I_1(t)+I_2(t)+I_3(t).\end{aligned}$$

Here we estimate

$$\begin{aligned}|I_1(t)| &= \left|\left[\frac{qc_f(t)}{V}+\frac{\rho}{M}-\frac{q\rho}{VM}\right]\left(\frac{1}{1-k_v\mu_1(t)}-\frac{1}{1-k_v\mu_2(t)}\right)\right| \\ &\le \left[\frac{qc_fT}{V}+\frac{\rho}{M}-\frac{q\rho}{VM}\right]\frac{k_v}{1-k_vK^2}|\mu_1(t)-\mu_2(t)| \\ &\le \left[\frac{qc_fT}{V}+\frac{\rho}{M}-\frac{q\rho}{VM}\right]\frac{k_v}{1-k_vK^2}TC_2||\mu_1'-\mu_2'||_\infty.\end{aligned}$$

Using the same estimation as for $|A_1(t)-A_1(t)|$, we obtain

$$|I_2(t)| = \left|\frac{k_v}{1-k_v\mu_1(t)}\mu_1'(t)-\frac{k_v}{1-k_v\mu_2(t)}\mu_2'(t)\right| \le C_1||\mu_1'-\mu_2'||_\infty.$$

Next we have

$$\begin{aligned}|I_3(t)| &= \frac{q\rho}{VM}\left|\frac{k_v}{1-k_v\mu_1(t)}\nu_1(t)-\frac{k_v}{1-k_v\mu_2(t)}\nu_2(t)\right| \\ &= \frac{q\rho}{VM}\left|\frac{k_v}{1-k_v\mu_1(t)}(\nu_1(t)-\nu_2(t))-k_v\nu_2(t)\left(\frac{k_v}{1-k_v\mu_2(t)}-\frac{k_v}{1-k_v\mu_1(t)}\right)\right| \\ &\le \frac{q\rho}{VM}\frac{k_v}{1-k_v\mu_1(t)}||\nu_1-\nu_2||_\infty+\frac{k_vK}{(1-k_vK)^2}||\mu_1-\mu_2||_\infty \\ &\le \frac{q\rho}{VM}\frac{k_v}{1-k_v\mu_1(t)}||\nu_1-\nu_2||_\infty+\frac{k_vK}{(1-k_vK)^2}TC_2||\mu_1'-\mu_2'||_\infty.\end{aligned}$$

These estimates give us

$$\begin{aligned}||\widetilde{b}_1(t)-\widetilde{b}_2(t)||_\infty \le& \left[\left(\frac{qc_fT}{V}+\frac{\rho}{M}-\frac{q\rho}{VM}\right)\frac{k_v}{1-k_vK^2}TC_2+C_1+\frac{k_vK}{(1-k_vK)^2}TC_2\right]||\mu_1'-\mu_2'||_\infty \\ &+\frac{q\rho}{VM}\frac{k_v}{1-k_v\mu_1(t)}||\nu_1-\nu_2||_\infty \le C_5||\mu_1'-\mu_2'||_\infty+C_6||\nu_1-\nu_2||_\infty,\end{aligned}$$

hence

$$||b_1-b_2||_\infty \le \left(e^{t\frac{k_vK}{1-k_vK}}C_{3t}t^2+C_5e^{t\frac{k_vK}{1-k_vK}}\right)T\|\mu_1'-\mu_2'||_\infty+e^{t\frac{k_vK}{1-k_vK}}C_6T||\nu_1-\nu_2||_\infty.$$

By the previous estimates we obtain

$$\begin{aligned}||c_1-c_2||_\infty &\le e^{Tk_vK/(1-k_vK)}\left(C_{3t}T^2C_3+C_5+c_0C_1\right)T\|\mu_1'-\mu_2'||_\infty+e^{t\frac{k_vK}{1-k_vK}}C_6T||\nu_1-\nu_2||_\infty \\ &\le C_7T\|\mu_1'-\mu_2'||_\infty+C_8T||\nu_1-\nu_2||_\infty,\end{aligned}$$

which establishes the desired contraction estimates for c_1-c_2. Naturally, we also get estimates for $G(c_1)-G(c_2)$ and $B(c_1)-B(c_2)$, which are

$$||G(c_1)-G(c_2)||_\infty \le k_g||c_1-c_2||_\infty \le k_gC_7T\|\mu_1'-\mu_2'||_\infty+k_gC_8T||\nu_1-\nu_2||_\infty \tag{39}$$

and

$$\begin{aligned}||B(c_1)-B(c_2)||_\infty &\le 2k_bC_T(1+C_T)T||c_1-c_2||_\infty \\ &\le 2k_bC_T(1+C_T)C_7T\|\mu_1'-\mu_2'||_\infty+2k_bC_T(1+C_T)C_8T||\nu_1-\nu_2||_\infty.\end{aligned} \tag{40}$$

(ii) Estimating characteristics $\phi^1-\phi^2$

From (39) we readily get an estimate for characteristics. Putting $\phi^i_{t_0,L_0}(t)=L_0+\int_{t_0}^t G(c_i(\tau))\,d\tau$, $i=1,2$, we have

$$\left|\phi^1_{t_0,L_0}(t)-\phi^2_{t_0,L_0}(t)\right| \le \int_{t_0}^t |G(c_1(\tau))-G(c_2(\tau))|\,d\tau \le |t-t_0|\|G(c_1)-G(c_2)||_\infty \le |t-t_0|C_9\left|m_1-m_2\right|.$$

