

# Mathematical Aspects of Coagulation-Fragmentation Equations\*

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**Abstract** We give an overview of the mathematical literature on the coagulation-like equations, from an analytic deterministic perspective. In Section 1 we present the coagulation type equations more commonly encountered in the scientific and mathematical literature and provide a brief historical overview of relevant works. In Section 2 we present results about existence and uniqueness of solutions in some of those systems, namely the discrete Smoluchowski and coagulation-fragmentation: we start by a brief description of the functional spaces, and then review the results on existence of solutions with a brief description of the main ideas of the proofs. This part closes with the consideration of uniqueness results. In Sections 3 and 4 we are concerned with several aspects of the solutions behaviour. We pay special attention to the long time convergence to equilibria, self-similar behaviour, and density conservation or lack thereof.

## 1 Introduction: some processes and models

Coagulation (coalescence, agglomeration, aggregation) and fragmentation phenomena are ubiquitous in many scientific disciplines, such as: Physical [41, 73, 83], Astronomical [194], Chemical [228], Atmospheric [188, 196], Biological [6, 174], Environmental [96, 151], as well as in several technological processes [24, 86, 97].

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Their quantitative modelling can be achieved by several mathematical approaches, such as those using stochastic processes, computer simulations, or by the mathematical or numerical analysis of certain types of differential equations, generally called coagulation-fragmentation equations. We will centre most of our attention in a class of these equations, the discrete coagulation-fragmentation equations, but will also refer to the so called continuous case. Our goal is to review the most important Mathematical Analysis results about existence, uniqueness and several properties of the solutions, with special attention to dynamical aspects, in a way that is accessible to anyone with a background in differential equations but with no previous contact with these equations. To this end, we shall try to present the results, the ideas of the proofs, and some of their details, in the most reader friendly way we can, often detailing simpler situations more deeply and just glossing over more technically demanding proofs, or even just referring the result and calling the reader's attention to the original articles. This, we think, will help the reader to gain a feel for the subject without getting too much bogged down in the technicalities, at the same time that will show him/her directions for a deeper study of the issues.

In order to provide an overview as broad as possible of the field, in this introductory part, in addition to those systems that will be the focus of our attention later on, we will present a number of coagulation-fragmentation equations that have been studied in the mathematical literature, although we will not enter into much detail, and, in several cases, will not refer to them again afterwards.

The mathematical literature on this type of equations has had a huge growth in the last two decades, so much so that a comprehensive review of the field is no longer possible in a work such as this. And this is not only true about the mathematical literature, but even more so about the mathematical modelling literature, as well as the extremely rich and variegated contributions coming from Physics and other scientific and technological areas. This scientific literature is also a seemingly inexhaustible fountain of interesting and difficult mathematical problems, and so every mathematician must spend as much (if not more) time being acquainted with it than he/she will spend with the mathematical literature. This feature is also reflected in the list of references of the present work.

Although the Mathematical Analysis of the coagulation-fragmentation equations is a relatively recent research area, there exists already a number of reviews that are useful to whoever wants to obtain an overview of the problems, methods, and existing results. Two of the most recent reviews, [133, 219], have important overlaps with topics we deal with in this work. Another recent review, largely (but not exclusively) centered on the study of coagulation-fragmentation-diffusion systems using duality arguments and entropy and entropy-dissipation methods is [68]. A very interesting introduction to some of the classical models in this field, including a discussion of the physical ideas involved, is [43]. Another interesting reference, although somewhat outdated, is [74]. Reference [72] was the first review article about mathematical modelling of these problems and although very few of its content is mathematically rigorous it is still an interesting source for the literature before the 1970s. Finally, although somewhat outside the scope of our work, it is important to point out the review article of David Aldous [2] that seems to have had a tremendous

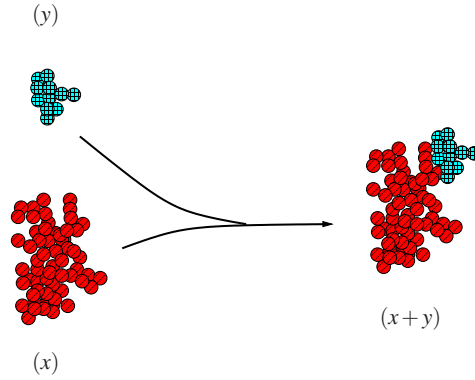
importance in calling the attention of probabilists to this area of research which resulted in the contemporaneous explosion of works using a stochastic approach to the coagulation and fragmentation processes (see, for instance, [21] and references therein). Similarly to what happens with the studies using a stochastic approach, those on numerical analysis (see, e.g., [88, 119, 141, 142, 189, 226]) are outside the scope of this work; a natural consequence of space limitations and also of my lack of expertise in those fields.

### 1.1 Coagulation and fragmentation processes

By coagulation (or coalescence, or agglomeration, or aggregation) one means a class of phenomena by means of which there is an increase in the size (or mass) of particles through their collision with other similar particles. In the overwhelming majority of cases simultaneous collision of more than two particles are extremely rare and are not considered.

In Figure 1 we present a schematic coagulation event between a particle of size (or mass)  $x$ , called  $x$ -cluster, and another of size (or mass)  $y$ ,  $y$ -cluster. As will be pointed out below there are modelling situation in which particle sizes vary continuously (in  $\mathbf{R}^+$ ) and others for which the size is assumed discrete, indexed in  $\mathbf{N}^+$ .

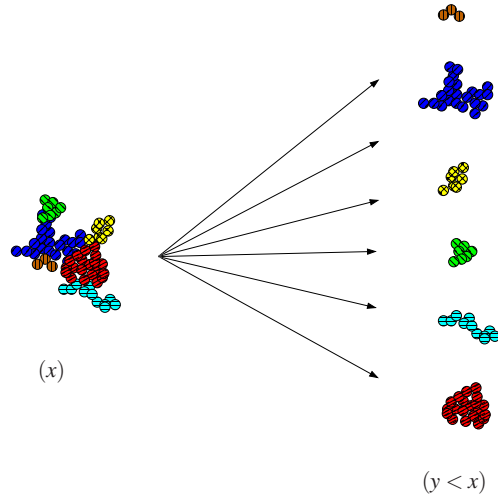
**Fig. 1** Scheme of the coagulation process of an  $x$ -cluster and a  $y$ -cluster.



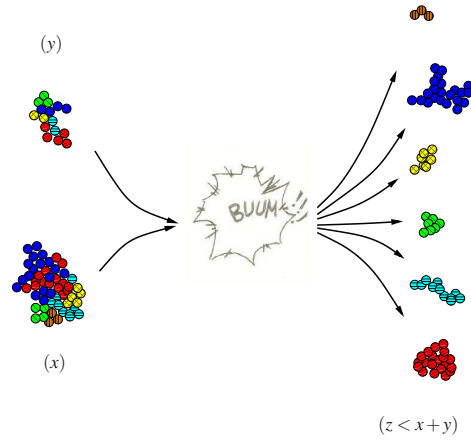
The reciprocal process of spontaneous fragmentation is, as the name implies, that by which a given particle breaks up and originates two, or more than two, smaller particles, and is schematically presented in Figure 2.

A different type of fragmentation that is sometimes considered in the literature is the collision induced fragmentation, also known as non-linear fragmentation: we can consider this process as made of two consecutive steps: a coagulation step forming an extremely unstable cluster that (in the time scale of the full process) instantaneously breaks into two or more smaller aggregates, as schematically illustrated in Figure 3.

**Fig. 2** Scheme of the spontaneous (or linear) multiple fragmentation process of an  $x$ -cluster into several smaller  $y$ -clusters.



**Fig. 3** Scheme of the collisional (or non-linear) fragmentation process.



The differential equations of coagulation-fragmentation type are one of the attempts at the mathematical modelling of the processes schematically represented in figures 1, 2, and 3. All these equations can be considered as equations of structured population dynamics, as they model the dynamics of systems of particles with some kind of internal structure (due to size, mass, or some other characteristic). Indeed, there are cases where standard equations of biological population dynamics pop up in the study of coagulation systems, although not in a direct and obvious way (e.g.: see [53, 61]), but the fact remains that, due to the special structure of the coagulation-fragmentation, the general methods of structured population dynamics

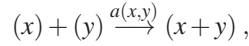
(as in, e.g., [221]) are usually not relevant (however, see Subsection 1.10.4). A much more important connection, as far as mathematical methods are concerned, is with kinetic theory, as is made plain in some of the most recent studies on the long time behaviour of solutions and self-similarity we shall refer to below.