That means

$$\left|\phi^1_{t_0,L_0}(t)-\phi^2_{t_0,L_0}(t)\right| \le C_9T\left|m_1-m_2\right| \tag{41}$$

for all $t_0,t\in[0,T]$ and every L_0. The estimate (39) for $G(c_1)-G(c_2)$ also leads to immediate estimates for $L(\tau)$ and $L(\xi)$, namely

$$|L_1(\tau)-L_2(\tau)| \le \int_\tau^t |G(c_1(\sigma))-G(c_2(\sigma))|d\sigma \le T||G(c_1)-G(c_2)||_\infty \tag{42}$$

$$|L_1(\xi) - L_2(\xi)| \quad \leq \quad \int_0^t |G(c_1(\sigma)) - G(c_2(\sigma))| d\sigma \leq T||G(c_1) - G(c_2)||_\infty. \tag{43}$$

(iii) Estimating $\widetilde{\mu}_1' - \widetilde{\mu}_2'$

Formula (31) decomposes into a sum of 6 expressions $\widetilde{\mu}'(t) = A(t) + B(t) + \cdots + F(t)$, and we estimate $A_1(t) - A_2(t), \ldots, F_1(t) - F_2(t)$ separately. Writing $A(t) = 3G(c_i(t)) \int_0^t \mathcal{I}(t,\tau)d\tau$ with the obvious meaning of $\mathcal{I}(t,\tau)$ in (31), we have

$$\begin{aligned} A_1(t) - A_2(t) &= 3G(c_1(t)) \int_0^t \mathcal{I}_1(t,\tau)\, d\tau - 3G(c_2(t)) \int_0^t \mathcal{I}_2(t,\tau)\, d\tau, \\ &= 3\left[G(c_1(t) - G(c_2(t))\right] \int_0^t \mathcal{I}_1(t,\tau)\, d\tau + G(c_2(t)) \int_0^t \left[\mathcal{I}_1(t,\tau) - \mathcal{I}_2(t,\tau)\right] d\tau. \end{aligned}$$

By (39) and boundedness of $\int_0^t \mathcal{I}_1(t,\tau)\, d\tau$, the first term on the right is bounded by $C\|G(c_1(t)) - G(c_2(t))\|_\infty \leq C'T \left| m_1 - m_2 \right|$. Similarly, in the second term on the right we use boundedness of $G(c_2(t))$, which leaves us to estimate the expression $\int_0^t \left[\mathcal{I}_1(t,\tau) - \mathcal{I}_2(t,\tau)\right] d\tau$. Now $\mathcal{I}(t,\tau)$ can be decomposed as $\mathcal{I}(t,\tau) = \left(B(c(\tau))L(\tau)^2 + \mathcal{N}(t,\tau)\right)\mathcal{M}(t,\tau)$. That means, suppressing arguments and writing $\mathcal{B} = B(c)L(\tau)^2$ for simplicity

$$\mathcal{I}_1 - \mathcal{I}_2 = (\mathcal{B}_1 + \mathcal{N}_1)\left[\mathcal{M}_1 - \mathcal{M}_2\right] + \left[\mathcal{B}_1 - \mathcal{B}_2 + \mathcal{N}_1 - \mathcal{N}_2\right]\mathcal{M}_2.$$

Then it suffices to estimate the terms $\mathcal{B}_1 - \mathcal{B}_2$, $\mathcal{N}_1 - \mathcal{N}_2$ and $\mathcal{M}_1 - \mathcal{M}_2$ separately, and use this in tandem with boundedness of $B, \mathcal{M}, \mathcal{N}$ over the set $t, \tau \in [0,T]$. While estimate $\mathcal{B}_1 - \mathcal{B}_2$ is handled using (40) and (42), we consider

$$\begin{aligned} \mathcal{M}_1(t,\tau) - \mathcal{M}_2(t,\tau) &= \exp\left(-\int_\tau^t h(\phi^1_{\tau,0}(\sigma))d\sigma\right) L_1(\tau)^2 - \exp\left(-\int_\tau^t h(\phi^2_{\tau,0}(\sigma))d\sigma\right) L_2(\tau)^2 \\ &= \exp\left(-\int_\tau^t h(\phi^1_{\tau,0}(\sigma))d\sigma\right)\left[L_1(\tau)^2 - L_2(\tau)^2\right] \\ &\quad + \left[\exp\left(-\int_\tau^t h(\phi^1_{\tau,0}(\sigma))d\sigma\right) - \exp\left(-\int_\tau^t h(\phi^2_{\tau,0}(\sigma))d\sigma\right)\right] L_2(\tau)^2. \end{aligned}$$