In the following parts of the present section we briefly present some of the coagulation and fragmentation models that have been more widely studied from the mathematical point of view.

## 1.2 Smoluchowski's coagulation equations

The coagulation differential equation was originally proposed in 1916 by the physicist Marian von Smoluchowski as a model for the kinetics of colloid formation [204, 205], and in spite of the fact that it is at present one of the best studied, there are still a number of important open mathematical problems about it.

Let us represent the coagulation process of Figure 1 in the following notation, usual in chemical kinetics:



where  $a(x,y)$  is the rate of the coagulation reaction among an  $x$ -cluster and a  $y$ -cluster, usually called the coagulation coefficient or coagulation kernel. Often these coefficients depend only on the mass of the clusters, but there are cases where it is important to consider dependences on some other characteristic (cf. cases in Subsections 1.9 and 1.10) or upon time [212, 223]. The only general mathematical property imposed by all physical situations is the symmetry and non-negativity of the coagulation kernel:  $a(x,y) = a(y,x) \geq 0$ .

When the cluster masses assume only discrete values, multiples of a smaller quantity considered as unity (the mass of the 1-cluster, or *monomer*), the usual notation for  $a(x,y)$  is  $a_{x,y}$ , and the usual letters to denote cluster sizes are  $i, j, k, \dots$ , instead of  $x, y, \dots$ .

In Table 1 we collected some coagulation kernels occurring in the literature (see references cited in [49, 72, 133]).

Let us start by considering the case of discrete masses, which was also the one considered by Smoluchowski. Assuming the system is spatially homogeneous, we represent the concentration (or density) at time  $t$  of the  $j$ -cluster by  $c_j = c_j(t)$ , and denote by  $c = (c_j)$  the vector of concentrations of all clusters. Assuming valid the mass action law of chemical kinetics the rate of change of  $c_j$  is given by the differential equation

$$\dot{c}_j = Q_c(c)(j) \tag{1}$$

where  $\dot{c}_j$  denotes the time derivative  $c_j$  and  $Q_c(c)(j)$  is the mathematical function that represents the coagulation (hence the subscript  $c$ ) reaction terms affecting the  $j$

**Table 1** Some coagulation kernels  $a(x, y)$  occurring in the literature

$a(x, y)$	Comments
1	Approximately Brownian coagulation Linear chain polymerization
$x + y$	Polymerization of branched chains of $ARB_{f-1}$ type ( $f \gg 1$ )
$x^{-2/3} + y^{-2/3}$	Limit case of gravitation coagulation Diffusional growth of supported metal crystalites
$xy$	Polymerization of branched chains of $RA_f$ type ( $f \gg 1$ ).
$x^\alpha y^\beta + x^\beta y^\alpha$	A general case including e.g. Golovin, Stockmayer, etc.
$(x^{1/3} + y^{1/3})(x^{-1/3} + y^{-1/3})$	Brownian coagulation (continuum regimen)
$(x^{1/3} + y^{1/3})^2 (x^{-1} + y^{-1})^{1/2}$	Brownian coagulation (free molecular regimen)
$(x^{1/3} + y^{1/3})^3$	Tangential coagulation (linear velocity profiles)
$(x^{1/3} + y^{1/3})^7$	Tangential coagulation (non-linear velocity profiles)
$(x^{1/3} + y^{1/3})^2  x^{1/3} - y^{1/3} $	Gravitational deposition (particles bigger than $\sim 50\mu m$ ).
$(x^\alpha + y^\alpha)^\beta  x^\gamma - y^\gamma $	Ballistic coagulation ( $\alpha, \beta, \gamma \geq 0, \alpha\beta + \gamma \leq 1$ )

component of the concentration vector  $c$ . There are two contributions to this reaction term:

1. the creation of  $j$ -clusters due to the reactions of smaller clusters with appropriate masses,  $(j-k) + (k) \rightarrow (j)$ , with  $k = 1, \dots, j-1$ , and  $j \geq 2$ , to which corresponds the term

$$Q_1(c)(j) := \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k, \quad (2)$$

and defining  $Q_1(c)(1) := 0$ ;

2. the destruction of  $j$ -clusters due to the coagulation reactions of a  $j$ -cluster and any other present in the system,  $(j) + (k) \rightarrow (j+k)$ , with  $k = 1, 2, \dots$ . Not imposing an *a priori* upper bound on the size of the clusters, to this process corresponds the term

$$Q_2(c)(j) := c_j \sum_{k=1}^{\infty} a_{j,k} c_k. \quad (3)$$

Hence, the right-hand side of (1) is

$$\begin{aligned} Q_c(c)(j) &:= Q_1(c)(j) - Q_2(c)(j) \\ &= \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k - c_j \sum_{k=1}^{\infty} a_{j,k} c_k, \quad j \in \mathbb{N}. \end{aligned} \quad (4)$$

The discrete Smoluchowski system is the system of a countable number of ordinary differential equations (1) with the right-hand side given by (4).

In several cases it is preferable to consider the version of Smoluchowski equations for which the cluster masses can be any positive real number. This continuous version, first considered by Hans Müller in 1928 [72, 164], can be written as the following integro-differential equation

$$\partial_t c(t, x) = Q_c(c)(t, x) \quad (5)$$

with  $c(t, x)$  the concentration (or density) of  $x$ -clusters at time  $t$ , and

$$\begin{aligned} Q_c(c)(t, x) &:= Q_1(c)(t, x) - Q_2(c)(t, x) \\ &:= \frac{1}{2} \int_0^x a(x-y, y) c(t, x-y) c(t, y) dy - c(t, x) \int_0^\infty a(x, y) c(t, y) dy. \end{aligned} \quad (6)$$

The first mathematical works about (1) seem to have been the papers by Brice McLeod [154] and [155]. On the continuous version (5) the first mathematical papers are those by Morgenstern [163] and Melzak [158] (this last one also including fragmentation, see Subsection 1.5). In the last two decades there has been a huge progress in our understanding of several questions about existence, uniqueness, regularity, and asymptotic behaviour of solutions to these equations, and part of these results will be presented below.

### 1.3 Oort-Hulst-Safronov coagulation equations

Another coagulation equation that has received some attention is the Oort-Hulst-Safronov equation [65, 177], [194, Chapter 8], that was first proposed to model astronomy phenomena. This equation has also the general form (5) but differs in the way  $Q_1(c)(t, x)$  and  $Q_2(c)(t, x)$  are defined:

1. the  $x$ -clusters creation rate depend on a kind of mean value size of the clusters, namely:

$$Q_1(c)(t, x) := -\partial_x \left( c(t, x) \int_0^x y a(x, y) c(t, y) dy \right); \quad (7)$$

2. the destruction of  $x$ -clusters due to the coagulation with other existing clusters only occurs through the reaction with higher masses, i.e., through a kind of sedimentation of smaller clusters onto larger ones:

$$Q_2(c)(t, x) := c(t, x) \int_x^\infty a(x, y) c(t, y) dy. \quad (8)$$

As a consequence, some solution properties are distinct from the corresponding ones in the Smoluchowski's system, the most notable of them is the finite speed of propagation of the support of solutions [75], which contrasts strongly to what happens in Smoluchowski's (see, for instance, [46] for the discrete case). Notwithstanding these differences in behaviour, both these equations are related and can be seen as limit cases of one-parameter families of cluster equations [75, 120].