Here the first line uses (42), while the second line uses $|e^{-a} - e^{-b}| \leq |a-b|$ for $a, b > 0$ and

$$\int_\tau^t h(\phi^1_{\tau,0}(\sigma)) - h(\phi^2_{\tau,0}(\sigma))d\sigma \leq \text{lip}(h) \int_\tau^t |\phi_{\tau,0}(\sigma) - \phi_{\tau,0}(\sigma)|d\sigma \leq \text{lip}(h)T^2C_9 \left| m_1 - m_2 \right|$$

where we assume that h is globally Lipschitz continuous on $[0,\infty)$ with constant lip(h). Now we use (41) to conclude that $|A_1(t) - A_2(t)| \leq CT \left| m_1 - m_2 \right|$ as claimed. For the term $B_1(t) - B_2(t)$ we decompose $B(t) = 3G(c(t)) \int_0^\infty \mathcal{J}(t,\xi)d\xi$, with $\mathcal{J}(t,\xi) = \left(n_0(\xi) + \mathcal{K}(\tau,\xi)\mathcal{E}(t,\xi)\right) L(\xi)^3$, then

$$B_1(t) - B_2(t) = 3G(c_1(t)) \int_0^\infty \left[\mathcal{J}_1(t,\xi) - \mathcal{J}_2(t,\xi)\right] d\xi + 3\left[G(c_1(t)) - G(c_2(t))\right] \int_0^\infty \mathcal{J}_2(t,\xi)d\xi,$$

so we have to show boundedness of $\int_0^\infty \mathcal{J}_2(t,\xi)d\xi$ over $t \in [0,T]$, and a Lipschitz estimate for the expression $\int_0^\infty \left[\mathcal{J}_1(t,\xi) - \mathcal{J}_2(t,\xi)\right] d\xi$. Boundedness requires $\int_0^\infty n_0(\xi)L(\xi)^2 d\xi < \infty$, and also $\sup_{0\leq t\leq T} \int_0^\infty w_i(L,t)L^2 dL < \infty$, which is clear since $w_1, w_2 \in E$. Now we concentrate on the Lipschitz estimate, where we have

$$\begin{aligned} \int_0^\infty \left[\mathcal{J}_1(t,\xi) - \mathcal{J}_2(t,\xi)\right] d\xi &= \int_0^\infty \left[\mathcal{K}_1(t,\xi)\mathcal{E}_1(t,\xi) - \mathcal{K}_2(t,\xi)\mathcal{E}_2(t,\xi)\right] L(\xi)^2 d\xi \\ &= \int_0^\infty \mathcal{K}_1(t,\xi)\left[\mathcal{E}_1(t,\xi) - \mathcal{E}_2(t,\xi)\right] L(\xi)^2 d\xi + \int_0^\infty \mathcal{E}_2(t,\xi)\left[\mathcal{K}_1(t,\xi) - \mathcal{K}_2(t,\xi)\right] L(\xi)^2 d\xi. \end{aligned}$$

Here the first term uses $\mathcal{E}_1(t,\xi) - \mathcal{E}_2(t,\xi) = \exp\left(-\int_0^t h(\phi^1_{0,\xi}(s))ds\right) - \exp\left(-\int_0^t h(\phi^2_{0,\xi}(s))ds\right)$, which is readily treated using (41) and the hypothesis that h is globally Lipschitz continuous. The second term requires estimation of

$$\mathcal{K}_1(t,\xi) - \mathcal{K}_2(t,\xi) = \int_0^t w_1(\phi^1_{0,\xi}(s),s) \exp\left\{\int_0^s h(\phi^1_{0,\xi}(\sigma)d\sigma\right\} - w_2(\phi^2_{0,\xi}(s),s) \exp\left\{\int_0^s h(\phi^2_{0,\xi}(\sigma)d\sigma\right\} ds,$$

which uses (41) in tandem with Lipschitz continuity of h and a uniform Lipschitz estimate $|w(L,t) - w(\hat{L},t)| \leq \|w\|_L |L - \hat{L}|$ on $t \in [0,T]$. Estimation of the third and fourth block in (31), that is $C_1 - C_2$ and $D_1 - D_2$, follows the same lines. From the last line in (31) we get the remaining two terms $E_1 - E_2$ and $F_1 - F_2$, which are estimated in much the same way as the corresponding $w_1 - w_2$ terms in the expression $B_1 - B_2$. We need once again the Lipschitz property of w_i in the first coordinate, and moreover, $\max_{0\leq t\leq T} \int_0^\infty w(L,t)L^3 dL < \infty$, which is guaranteed by $w_i \in E$. This concludes the estimation of $\widetilde{\mu}_1 - \widetilde{\mu}_2$.

(iv) Estimating the remaining terms

We have to estimate in much the same way the expressions $\widetilde{\nu}_1 - \widetilde{\nu}_2$ and $\widetilde{w}_1 - \widetilde{w}_2$. The first expression clearly follows the same line as in the previous section, now based on (28), while the second estimate uses (32). Naturally, for $\widetilde{w}_1 - \widetilde{w}_2$ we have to repeat this three times for the different norms involved, the principle being the same.

Altogether, we have shown that $\mathcal{Q} : X_T \to X_T$ is a contraction with respect to the metric (34) on X_T. Consequently, $\mathcal{Q}$ has a fixed point (w^*, μ^*, ν^*). □

It is now clear from inspecting (33) that c^*, defined through (19), and n^*, defined through (25), are solutions of the crystallizer model (11) – (16) on $[0, T]$.