### 1.4 Fragmentation equations

The first references to fragmentation processes took place in the context of chemical studies on polymer degradation (see, e.g., [197]). The first reference to the mathematical modelling of the spontaneous fragmentation process seems to have been done, using probabilistic methods, by Kolmogorov [112], who also suggested and supervised the later study [89] by A.F. Filippov. The first non-probabilistic mathematical reference to these processes is included in Melzak work [158] about the continuous coagulation-fragmentation system. For the discrete version the first reference seems to be the paper [206] by Spouge. As the spontaneous fragmentation process can be modelled by a linear equation (see below), the modern approach to these problems is intimately connected with tools and methods from Linear Functional Analysis, and an excellent introduction to them can be seen in [7, Chapters 8 and 9].

In this section we will consider the spontaneous fragmentation case; other processes, such as collisional fragmentation or volumetric dispersion, which are also related with coagulation processes will be presented later on in Subsection 1.7.

We can represent the fragmentation process of Figure 2 by<sup>3</sup>

$$(x) \xrightarrow{B(x)} (y_1) + (y_2) + \dots,$$

where  $B(x)$  is the rate of fragmentation of  $x$ -clusters. Let  $\psi(x, y)$  be the average number of  $y$ -clusters produced by the fragmentation of an  $x$ -cluster. In the case of discrete masses, denoted by  $i, j, k, \dots$ , we use the traditional notation  $B_j$  and  $\psi_{j,k}$ , instead of  $B(j)$  and  $\psi(j, k)$ .

Mass conservation in each simple fragmentation reaction implies that the total mass of daughter particles must be equal to the mass of the original particle, namely

$$\int_0^x y \psi(x, y) dy = x \quad \text{or} \quad \sum_{k=1}^{j-1} k \psi_{j,k} = j, \quad (9)$$

for the continuous and for the discrete cases, respectively.

For definiteness, let us consider the discrete case. Assuming the mass action law the rate of change of  $c_j$  due to spontaneous fragmentation processes is given by the differential equation

$$\dot{c}_j = Q_f(c)(j), \quad (10)$$

where  $Q_f(c)(j)$  encodes the fragmentation reaction contributions (hence the subscript  $f$ ) to the evolution of the  $j$ -cluster concentration, that can be expressed by

$$Q_f(c)(j) := -Q_3(c)(j) + Q_4(c)(j), \quad j \in \mathbf{N}, \quad (11)$$

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<sup>3</sup> The notation is not very good since it suggests there can be at most a countable number of daughter particles  $(y_k)$ : in fact, there is no *a priori* reason preventing the distribution to be continuous.



where

1. the destruction of  $j$ -clusters due to the fragmentation  $(j) \rightarrow (k) + \dots$ , is given by

$$Q_3(c)(j) := B_j c_j \quad \text{and} \quad Q_3(c)(1) := 0; \quad (12)$$

2. the creation of  $j$ -clusters due to fragmentation of bigger clusters is modelled by

$$Q_4(c)(j) := \sum_{k=1}^{\infty} B_{j+k} \psi_{j+k,j} c_{j+k}. \quad (13)$$

A frequent assumption, valid, for instance, in the degradation of polymers [228], is that of binary fragmentation, i.e., each fragmenting cluster produces only two daughter particles, and thus, by the symmetry of the physical process,  $\psi_{i,j} = \psi_{i,i-j}$ . Hence, (9) implies  $\sum_{k=1}^{j-1} \psi_{j,k} = 2$ , which has the obvious interpretation that the average number of daughter particles in each fragmentation is equal to two<sup>4</sup>.

In this case, denoting by  $b_{j,k}$  the rate constant for the binary fragmentation reaction  $(j+k) \rightarrow (j) + (k)$ , i.e.,  $b_{j,k} := B_{j+k} \psi_{j+k,k}$ , we conclude that  $B_j = \frac{1}{2} \sum_{k=1}^{j-1} b_{j-k,k}$  and thus the right-hand side of the binary fragmentation reaction is

$$Q_f(c)(j) := -\frac{1}{2} \sum_{k=1}^{j-1} b_{j-k,k} c_j + \sum_{k=1}^{\infty} b_{j,k} c_{j+k}. \quad (14)$$

As with the case of Smoluchowski's equations, the continuous versions of the fragmentation equations consist in integro-differential equations obtained formally by substituting the sums by integrals.

The fragmentation mechanism is mathematically encoded in the functions  $B(x)$ ,  $\psi(x,y)$  and  $b(x,y)$  and the only general property these functions must obey, on physical grounds, is non-negativity. Furthermore, the binary fragmentation coefficients must be symmetric:  $b(x,y) = b(y,x)$ .

In the mathematical literature it is common to assume growth conditions such as  $b(x,y) \leq (x+y)^\gamma$ , or  $b(x,y) \leq x^\gamma + y^\gamma$ , or  $b(x,y) \geq (x+y)^\gamma$ , etc., or other conditions, such as the *strong fragmentation* [34, 45], and the *weak fragmentation* [29, 30, 36, 48]. In models of some specific phenomena particular fragmentation kernels need to be considered (e.g.: see [103]).

An assumption that is particularly important from the physical viewpoint, corresponding to the occurrence of microscopic reversibility, is called the detailed balance condition. This presupposes the simultaneous existence of coagulation and fragmentation processes (cf. next section) and, informally, it says that it must exist an equilibrium (i.e., time independent) solution to each of the individual reactions

$$(j) + (k) \rightleftharpoons (j+k).$$

The detailed balance condition is the following: there exists a positive sequence  $(M_j)$ , with  $M_1 = 1$ , such that

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<sup>4</sup> As it should, in a binary fragmentation...

$$a_{j,k}M_jM_k = b_{j,k}M_{j+k}. \quad (15)$$

The sequence  $(M_j)$  is physically interpreted as the system's partition function [13, 36].

### 1.5 Coagulation-fragmentation equations

The coagulation-fragmentation equations are the system that describes phenomena where coagulation and fragmentation processes are simultaneously present. As such, it has the form

$$\dot{c}_j(t) = Q_c(c(t))(j) + Q_f(c(t))(j), \quad (16)$$

or

$$\partial_t c(t, x) = Q_c(c)(t, x) + Q_f(c)(t, x), \quad (17)$$

in the discrete and in the continuous case, respectively.

Possibly the first explicit reference to this system in the literature is in [23] treating phenomena of polymerization and de-polymerization in chemistry. The authors consider the discrete version of the equations, binary fragmentation, and reaction kernels independent of the cluster sizes,  $a_{j,k} \equiv a$ ,  $b_{j,k} \equiv b$ .

The first mathematical study about the existence of solutions is Melzak's 1957 paper cited in the last section [158], that considers the continuous system with bounded kernels. The extension to unbounded kernels started more than three decades latter in Iain Stewart's papers [210, 211]. For the discrete system the first existence result was published by John Spouge [206] in 1984, valid for bounded fragmentation coefficients. More general results were obtained by John Ball and Jack Carr [12], by Philippe Laurençot [126], by the author [45], among many others. Analogous existence results were obtained for similar equations modelling an Ising spin system with Glauber dynamics by Markus Kreer [116].

Long time behaviour of solutions to coagulation-fragmentation systems is a rather difficult problem, not yet completely understood. The first significant contribution was by Michael Aizenman e Thor Bak in 1979, for the continuous system with constant coefficients, [1]. In the last couple of decades a number of important papers have greatly advanced our understanding of the dynamic behaviour of solutions of both the discrete and the continuous coagulation-fragmentation equations. Some of these contributions will be analysed below.

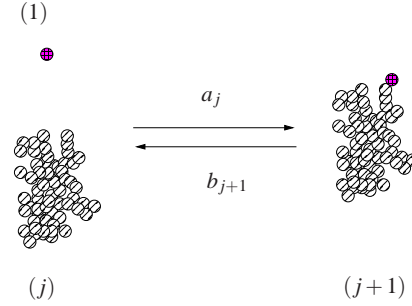
The vast majority of the mathematical studies have considered binary fragmentation, but some other types of fragmentation processes have also been considered [84, 85, 86, 139, 215] and we shall briefly refer to them in Section 1.7.

## 1.6 Becker-Döring equations

The original Becker-Döring model was proposed in 1935 in the context of nucleation studies [17] (formation of liquid droplets in a supersaturated vapour) in which the concentrations of large clusters are so small that one assumes the only relevant reactions are those of coagulation between a cluster and a monomeric particle, and the fragmentation of a cluster by shedding off a single monomeric particle at a time, as schematically illustrated in Figure 4.