4. Global existence of solutions

In this section we investigate the existence of global solutions. Recall that our model assumes that the mass of solvent is invariant, which includes the case of batch crystallization, or the continuous mode. Since the molar masses of solids and liquid of the constituent are assumed equal, as is the case in KCl-crystallization used in our simulation study, the total mass of constituent $m_{\text{solute}} + m_{\text{solid}}$ is also constant. This gives the lower bound

$$\epsilon(t) \geq \frac{V_{\text{solvent}}}{V} > 0 \tag{44}$$

for the void fraction $\epsilon(t)$ at all times t. Using (52) and the above mass conservation of constituent, we have

$$V\epsilon c M \leq V\epsilon_0 c_0 M + m_{\text{solid}}(0),$$

which by (44) gives the upper bound

$$c(t) \leq \frac{\epsilon_0 c_0}{\epsilon(t)} + \frac{m_{\text{solid}}(0)}{V M \epsilon(t)} \leq \frac{V\epsilon_0 c_0}{V_{\text{solvent}}} + \frac{m_{\text{solid},0}}{V_{\text{solvent}} M} =: c_\infty.$$

Since $c(t) > c_s$, we deduce that $c(t)$ is bounded.

Theorem 4.1 *Suppose the total volume of slurry V and the total mass of the constituent are held constant during the process. Then the crystallizer model (11) – (16) has a global solution.*

Proof 1) The proof of the local existence theorem shows that for initial data $c_0 > c_s$ and $n_0(L)$ there exists times $T > 0$ small enough such that $\mathcal{Q} : X_T \to X_T$ is a contraction with respect to the distance (34). As the construction only depends on the norm $\|n_0(L)\|_1$ of the initial seed, we may define $T_{1/2}(c_0, \nu_0)$ as the largest $T > 0$ such that for initial data $c_s < c(0) \leq c_0$ and $\|n_0\|_1 \leq \nu_0$, we have $\text{dist}(\mathcal{Q}(m_1), \mathcal{Q}(m_2)) \leq \frac{1}{2}\text{dist}(m_1, m_2)$. In particular, the solution $c(t)$, $n(L, t)$ of (11) – (16) is guaranteed to exist at least on the interval $[0, T_{1/2}(c(0), \|n_0(\cdot)\|_1)]$.

As a consequence of the above, if we let $\ell(c_0, \|n_0\|_1, T)$ be the Lipschitz constant of $\mathcal{Q}$ on X_T with initial data c_0 and $n_0(L)$, then by construction $\ell\left(c_0, \|n_0\|_1, T_{1/2}(c_0, \|n_0\|_1)\right) = \frac{1}{2}$.

2) Using the boundedness of $c(t)$, there exist constants G_0, B_0 such that $G(c(t)) \leq G_0$ and $B(c(t))/G(c(t)) \leq B_0$. Now let $\widetilde{n}(L, t)$ be the solution of (11) with constant speed of growth G_0 and constant birth rate $n(0, t) = B_0$. Then $n(L, t) \leq \widetilde{n}(L, t)$ on any finite interval of existence of the solution $n(L, t)$. Indeed, $\widetilde{n}$ is the solution of a linear population balance equation which exists on any finite interval. Moreover, since in the model for $\widetilde{n}$ more crystals nucleate and the speed of growth is faster, while breakage, fines dissolution and product removal are treated the same way, the linear model has a larger number of crystals at all sizes L.

3) Let us define $\widetilde{n}(T) = \sup_{t \leq T} \int_0^\infty \widetilde{n}(L, t) dL$, then $\|n(\cdot, t)\|_1 \leq \widetilde{n}(T)$ for all L and all $t \leq T$ as long as $n(L, t)$ exists on $[0, T]$. Now fix $T^* > 0$. We will prove that the solutions $c(t)$ and $n(L, t)$ exist on $[0, T^*]$. Put $T_{1/2} := T_{1/2}(c_\infty, \widetilde{n}(T^*))$. Then the solution $c(t)$, $n(L, t)$ exist on $[0, T_{1/2}]$. If $T_{1/2} \geq T^*$ we are done, so suppose $T_{1/2} < T^*$.

In order to extend the solution further to the right, we take $c(T_{1/2})$ and $n(\cdot, T_{1/2})$ as new initial data. Since $c(T_{1/2}) \leq c_\infty$ and $\|n(\cdot, T_{1/2})\|_1 \leq \widetilde{n}(T_{1/2}) \leq \widetilde{n}(T^*)$, the new initial data are still bounded by c_∞ and $\widetilde{n}(T^*)$. That means, the new solution will exist at least on $[0, T_{1/2}]$ with $T_{1/2} = T_{1/2}(c_\infty, \widetilde{n}(T^*))$ the same as before. Patching together, we have now existence of a solution of (11) – (16) on $[0, 2T_{1/2}]$. If $2T_{1/2} \geq T^*$ we are done, otherwise we use $c(2T_{1/2})$ and $n(L, 2T_{1/2})$ as new initial data. Then we still have $c(2T_{1/2}) \leq c_\infty$ and $\|n(\cdot, 2T_{1/2})\|_1 \leq \widetilde{n}(2T_{1/2}) \leq \widetilde{n}(T^*)$, so we have the same bound as before, and the local existence proof will again work at least on $[0, T_{1/2}]$. After a finite number of steps we reach T^*, which proves that the solution exists on $[0, T^*]$. □

Remark 3 *Since $G(c)$ and $B(c)$ are only locally Lipschitz functions in general, we should not expect global existence of solutions in all cases. For instance, in evaporation crystallization the solvent mass is driven to 0 in finite time T, which leads to $c(t) \to \infty$ as $t \to T$, even though the mass of constituent may remain bounded.*

5. Numerical tests

In this section we present a numerical simulation for the continuous crystallizer in steady state with fines dissolution, product classification, and negligible attrition. This case is instructive as it shows that the system has a one-parameter family of possible steady states, which are oscillatory or even unstable and in practice need feedback control. See e.g. [1, 5]. A second interest in this study lies in the fact that in this case an explicit solution can be computed, which allows to validate the numerical scheme.