Even at very low densities it is not physically reasonable to expect the Becker-Döring to be a good approximation [182]. However, the rich (and difficult) mathematics of the Becker-Döring system, together with the fact that some of its properties are believed to also hold in more general systems whose mathematical study is considerably more complex, have turned the Becker-Döring system into a paradigmatic model in coagulation-fragmentation studies whose importance and contribution to the understanding of the issues involved can hardly be overstated [202]. Even from the physical and mathematical modelling points of view, Becker-Döring like systems continue to this day to be proposed and studied [73, 107].

**Fig. 4** Scheme of the Becker-Döring processes.



In the original version of the Becker-Döring system the monomer concentration was assumed to be time independent. It was J.J. Burton [28] and Oliver Penrose and Joel Lebowitz [182] who first considered the current version of the equations, in which the mass of the system is formally constant and thus the monomer concentration has to change with time, and by this turning them into a non-linear system of the differential equations that is a particular case of the coagulation-fragmentation system with the rate kernels satisfying the restriction<sup>5</sup>

$$a_{j,k} = b_{j,k} = 0 \quad \text{if } j \wedge k > 1. \quad (18)$$

Due to historical reasons, the notation used in Becker-Döring system is slightly different from the one that would be obtained by substituting (18) into (4),(14)

<sup>5</sup> In this work we shall use the notation  $x \wedge y = \min\{x, y\}$  and  $x \vee y = \max\{x, y\}$  and analogously for the comparison of more than two numbers.

and the result into (16). For  $j > 1$  let us define  $a_j := a_{j,1}$  and  $b_{j+1} := b_{j+1,1}$ . let  $a_1 = \frac{1}{2}a_{1,1}$ ,  $b_2 = \frac{1}{2}b_{2,1}$ , and remember that the rate coefficients  $a_{j,k}$  e  $b_{j,k}$  are invariant under permutation of the subscripts. Thus, the Becker-Döring system is usually written as

$$\begin{cases} \dot{c}_1 = -J_1(c) - \sum_{j=1}^{\infty} J_j(c), \\ \dot{c}_j = J_{j-1}(c) - J_j(c), \quad j \geq 2, \end{cases} \quad (19)$$

where  $J_j(c) := a_j c_1 c_j - b_{j+1} c_{j+1}$ .

### 1.7 Equations with non-linear fragmentation

As pointed out in Section 1.1 there is a fragmentation mechanism that is quite different from the spontaneous fragmentation considered in subsections 1.4–1.6, and that mathematically originates non-linear contributions to the equations. There are some situations in astrophysics and atmospheric sciences where this non-linear fragmentation model has been used [194, 208, 209] and also in the mathematics literature there has been some interest (see, for example [42, 76, 213, 222]).

Considering the discrete case, assume that the collision between a  $k$ -cluster and a  $(j-k)$ -cluster can give rise to a  $j$ -cluster with probability  $w_{j-k,k}$ , or, with probability  $1 - w_{j-k,k}$ , to a variable number of daughter particles with total mass equal to  $j$ . Observe that, in contradistinction to spontaneous fragmentation, in this collisional fragmentation process some of the daughter particles can be *larger* than any of the original clusters.

As usual, the equations are of the type

$$\dot{c}_i = Q_d(c)(i) \quad (20)$$

where the reaction term has the following additive contributions:

1. formation of  $i$ -clusters by coagulation of smaller clusters of appropriate size, say  $i-j$  and  $j$ , with probability  $w_{i-j,j}$ , to which corresponds the term

$$\frac{1}{2} \sum_{j=1}^{i-1} w_{i-j,j} a_{i-j,j} c_{i-j} c_j; \quad (21)$$

2. destruction of  $i$ -clusters by their collision with any other cluster, independently of the final result be a coagulation or a fragmentation, which corresponds to a contribution  $Q_2(c)(i)$  given by (3);
3. the formation of an  $i$ -cluster as the result of a collision followed by instantaneous fragmentation, with probability complementary to the one above, whose term reads as

$$\frac{1}{2} \sum_{j=i+1}^{\infty} \sum_{k=1}^{j-1} \Psi_{j-k,k}^i (1 - w_{j-k,k}) a_{j-k,k} c_{j-k} c_k, \quad (22)$$

where  $\Psi_{j-k,k}^i$  gives the distribution of size  $i$  fragments produced by collisional fragmentation of  $j-k$  and  $k$ -clusters. Thus, this function is analogous to the function  $\psi$  in the spontaneous fragmentation models (cf. Section 1.4). Observe that  $\Psi$  has to satisfy the identity  $\Psi_{j,k}^i = \Psi_{k,j}^i$ , and, considering that each reaction conserves mass, also

$$\sum_{i=1}^{j+k-1} i \Psi_{j,k}^i = j + k.$$

Hence, the general coagulation-fragmentation system with collisional fragmentation (20) is

$$\begin{aligned} \dot{c}_i = & \frac{1}{2} \sum_{j=1}^{i-1} w_{i-j,j} a_{i-j,j} c_{i-j} c_j - c_i \sum_{j=1}^{\infty} a_{i,j} c_j + \\ & + \frac{1}{2} \sum_{j=i+1}^{\infty} \sum_{k=1}^{j-1} \Psi_{j-k,k}^i (1 - w_{j-k,k}) a_{j-k,k} c_{j-k} c_k. \end{aligned}$$

Naturally, to the right-hand side of this system one can add the spontaneous fragmentation term  $Q_f(c)(i)$  given by (11).

The specification of the functions  $\Psi_{j,k}^i$ ,  $w_{j,k}$  and  $a_{j,k}$  allows the modelling of particular cases of interest such as, for example, that considered in [209] where collisional fragmentation always produces only monomers.

The first mathematical study of these equations is due to Philippe Laurençot and Dariusz Wrzosek [139]. An analogous (continuous) system, with the imposition of a maximum cluster size was proposed by Antonio Fasano and co-workers in the context of liquid-liquid dispersions in chemical engineering [84, 85, 86] (see also [215]). Another similar system was considered in studies of polymerization reactions with catalysed fragmentation [111].

## 1.8 Diffusive coagulation-fragmentation equations

The previous approaches to coagulation-fragmentation processes assumed spatially homogeneous systems and so the cluster densities are independent of the spacial location. However, the spacial dependence of the cluster densities, and in particular the consideration of diffusive effects, has been recognized important in several situations [22, 70, 178].

The discrete version of these systems can be written as

$$\dot{c}_j = \nabla_z (d_j \nabla_z c_j) + Q_c(c)(j) + Q_f(c)(j), \text{ in } \Omega \times \mathbf{R}^+ \subseteq \mathbf{R}^n \times \mathbf{R}^+ \quad (23)$$

where the diffusion coefficients  $d_j = d_j(z, c)$  are non-negative functions, and adequate conditions are imposed to  $c_j = c_j(z, t)$  on the boundary  $\partial\Omega$ , or to their decay at infinity.

The first mathematical study about these systems seems to be by Marshall Slemrod [199], but the first general existence and uniqueness result (without fragmentation,  $Q_f(c)(j) \equiv 0$ ) is [19] by Philippe Bénilan and Dariuz Wrzosek. In the last decade a growing number of papers have been published about systems like (23), or its continuous analogue, dealing with existence, uniqueness, and behaviour of solutions. In this context the contributions by Amann, Laurençot, Mischler, Wrzosek, among others, are extremely important (see, for example, [3, 4, 5, 68, 78, 121, 129, 132, 137, 138, 224, 225]). In the present chapter we shall not further consider these works.

### 1.9 Equations with kinetic and transport terms

Another type of space dependence in these cluster equations is the introduction of transport terms, first considered in meteorological studies, in particular in models of cloud formation [20, 143]. The goal is to model the convection of clusters due to a given velocity field. We shall exemplify with the case studied in [74, Chapters 10 and 11]. Let  $z \in \Omega \subseteq \mathbf{R}^3$  denote the space variable,  $v(z, t, x) \in \mathbf{R}^3$  the velocity of the  $x$ -cluster at time  $t$  and position  $z$ , and let  $r(z, t, x) \in \mathbf{R}$  be the rate of change of the concentration  $c(z, t, x)$  by condensation or evaporation of droplets of size  $x$  in the space-time location  $(z, t)$ . The equation, first proposed by Levin and Sedunov, and by Berry, is

$$\partial_t c(z, t, x) + \partial_x(r(z, t, x)c(z, t, x)) + \nabla_z(v(z, t, x)c(z, t, x)) = Q_c(c)(t, x), \quad (24)$$

where  $Q_c(c)$  is defined by (6), with the concentrations also dependent of the  $z$  variable, but the coagulation kernel only dependent on the masses.

In this model the velocity field is an “exterior” field where the clusters are embedded. In particular, the coagulation reactions are not influenced by the velocity field  $v$ . A different possibility is to consider the coagulation process depending on the local velocity, that is, considering the velocity field not as some field carrying the clusters, but essential as the field that describes the local velocity of each cluster. This more detailed model, analogous to the viewpoint used in kinetic theory, was first considered in the context of discrete velocity models by Slemrod and co-workers [201, 203], and, more recently, in the general case, among others by Escobedo, Laurençot and Mischler in [79] where they proved results on global existence of weak solutions and their convergence as  $t \rightarrow \infty$ .

Let  $c(z, t, x, p)$  be the concentration of  $x$ -clusters with linear momentum  $p$ , and located at  $z$  at time  $t$ . The system studied in [79] is

$$\partial_t c + v \cdot \nabla_z c = Q_c(c), \quad (25)$$

where  $v = p/x$ , and the coagulation term  $Q_c = Q_1 - Q_2$  is defined by

$$\begin{aligned} Q_1(c) &:= \frac{1}{2} \int_{\mathbf{R}^3} \int_0^x a(y', y - y') c(\cdot, \cdot, y') c(\cdot, \cdot, y - y') dx' dp' \\ Q_2(c) &:= \int_{\mathbf{R}^3} \int_0^x a(y, y') c(\cdot, \cdot, y) c(\cdot, \cdot, y') dx' dp' \end{aligned}$$

where  $y := (x, p) \in \mathbf{R}^+ \times \mathbf{R}^3$ , etc.

A similar model, without the space dependence, was considered before by Baranger in [15] and by Roquejoffre and Villedieu in [193]. Still another model was studied by Nicolas Fournier and Stéphane Mischler [94, 95], in which, although there is also no spacial dependence, there are, additionally to the binary collisions resulting in coagulating events, other binary elastic collisions (modelled by Boltzmann collision operator) and inelastic collisions (modelled by a granular collision operator).

Naturally, the analysis of this type of equations makes use of methods closely related to those used in studies of Boltzmann's and related kinetic equations. In this chapter we will not consider these works further.

## 1.10 Other models

Other models have been considered in the literature. We shall now describe some of them.

### 1.10.1 Multi-index cluster models

In the systems of previous sections clusters were characterized by a single “internal” quantity, their mass or volume, and, in some cases, by some “external” ones, such as the spacial position or velocity. However, for some applications one needs to characterize the existing clusters by additional variables identifying relevant physical quantities.

One obvious case is in co-polymerization reactions when there are two monomeric species,  $A$  and  $B$  say, and it is important to keep track of the way a given cluster is made, not only by the total number of monomeric particles, but accounting for how many of each monomeric species a given cluster is formed. Thus, in the simplest situation a cluster has to be described by a vector subscript  $(i_A, i_B)$  informing that the cluster is made by  $i_A$  units of the monomeric species  $A$  and by  $i_B$  units of  $B$ . This approach was used in kinetic studies of micelles and vesicle formation (cf. e.g. [64]) where the following two component Becker-Döring like system was proposed:

$$\frac{d}{dt} c_{i,j} = J_{i-1,j}^A(c) - J_{i,j}^A(c) + J_{i,j-1}^B(c) - J_{i,j}^B(c), \quad i, j \in \mathbf{N}^+ \setminus \{1\},$$

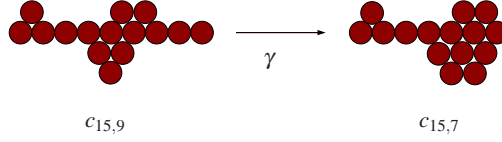
with the terms of microscopic balance for monomer  $A$  given by  $J_{i,j}^A(c) := a_{i,j}c_{1,0}c_{i,j} - b_{i+1,j}c_{i+1,j}$  and those for  $B$  by  $J_{i,j}^B(c) := \alpha_{i,j}c_{0,1}c_{i,j} - \beta_{i,j+1}c_{i,j+1}$ , with the meaning of the symbols analogous to that in the usual Becker-Döring equation (19).

A similar case occurs when the clusters are made of two phases of the same substance and one needs to keep track of the quantities of each of them. An example is presented in [186], where it is considered that each particle has a continuously varying mass  $x$ , of which  $\alpha \leq x$  is the mass of one of the phases (ice or liquid water). Using a notation analogous to that in Section 1.2, the coagulation operator correspondent to (6) is now

$$\begin{aligned} Q_c(c)(x, \alpha) &= \frac{1}{2} \int_0^x \int_0^\alpha a(x-y, \alpha-\beta; y, \beta) c(\cdot, x-y, \alpha-\beta) c(\cdot, y, \beta) dy d\beta - \\ &= -c(\cdot, x, \alpha) \int_0^\infty \int_0^\infty a(x, \beta; y, \beta) c(\cdot, y, \beta) dy d\beta. \end{aligned}$$

Other model requiring a multi-index is considered in [220], where each cluster is characterized by its mass  $j$  and also by another subscript  $k \leq j$  describing its shape in the sense that it reflects its diameter. In this case a cluster can not only be subject to coagulation and fragmentation reactions, but also to internal rearrangement “reactions” that are a mere change in its geometry, as Figure 5 attempts to exemplify.

**Fig. 5** An example of an internal geometric rearrangement “reaction” of a  $(15, 9)$ -cluster to a  $(15, 7)$ -cluster, at a rate  $\gamma c_{15,9}$ .



A further model of this type was used in the study of surface capping in cell-antibody interactions [37, 63]. In this case a  $j$ -cluster can be represented by a graph for which each of the  $j$  nodes stands for a monomeric unit in the cluster. All nodes are potentially of maximum degree three but not all of them have this valency at a given particular time. The  $k < j$  nodes with degree one (the leaves of the graph) are particularly important in this model and so the clusters are characterized by the pair  $(j, k)$  and their dynamic was studied using an adapted version of the Becker-Döring system.

Finally, another case with an associated graph (in this case a tree) was studied in [56], motivated by the study of self-organized criticality in [100]. The model consists in a coagulation system for the evolution of clusters described by a pair  $(p, q)$ , where  $p$  is the “order” and  $q$  its mass, and where the reactions are schematically represented by

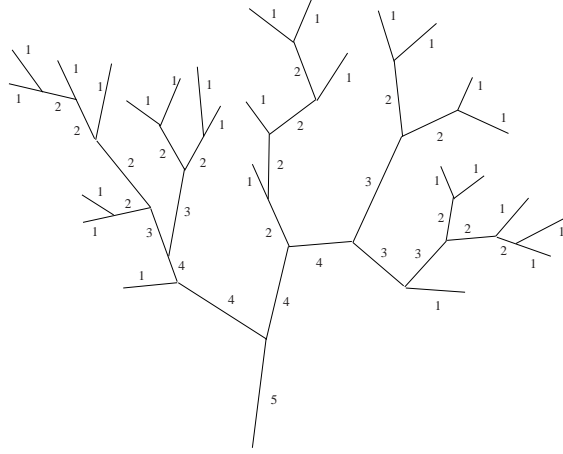
$$(i, j) + (k, m) \rightarrow (\vee(i, k, (i \wedge k) + 1), j + m).$$

This means that the mass satisfies the usual additivity, and the order satisfy the Horton-Strahler rules. Each cluster is represented by an edge of a tree and a reaction between two clusters corresponds to the respective edges concurring in a node (cf.