The population and mole balance equations at the equilibrium $(c_{ss}, n_{ss}(L))$ are maintained through the steady-state feed concentration c_{fss}, where c_{ss} and $n_{ss}(L)$ are respectively the corresponding steady state values for solute $c(t)$ and CSD $n(L,t)$. For fixed c_{ss}, the population balance equation has a unique solution n_{ss}

$$n_{ss}(L) = n_{ss}(0)\exp\left\{-\frac{q}{Vk_g(c_{ss}-c_s)}\int_0^L h_{f+p}(l)dl\right\} \tag{45}$$

where $n_{ss}(0) = \dfrac{B(c_{ss})}{G(c_{ss})}$. Then we obtain

$$n_{ss}(L) = \frac{B(c_{ss})}{G(c_{ss})}\begin{cases} \exp\left\{-\dfrac{q}{Vk_g(c_{ss}-c_s)}(1+R_f)L\right\} & 0 \le L < L_f \\ \exp\left\{-\dfrac{q}{Vk_g(c_{ss}-c_s)}(R_fL_f+L)\right\} & L_f \le L < L_p \\ \exp\left\{-\dfrac{q}{Vk_g(c_{ss}-c_s)}(R_fL_f-R_pL_p+(1+R_p)L)\right\} & L_p \le L \end{cases} \tag{46}$$

with

$$\epsilon_{ss} = 1 - k_v\int_0^\infty n_{ss}(L)L^3dL$$

and

$$\eta_{ss} = k_v\int_{L_p}^\infty n_{ss}(L)L^3dL,$$

which as we realize are both functions of c_{ss} alone. Our fundings are

$$\int_0^\infty n_{ss}(L)L^3dL = I_1+I_2+I_3, \qquad \int_0^\infty n_{ss}(L)L^3dL = I_3$$

where

$$\begin{aligned} I_1 &= \frac{k_b}{k_g}(c_{ss}-c_s)^3\int_0^{L_f} e^{-aL}L^3dL \\ &= \frac{k_b}{k_g}(c_{ss}-c_s)^3\left(e^{-aL_f}\left(-\frac{L_f^3}{a}-\frac{3L_f^2}{a^2}-\frac{6L_f}{a^3}-\frac{6}{a^4}\right)+\frac{6}{a^4}\right) \end{aligned}$$

with

$$a = \frac{q}{Vk_g(c_{ss}-c_s)}(1+R_1)$$

and

$$\begin{aligned} I_2 &= \frac{k_b}{k_g}(c_{ss}-c_s)^3 e^{-\frac{q}{Vk_g(c_{ss}-c_s)}}\int_{L_f}^{L_p} e^{-bL}L^3dL \\ &= \frac{k_b}{k_g}(c_{ss}-c_s)^3 e^{-\frac{q}{Vk_g(c_{ss}-c_s)}}\left(e^{-bL_p}\left(-\frac{L_p^3}{b}-\frac{3L_p^2}{b^2}-\frac{6L_p}{b^3}-\frac{6}{b^4}\right)\right. \\ &\quad \left.+\,e^{-bL_f}\left(\frac{L_p^3}{b}+\frac{3L_p^2}{b^2}+\frac{6L_p}{b^3}+\frac{6}{b^4}\right)\right) \end{aligned}$$

with

$$b = \frac{q}{Vk_g(c_{ss}-c_s)}$$

and

$$\begin{aligned} I_3 &= \frac{k_b}{k_g}(c_{ss}-c_s)^3 e^{-\frac{q}{Vk_g(c_{ss}-c_s)}(R_1L_f-R_2L_p)}\int_{L_p}^\infty e^{-cL}L^3dL \\ &= \frac{k_b}{k_g}(c_{ss}-c_s)^3 e^{-\frac{q}{Vk_g(c_{ss}-c_s)}(R_1L_f-R_2L_p)}e^{-cL_p}\left(\frac{L_p^3}{c}+\frac{3L_p^2}{c^2}+\frac{6L_p}{c^3}+\frac{6}{c^4}\right) \end{aligned}$$

with

$$c = \frac{q}{Vk_g(c_{ss} - c_s)}(1 + R_2).$$

Now we have to substitute these numbers in the steady state mole balance equation, $\frac{dc(t)}{dt} = 0$, $\frac{d\epsilon(t)}{dt} = 0$. This gives

$$c_{fss} = \rho(1 + R_2\eta_{ss}) - (\rho - Mc_{ss})\epsilon_{ss}. \tag{47}$$

This means that for a given steady state concentration, c_{ss}, there exists a unique possible steady state feed concentration c_{fss}. Of course there are limiting values, determined by the constraint $0 \leq \epsilon_{ss} \leq 1$. In fact, the case $c_{ss} = c_s$ gives $n_{ss} = 0$, $\epsilon_{ss} = 1$, so no crystal production at all. This correspond to the feed $c_{fss} = c_{ss} = c_s$. In the other end, the limiting case $\epsilon_{ss} = 0$ means no liquid left, everything is crystal.