Figure 6). In [56] a coagulation system for the time evolution of the concentrations

**Fig. 6** Illustration of the Horton-Strahler rules in the orders of the edges of a tree.



$c_{i,j}$  of the  $(i,j)$ -clusters is studied, as well as the evolution of some mesoscopic quantities, like the total number of clusters of a given order.

### 1.10.2 Annihilation models

We now briefly consider a class of systems in which clusters are also made up of two monomer species but merit a reference outside Section 1.10.1 because part of the physical processes involved are significantly different from the usual coagulation in that cluster can annihilate each other. The two-species coagulation-annihilation system describe the time evolution of the concentration of clusters of two different particle species ( $A$  and  $B$ , say) in which the  $A$ -particle clusters [resp.,  $B$ -particle clusters] undergo coagulation between themselves, symbolically

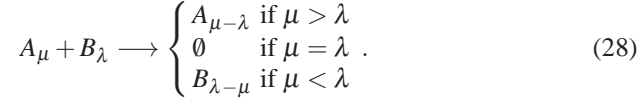
$$A_\mu + A_\lambda \xrightarrow{K_a} A_{\mu+\lambda} \quad [\text{resp.}, B_\mu + B_\lambda \xrightarrow{K_b} B_{\mu+\lambda}], \quad (26)$$

but when an  $A$ -particle cluster and a  $B$ -particle cluster come together, they annihilate each other, and in the simplest such model the annihilation is complete, i.e., for all  $\mu$  and  $\lambda$ ,

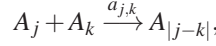
$$A_\mu + B_\lambda \xrightarrow{L} \emptyset, \quad (27)$$

where  $\emptyset$  represents a physically inert species. In the physics literature these processes have been approached through a variety of techniques. Using a mass action approach as in coagulation studies, one of the first works seems to be Ben-Naim and Krapivsky [18] where, for the case of discrete cluster sizes, it is assumed that reactions rates are independent of the cluster sizes and all have the same value,  $K_a(\mu, \lambda) = K_b(\mu, \lambda) = L(\mu, \lambda) = 2$ . In that work, the authors inves-

tigated the time evolution of the system and the existence, or non-existence, of a universal similarity behaviour of the solutions. More recently, Laurençot and van Roessel [135] considered these same issues in the case of continuous cluster sizes with reaction rates still independent of the cluster sizes but with the coagulation rates  $K_a(\mu, \lambda) = K_b(\mu, \lambda) = k$  possibly different from the annihilation kernel  $L(\mu, \lambda) = L$ , a case that had already been considered by Krapivsky in [115] for the discrete case with  $k = 2$ . Still within the context of rate coefficients independent of cluster sizes, in [61] da Costa, Pinto, van Roessel, and Sasportes extended [135] by considering the possibility of the coagulation rates of  $A$ -clusters and of  $B$ -clusters to be different from each other, i.e.,  $K_a(\mu, \lambda) = K_a$ ,  $K_b(\mu, \lambda) = K_b$ , and  $L(\mu, \lambda) = L$ , where  $K_a, K_b, L$  are positive constants, otherwise unrestricted. A slightly more general process has also been proposed consisting in an incomplete annihilation between  $A$  and  $B$ -particles (see, e.g., [115]), which means that the  $A$  and  $B$  species still annihilate each other but only to the extent corresponding to their respective sizes, which is represented schematically by

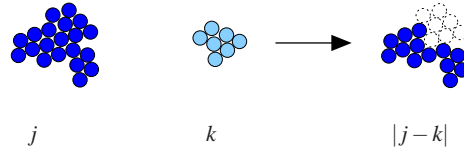


Coagulation-annihilation with incomplete annihilation are hard to analyse mathematically and the method used for the complete annihilation case, based on Laplace transform techniques, do not seem to work. This difficulty has led to the consideration of some toy models that still show some interesting behaviour. One such model was first introduced by Redner, Ben-Avraham and Kahng in [191] and a similar but more general one was introduced in [108]. These toy models retain the incomplete annihilation process but get rid of both, the two different monomeric species, and the coagulation reactions. The process is schematically represented by



where  $A_0 := \emptyset$ . Still assuming that there is no destruction of mass in each individual reaction, it now makes more sense to think of  $j$  as the size of the cluster “active part”, being the difference between  $j+k$  and  $|j-k|$  the size of the resulting cluster that has become inactive after the reaction. One illustration of this is in Figure 7

**Fig. 7** Schematic reaction in the RBK coagulation-annihilation model.



The dynamics of this cluster system is governed by the following equations, called the RBK cluster system in [59],

$$\dot{c}_j = \sum_{k=1}^{\infty} a_{j+k,k} c_{j+k} c_k - \sum_{k=1}^{\infty} a_{j,k} c_j c_k, \quad j = 1, 2, \dots, \quad (29)$$

whose mathematical study started only recently [59, 60]. It is worth noticing the similarities, and also the differences between the RBK system (29) and Smoluchowski's coagulation equations.

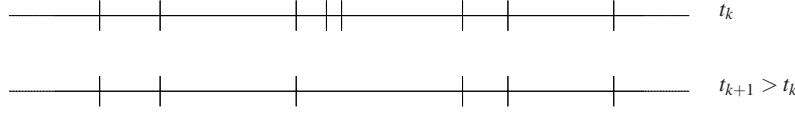
### 1.10.3 Coagulation of intervals in the real line

The models considered in this section are toy models for maturation and ageing processes in physical systems far from equilibrium and have been considered extensively in the Physics literature. Below we shall concentrate exclusively on mathematical works. It is also interesting to remark that some of these models can be seen as a kind of “dual” processes to the RBK system with mono-disperse initial data [108].

The first of these models was studied in 1992 by Jack Carr and Robert Pego [39]. Their motivation was the studies of metastability in solutions to reaction-diffusion systems of Chafee-Infante type  $u_t = \varepsilon^2 u_{xx} + u - u^3$  in the bounded interval  $(0, 1)$  with homogeneous Neumann conditions at the boundary, and very small diffusion coefficient  $\varepsilon$ . This rather interesting behaviour had been discovered and studied by the same authors and by Giorgio Fusco e Jack Hale in a series of remarkable papers (cf. [38, 98] for an introduction to those results). It consists in the fact that a typical solution rapidly approaches functions that, in spite of not being equilibria (and being far from one) are practically time independent for an extraordinarily large interval of time (of the order  $e^{1/\varepsilon}$ ). The graph of these functions are essentially constant but for what happens in the neighbourhood of a finite number  $N$  of points of  $(0, 1)$ , where very sharp transitions take place. When, due to the extra slow dynamics, two of these transition layers finally come close to one another, the dynamics has a markedly increase in its speed in such a way that the transition layers suddenly collapse and disappear, after which the dynamics returns to its exponentially slow pace.

The mean field coagulation like model [39, 99] for this behaviour was first derived by [165] and is the following: consider  $N \gg 1$  points arbitrarily chosen in the interval  $(0, 1)$  (these points represent the location of the transition layers in the solution to the reaction-diffusion equation); assume the following process with discrete time: in each time unit look for the shortest interval in the partition of  $(0, 1)$  defined by the  $N$  chosen points and eliminate the two points that are its boundary, thus producing the fusion of that interval with its nearest neighbours (this corresponds to the quick collapse of the transition layers) and having as a result the reduction of the number of points to  $N - 2$  in the next time unit (cf. Figure 8). Denoting by  $f(x, t)$  the density, at time  $t$ , of the distribution of the number of intervals by unit length, the total number of intervals by unit length is

$$N(t) = \int_0^{\infty} f(x, t) dx.$$



**Fig. 8** A schematic interval's coagulation process.

Let  $\mathcal{L}(t)$  be the smallest interval at time  $t$ . The time evolution of  $f$  due to the process described above has the following two contributions:

1. formation of an interval with length  $x$  by coalescence of an interval of length  $\mathcal{L}(t)$  with two intervals of lengths  $y$  and  $x - y - \mathcal{L}(t)$ ,
2. disappearance of an interval of length  $x$  by coalescence with any other interval.

The differential equation for  $f$  is

$$\partial_t f(x, t) = \frac{f(\mathcal{L}(t), t) \dot{\mathcal{L}}}{N^2(t)} \left[ \int_0^\infty f(y, t) f(x - y - \mathcal{L}(t), t) dy - 2f(x, t) N(t) \right], \quad (30)$$

where  $f(x, t) = 0$  if  $x < \mathcal{L}(t)$ . In [39] the time scale was chosen so that the expected number of coagulation events per unit time is  $f(\mathcal{L}(t), t) \frac{d\mathcal{L}}{dt} = 1$ . In [99] the time was parametrized by the smallest interval size, i.e.,  $\mathcal{L}(t) = t$ , and the system was written for the probability density  $\rho_t(x) := f(x, t)/N(t)$ , instead of  $f$ .

A more general model, also studied by Carr and Pego [40], is a generalization of previous models by Derrida, Godrèche, and Yekutieli [67] and by Pesz and Rodgers [185]. The difference relative to the previous model is that now, in each unit of time, the smallest interval is divided in  $\alpha^{-1}$  parts according to a probability density  $d\nu(\alpha)$  and these parts are randomly redistributed by the remaining intervals in the partition.

Let  $X(t)$  be the size of the smallest interval at time  $t$ ,  $\varphi(x, t)$  be the expected number of intervals with length larger than or equal to  $x$  at time  $t$  normalize by the initial number of intervals, and let  $N(t)$  be the normalized total number of intervals at time  $t$ . Then, the dynamics is determined by the equation

$$\partial_t \varphi(x, t) = -\frac{\dot{N}(t)}{N(t)} \int_0^\infty \left( \varphi(x - \alpha X(t), t) - \varphi(x, t) \right) \alpha^{-1} d\nu(\alpha)$$

subject to

$$N(t) = \varphi(x, t), \quad \text{for } -\infty < x \leq X(t).$$

Another model was proposed and rigorously studied by Menon, Niethammer and Pego [162]. The corresponding process is the following: at each time step choose an integer  $k \geq 1$  at random with probability  $p_k$ , and merge the smallest interval with  $k$  randomly chosen intervals.

With  $\rho_t^{*k}$  denoting the  $k$ -fold self convolution of  $\rho_t$ , and the remaining variables as above, the dynamics is described by the equation

$$\partial_t f(x, t) = f(\mathcal{L}(t), t) \mathcal{L} \sum_{k=1}^{\infty} p_k \left( \rho_t^{*k}(x - \mathcal{L}) - k \rho_t(x) \right), \quad \text{with } x > \mathcal{L}(t).$$

In [162] the time scale was taken as  $t = N(t)^{-1}$ , and the analysis was based on the method of Gallay and Mielke [99].

#### 1.10.4 Proliferation models in population dynamics

The mathematical studies about proliferation processes in biological populations, being them of individuals, cells, or biochemical molecules, have resulted in an appreciable diversity of differential equation used as models [184].

One of these equations, to model the time evolution of a cell population undergoing mitosis, by which a cell of size  $x$  is broken into two of sizes  $x/2$  at a rate  $B(x)$ , is the following [184, Chapter 4]

$$\partial_t n(x, t) + \partial_x n(x, t) = -B(x)n(x, t) + 4B(2x)n(2x, t),$$

where  $n(x, t)$  is the density of cells of size  $x$  at time  $t$ . It is possible to generalize this process by assuming that a cell can be broken into  $\alpha$  equal daughter cells with sizes  $x/\alpha$  [55]. On the other hand, if the fragmentation process allows the two daughter cells to have distinct sizes, the differential equation for the density  $n(x, t)$  has the typical form of a fragmentation equation with mass transport [184] (cf. sections 1.4 and 1.9 above):

$$\partial_t n(x, t) + \partial_x n(x, t) = -B(x)n(x, t) + \int_x^{\infty} b(x, y)n(y, t)dy.$$

In order to model more specific situations, the mathematical models can become correspondingly more complex. As an example that has recently received some attention, we can point to models of growth and proliferation of prions (i.e., of proteins with transmissible pathological conformations) responsible for the Bovine Spongiform Encephalopathy (“Mad Cow Disease”) [102, 187] and several mathematical models have already been object of a rigorous analysis [136, 198, 216]. According to the contemporary biological understanding, there are two basic prion forms, a normal, non-infectious, monomeric one (denoted by  $\text{PrP}^{\text{C}}$  in the literature) and an infectious polymeric form ( $\text{PrP}^{\text{Sc}}$ ) formed by the polymerization of the monomeric form. Above a certain critical size  $n$ , the  $\text{PrP}^{\text{Sc}}$  seems to have the strong tendency to rapidly bond with the monomers. The  $\text{PrP}^{\text{Sc}}$  has also break up into polymers below the critical size that are quickly degraded into  $\text{PrP}^{\text{C}}$  monomers. Denoting by  $y_0(t)$  the  $\text{PrP}^{\text{C}}$  concentration and by  $y_i$  the concentration of  $\text{PrP}^{\text{Sc}}$  polymeric chains made up of  $i$  monomers, the differential equation model is the following [102, 187]

$$\begin{aligned}\dot{y}_0 &= \lambda - dy_0 - y_0 \sum_{i=n}^{\infty} \beta_i y_i + 2 \sum_{j=1}^{n-1} \sum_{i=n+j}^{\infty} j b_i y_i + 2 \sum_{j=1}^{n-1} \sum_{i=n}^{n+j-1} i b_i y_i \\ \dot{y}_i &= \beta_{i-1} y_0 y_{i-1} - \beta_i y_0 y_i - a_i y_i - (i-1) b_i y_i + 2 \sum_{j=i+1}^{\infty} b_j y_j,\end{aligned}$$

where  $a_i, b_i, \beta_i, \lambda \in \mathbb{R}^+$  are positive constants. Continuous mass versions of these equations were also considered in the literature [102, 136, 198, 216].

### 1.11 Other problems about coagulation and fragmentation models: relation with particle models

To finish this introductory part, we will refer to a different type of mathematical studies of coagulation and fragmentation equations. So far, the mathematical works that we have referred to were those that, starting with a given differential equation, have as goal the study of (some) properties of its solutions (say: existence, uniqueness, regularity, mass conservation, long time behaviour, self-similarity). In the following sections of this chapter this is also the theme we will be interested in, but in the present section we consider another important class of problems that have attracted some attention: starting from more fundamental non-equilibrium Statistical Physics assumptions in terms of stochastic processes, to obtain the coagulation equations as some kind of thermodynamic limit of these processes. Here we will just present, in a brief way adapted from [192], the kind of approach used, and direct the interested reader to the works of, among others, Flavius Guiaş [105], James Norris [175], Großkinsky and co-workers [104], Vassili Kolokoltsov [114], Fraydoun Rezakhanlou [192], and Nicolas Fournier and co-workers [66, 92].

In the microscopic model one initially considers a collection of  $N \gg 1$  particles randomly distributed in points  $x_i \in \mathbb{R}^d$ , with  $d \geq 2$  and  $i \in I = \{1, 2, \dots, N\}$ . Each particle has an integer mass  $m_i \in \mathbb{N}^+$  and is animated with a Brownian motion with diffusion constant  $2d(m_i)$ . When two particles of masses  $m_i$  and  $m_j$  are at a distance from one another equal to  $\|x_i - x_j\| = \varepsilon > 0$  they can coagulate to make a particle of mass  $m_i + m_j$ , randomly located in any of the positions  $x_i$  or  $x_j$ , with probability dependent of the masses of the original particles<sup>6</sup>; Due to this coagulation process the number of particles in the system diminishes with time and so the indexing set is time dependent,  $I_{q(t)} \subset I$ . One assumes that the dynamics of this particle system  $q(t) := \{(x_i(t), m_i(t)) | i \in I_{q(t)}\}$  is a Markov process with infinitesimal generator  $\mathcal{L} = \mathcal{A}_{\text{dif}} + \mathcal{A}_{\text{c}}^\varepsilon$ , where  $\mathcal{A}_{\text{dif}}$  is the contribution of the Brownian motion between collisions, and  $\mathcal{A}_{\text{c}}^\varepsilon$  is the coagulation term. For this microscopic process one defines the empirical measure