For the simulation, we use the finite difference upwind scheme [30]. We then transform the system into an ODE system and run an ODE solver. A simulation of the nonlinear model is shown in figure 2. For visualization we use the mass density function $m(L, t) = \rho k_v n(L, t)L^3$ and the number density function $n(L, t)$ representing the crystal size distribution. The feed concentration is kept constant at $c_{fss} = 4.4\ mol/l$. The corresponding steady state value for solute concentration $c(t)$ is $c_{ss} = 4.091\ mol/l$.

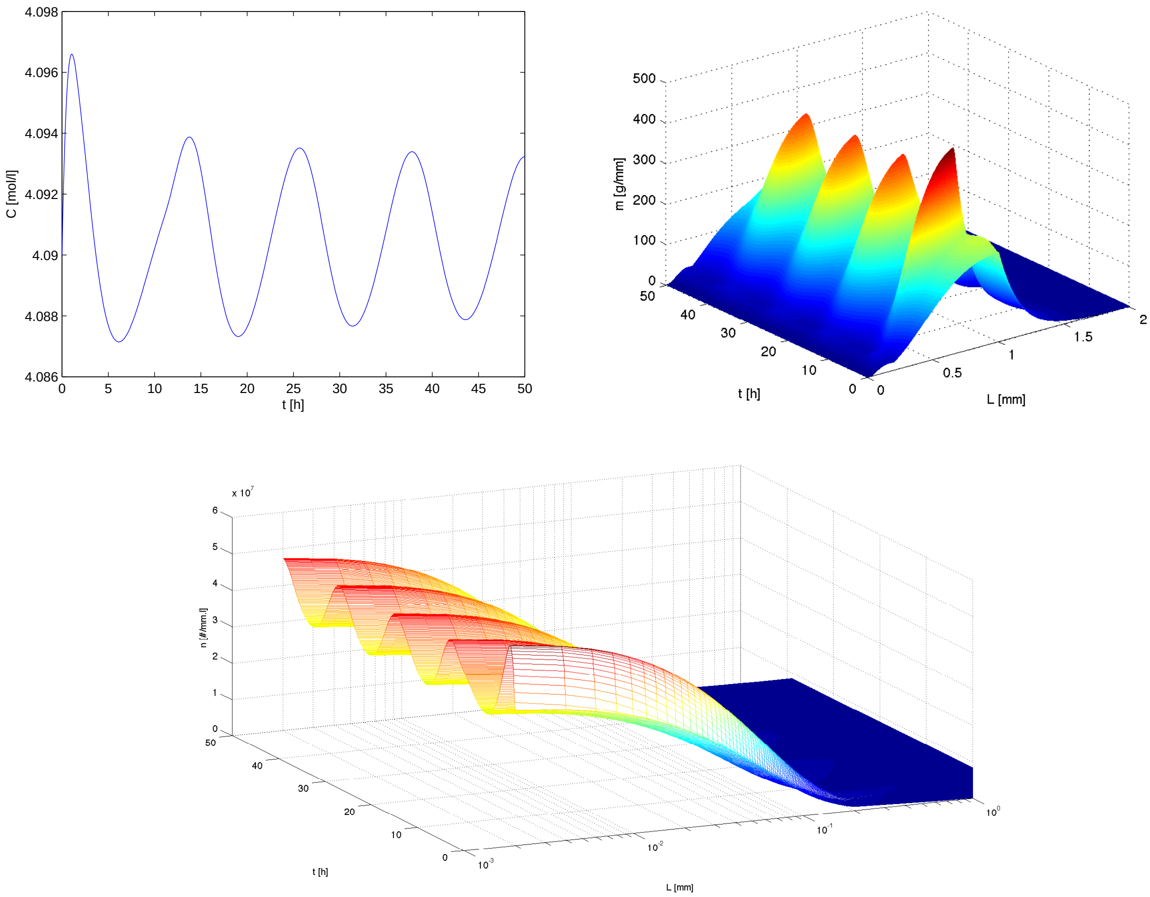

Figure 2. Simulation of continuous crystallizer with fines dissolution, product removal, and negligible attrition at steady state. Solute concentration (left), mass density $m(L, t) = \rho k_v n(L, t)L^3$ (right) and number density function $n(L, t)$ (at the bottom) show sustained oscillations and even instability, which may degrade product quality.

6. Discussion and conclusion

We have derived a mathematical model of a mixed suspension mixed product removal crystallizer based on population and mass balance equations. Consistency of the model was shown by proving local existence and uniqueness of solutions using the method of characteristics. Global existence of solutions for continuous and batch mode was also established using a prior bound on the solute concentration derived from physically meaningful conditions. Our method of proof indicates that when growth rate

$G(c)$ and nucleation rate $B(c)$ are only locally Lipschitz functions, we should not expect global existence of solutions as a rule. The complexity of the continuous crystallization mode with fines dissolution, classified product and breakage dependent on the Lipschitz behavior of growth and birth rates is a novel aspect of our study compared to models discussed in mathematical biology [17, 15, 20]. Our work contributes the first complete mathematical study of crystallization based on a model including population, molar balances and breakage phenomenon. We have also presented numerical simulations of the continuous crystallizer in steady state. The result of a simulation study of continuous crystallizer in steady state shows that feedback control is needed to avoid oscillations that may degrade product quality.

Appendix

In this appendix, we give details how the mole balance equation is obtained by investigating the mass balance within the crystallizer. The total mass m of the suspension in the crystallizer is decomposed as

$$m = m_{\text{liquid}} + m_{\text{solid}} = m_{\text{solvent}} + m_{\text{solute}} + m_{\text{solid}}. \tag{48}$$

In this study we consider non-solvated crystallization, where solute molecules transit directly into solid state without integrating (or capturing) solvent molecules. We therefore have

$$\dot m^+_{\text{solvent}} = \dot m^-_{\text{solvent}}. \tag{49}$$

In batch mode $q = 0$ the total mass is preserved, $\frac{dm}{dt} = 0$, and the system is closed. We will then obtain the relation $\frac{dm_{\text{solute}}}{dt} = -\frac{dm_{\text{solid}}}{dt}$. In continuous or semi-batch mode this equation has to be completed by external sources and sinks. Using (49) this takes the form

$$\frac{dm}{dt} = \frac{dm_{\text{solute}}}{dt} + \frac{dm_{\text{solid}}}{dt} = \dot m^+_{\text{solute}} - \dot m^-_{\text{solute}} + \dot m^+_{\text{solid}} - \dot m^-_{\text{solid}}. \tag{50}$$

We will now have to relate this equation to the population balance equation (1). In analogy with (48) we decompose the total volume V of the suspension as $V = V_{\text{liquid}} + V_{\text{solid}} = V_{\text{solute}} + V_{\text{solvent}} + V_{\text{solid}}$. The liquid section is the dimensionless quantity

$$\varepsilon = \frac{V_{\text{liquid}}}{V} = 1 - \frac{V_{\text{solid}}}{V}. \tag{51}$$

A natural physical quantity to describe the solute population is the molar concentration of solute $c_m = \frac{V_{\text{solute}}}{V}\frac{\rho}{M}$, which quantifies the amount of the solute constituent per volume of the suspension. Here $\rho := \rho_{\text{solute}} = \rho_{\text{solid}}$ is the density of solute, and by assumption also the crystal density, M is the molar mass of the constituent, V the total volume of the suspension, and V_{solute} is the volume of solute in the suspension. The unit of c_m is [mol/ℓ].This allows us now to introduce the molar concentration of solute in the liquid phase $c = \varepsilon^{-1} c_m = \frac{V_{\text{solute}}}{V_{\text{liquid}}}\frac{\rho}{M}$, whose dimension is again [mol/ℓ]. This quantity leads to a more complicated form of the mole balance equation, but its use is dictated by the fact that growth term in the population balance (1) depends on c and not directly on c_m. We deduce the relation

$$m_{\text{solute}} = V\varepsilon c M. \tag{52}$$

Since we consider the total volume V of the suspension as constant, we obtain the formula

$$\frac{dm_{\text{solute}}}{dt} = \frac{d(V\varepsilon c M)}{dt} = V\frac{d\varepsilon}{dt}cM + V\varepsilon\frac{dc}{dt}M. \tag{53}$$

Let us now get back to (50). We start by developing the expressions on the right hand side. Decomposing $\dot m^{\pm}_{\text{solid}} = \dot m^{\pm}_{\text{fines}} + \dot m^{\pm}_{\text{product}} + \dot m^{\pm}_{\text{general}}$, we have $\dot m^{+}_{\text{fines}} + \dot m^{+}_{\text{product}} + \dot m^{+}_{\text{general}} = 0$, meaning that we do not add crystals during the process. For crystal removal we have

$$\dot m^-_{\text{product}} = qk_v\rho\int_0^\infty h_p(L)n(L,t)L^3\,dL,$$

which means that crystals of size L are filtered with a certain probability $h_p(L)$ governed by the product classification function. Similarly, fines are removed according to

$$\dot m^-_{\text{fines}} = qk_v\rho\int_0^\infty h_f(L)n(L,t)L^3\,dL$$

where $h_f(L)$ is the fines removal filter profile.

The term $\dot m^-_{\text{general}} = qk_v\rho\int_0^\infty n(L,t)L^3\,dL$ corresponds to a size indifferent removal of particles caused by the flow with rate q. The external terms for solute include $\dot m^-_{\text{solute}} = \frac{q}{V}m_{\text{solute}}$, meaning that due to the flow with rate q a portion of the solute mass is lost.

In the input we have $\dot{m}^+_{\text{solute}} = qc_f M + \dot{m}^-_{\text{fines}}$, where $qc_f M$ means solute feed and is a control input. The second term $\dot{m}^-_{\text{fines}}$ means that the mass which is subtracted from m_{solid} in the dissolution phase is recycled and added to m_{solute}. Altogether, fines therefore do not alter the mass balance. We now have related the dotted expressions to quantities used in the population balance equation.

Our next step is to relate the internal dynamics of the mass balance to the population balance equation. We start by noting that $m_{\text{solid}}(t) = k_v \rho V \int_0^\infty n(L,t)L^3\,dL$. Differentiating with respect to time and substituting the integrated right hand side of the population balance equation $\int_0^\infty \{\dots\}L^3 dL$ in (1) gives

$$\begin{aligned}
\frac{dm_{\text{solid}}(t)}{dt} &= k_v \rho V \int_0^\infty \frac{\partial n(L,t)}{\partial t} L^3 dL \\
&= -k_v \rho V G(c(t)) \int_0^\infty \frac{\partial n(L,t)}{\partial L} L^3 dL - k_v \rho q \int_0^\infty \left(1 + h_f(L) + h_p(L)\right) n(L,t) L^3\,dL \\
&= 3k_v V \rho G(c(t)) \int_0^\infty n(L,t) L^2\,dL - k_v \rho q \int_0^\infty \left(1 + h_f(L) + h_p(L)\right) n(L,t) L^3\,dL,
\end{aligned} \tag{54}$$

where the third line uses integration by parts and also the fact that breakage conserves mass at all times

$$\int_0^\infty \left(\mathcal{Q}^+_{\text{break}}(L,t) - \mathcal{Q}^-_{\text{break}}(L,t)\right) L^3 dL = 0,$$

so that integrated terms related to breakage cancel. Altogether, equation (50) becomes

$$\begin{aligned}
&\frac{d(V\epsilon c M)}{dt} + 3k_v \rho V G(c(t)) \int_0^\infty n(L,t)L^2\,dL - k_v \rho q \int_0^\infty \left(1 + h_f(L) + h_p(L)\right) n(L,t)L^3\,dL \\
&\quad = -\frac{q}{V} V \epsilon c M + q c_f(t) M - q k_v \rho \int_0^\infty (1 + h_p(L)) n(L,t) L^3 dL.
\end{aligned}$$

Subtracting the term $qk_v\rho \int_0^\infty (1+h_p(L))n(L,t)L^3 dL$ on both sides gives

$$\frac{d(V\varepsilon c M)}{dt} = qc_f M - qc\varepsilon M - 3Vk_v\rho G(c(t)) \int_0^\infty n(L,t)L^2 dL + qk_v\rho \int_0^\infty h_f(L)n(L,t)L^3 dL, \tag{55}$$

where $c_f(t)$ is the feed concentration and $\varepsilon(t)$ is the void fraction (51), which takes the form

$$\varepsilon(t) = 1 - k_v \int_0^\infty n(L,t)L^3 dL, \tag{56}$$

where k_v is the volume shape factor of crystals (see Table 1).

Differentiating (56) with respect to time t yields

$$\frac{d\varepsilon(t)}{dt} = -k_v \int_0^\infty \frac{\partial n(L,t)}{\partial t} L^3 dL. \tag{57}$$

By substituting the population balance Eq.(1) in first step and using the partial integration in second step, the temporal variation of the liquid fraction is

$$\begin{aligned}
\frac{d\varepsilon(t)}{dt} &= k_v G(c(t)) \int_0^\infty \frac{\partial n(L,t)}{\partial L} L^3 dL + \frac{q}{V} k_v \int_0^\infty (1 + h_f(L) + h_p(L)) n(L,t) L^3 dL \\
&= -3k_v G(c(t)) \int_0^\infty n(L,t)L^2 dL + \frac{q}{V} k_v \int_0^\infty (1 + h_f(L) + h_p(L)) n(L,t) L^3 dL.
\end{aligned} \tag{58}$$

Solving (58) for $3k_v G(c(t)) \int_0^\infty n(L,t)L^2 dL$ and multiplying by ρ, the mass balance of the solute in the liquid in (55) simplifies to

$$\frac{d(V\varepsilon c M)}{dt} = qc_f M - qc\varepsilon M + V\rho \frac{d\varepsilon}{dt} - q\rho k_v \int_0^\infty (1 + h_p(L)) n(L,t) L^3 dL. \tag{59}$$

Finally, using (53), equation (59) is transformed to

$$M\frac{dc(t)}{dt} = \frac{q(\rho - Mc(t))}{V} + \frac{\rho - Mc(t)}{\varepsilon(t)} \frac{d\varepsilon(t)}{dt} + \frac{qc_f(t)M}{V\varepsilon(t)} - \frac{q\rho}{V\varepsilon(t)} \left(\varepsilon(t) + k_v \int_0^\infty (1 + h_p(L)) n(L,t) L^3 dL\right). \tag{60}$$

By substituting (56) into the last term of the right hand side in this last equation, the mass balance of the solute in the liquid phase is finally obtained as

$$M\frac{dc}{dt} = \frac{q(\rho - Mc)}{V} + \frac{\rho - Mc}{\varepsilon}\frac{d\varepsilon}{dt} + \frac{qc_f M}{V\varepsilon} - \frac{q\rho}{V\varepsilon}(1 + k_v \nu(t)) \tag{61}$$

with

$$\nu(t) = \int_0^\infty h_p(L)n(L,t)L^3 dL. \tag{62}$$

This is now equation (10) in the text, which is thereby justified. We shall assume $0 \le h_p(L) \le R_p$ for all L, so that $\nu(t) \le R_p\mu(t)$ for all t. In the case where h_p in Eq.(62) is an ideal high pass filter, we obtain

$$\nu(t) = R_p \int_{L_p}^\infty n(L,t)L^3 dL. \tag{63}$$

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