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<sup>6</sup> In other versions of this coagulation process of stochastic particles it is assumed the resulting particle is located at the centre of mass  $\frac{x_i m_i + x_j m_j}{m_i + m_j}$  [176], in still others coagulation can happen within a whole interval of distances between the particles and not only at the distance  $\varepsilon$  [104]

$$g_n(dx, t) = \frac{1}{K_\varepsilon} \sum_i \delta_{x_i(t)}(dx) \mathbf{1}(m_i(t) = n).$$

Being  $K_\varepsilon \xrightarrow{\varepsilon \rightarrow 0} \infty$  an appropriate scaling factor and  $Z$  a constant (the total macroscopic density), one can prove that in the thermodynamic limit, i.e., when  $N \rightarrow \infty$  keeping  $N/K_\varepsilon = Z$ , the measure  $g_n(dx, t)$  converges to a measure  $c_n(x, t)dx$  in the following sense

$$\lim_{N \rightarrow \infty} \mathbf{E}_N \left| \int_{\mathbf{R}^d} J(x, t) (g_n(dx, t) - c_n(x, t)dx) \right| = 0,$$

for all test functions  $J$  bounded and continuous in  $\mathbf{R}^d \times [0, \infty)$ . The density  $c_n(x, t)$  of the limit measure solves the coagulation equation with diffusive terms (23).

## 2 Existence and uniqueness of solutions to discrete coagulation-fragmentation systems

In this section we shall review results about existence and uniqueness of solutions to coagulation-fragmentation systems, with special emphasis to the discrete case. Note, however, that most of the results in one case have equivalent in the other, and a rigorous relation between the two can be established [131]. We start with the special case of Smoluchowski's coagulation equation because of its importance, historically and conceptually. Most of the section will be devoted to existence results. Uniqueness will be treated in the last part.

We start by briefly presenting the most relevant spaces needed afterwards.

### 2.1 Finite density spaces

With the notation introduced in Subsection 1.2, let  $c_j(t)$  be the concentration of  $j$ -clusters at time  $t$  and, without loss of generality, assume the mass of a  $j$ -cluster is  $j$ . Thus, the quantity  $\rho(t) := \sum_{j=1}^{\infty} j c_j(t)$  can be interpreted as the total density of the system (total mass, assuming the volume is constant) and it is reasonable to impose that solutions to (16) must have finite density, which means that, for all  $t \geq 0$ , the solution must be an element of the Banach space  $X_1 \subset \ell^1$  of finite density solutions defined by

$$X_1 := \left\{ c = (c_j) \in \mathbf{R}^{\mathbf{N}^+} : \|c\|_1 := \sum_{j=1}^{\infty} j |c_j| < \infty \right\}. \quad (31)$$

In many situations it is important to consider other Banach spaces, namely

$$X_\alpha := \left\{ c = (c_j) \in \mathbf{R}^{\mathbf{N}^+} : \|c\|_\alpha := \sum_{j=1}^{\infty} j^\alpha |c_j| < \infty \right\}, \quad \alpha \geq 0. \quad (32)$$

Some of these spaces have also physical meaning, for example, the norm in  $X_0 = \ell^1$  is a quantity proportional to the total number of clusters. Due to the physical meaning associated to the coagulation and fragmentation equations we will consider only non-negative solutions, i.e., those remaining in the non-negative cone of the relevant space  $X_\alpha$ ,

$$X_\alpha^+ := \left\{ c \in X_\alpha : c_j \geq 0, \forall j \right\}. \quad (33)$$

It is not hard to prove [44, Theorem 1.2.1] that the spaces  $X_\alpha$  with the norms  $\|\cdot\|_\alpha$  constitute a compact and normal scale of Banach spaces, which means that, for all  $\beta > \alpha \geq 0$ ,  $X_\beta \subset X_\alpha$  with the inclusions being continuous, dense, and compact, and for all  $c \in X_\beta$  it holds that  $\|c\|_\alpha \leq \|c\|_\beta$ , and the following interpolation inequality is also valid

$$\forall 0 \leq \alpha < \beta < \gamma, \forall c \in X_\gamma, \quad \|c\|_\beta^{\gamma-\alpha} \leq \|c\|_\alpha^{\gamma-\beta} \|c\|_\gamma^{\beta-\alpha}.$$

This scale is also regular, meaning that the norm of the dual spaces  $X'_\alpha$  is a logarithmically convex function of the parameter  $\alpha$ , but this result is not needed in what follows.

For the continuous version of the coagulation-fragmentation equations (17), where the cluster masses are in  $\mathbf{R}^+ = (0, \infty)$ , one defines the relevant spaces in an analogous way, but with the difference that the need to control what happens to very small clusters, and the non-existence of an inclusion relation in the  $L^p(\mathbf{R}^+)$  spaces similar to what exists in the  $\ell^p$ , leads to the following finite density space

$$Y_1 := L^1(\mathbf{R}^+, (1+y)dy) = L^1(\mathbf{R}^+, dy) \cap L^1(\mathbf{R}^+, ydy),$$

where  $dy$  is the Lebesgue measure on  $\mathbf{R}$ . The norm in this space is

$$\|\cdot\|_{Y_1} := \|\cdot\|_{L^1(\mathbf{R}^+, dy)} + \|\cdot\|_{L^1(\mathbf{R}^+, ydy)}.$$

## 2.2 Discrete Smoluchowski equations

Let us consider the Cauchy problem for Smoluchowski's coagulation system (1)-(4),

$$\begin{aligned} \dot{c}_j &= \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k - c_j \sum_{k=1}^{\infty} a_{j,k} c_k, \\ c_j(0) &= c_{j0}, \end{aligned} \quad (34)$$

The approach to questions of existence to (34) that have been most fruitful so far consists in its approximation by finite  $n$ -dimensional truncations for which one can prove that their solutions  $c^n(t)$  approach, in an adequate sense, a function  $c(t)$  which can be proved to be a solution of the infinite dimensional system (34). This approach was used from the very first mathematical works, in the coagulation sys-



tem by McLeod [154], in the coagulation-fragmentation systems by Spouge [206] and in the Becker-Döring, by Ball, Carr and Penrose [13].

A different approach that has been occasionally used in continuous coagulation-fragmentation systems consists in the use of fixed point theorems and operator semi-group theory, techniques that were pioneered by Melzak [158] and by Aizenman e Bak [1]. The approach using semi-group theory has been very successful in the study of (linear) fragmentation systems (cf., for example, [14, 153]).

In the present chapter we will only use the approach based on truncation. There are essentially two finite  $n$ -dimensional truncations used in the literature: the  $n$ -maximal truncation and the  $n$ -minimal truncation, in the designation introduced in [49]. The first one corresponds to the following system of  $n$  ordinary differential equations for the phase space vector  $(c_1, c_2, \dots, c_n)$ :

$$\dot{c}_j = \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k - c_j \sum_{k=1}^{n-j} a_{j,k} c_j c_k, \quad j \in \{1, \dots, n\}. \quad (35)$$

The second corresponds to the system

$$\dot{c}_j = \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k - \sum_{k=1}^n a_{j,k} c_j c_k, \quad j \in \{1, \dots, n\} \quad (36)$$

for the same phase space vector. A  $2n$ -dimensional truncation analogous to the  $n$ -minimal truncation, for which the  $2n$ -dimensional vector is  $(c_1, c_2, \dots, c_{2n})$ , is the following, [123]:

$$\begin{aligned} \dot{c}_j &= \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k} c_k - \sum_{k=1}^n a_{j,k} c_j c_k, \quad j \in \{1, \dots, n\} \\ \dot{c}_j &= \frac{1}{2} \sum_{k=j-n}^n a_{j-k,k} c_{j-k} c_k, \quad j \in \{n+1, \dots, 2n\}. \end{aligned} \quad (37)$$

The starting point of the analysis consists in considering an appropriate and rigorous version of the formal identity, which is a weak version of the coagulation equation:

$$\sum_{j=1}^{\infty} g_j c_j(t) - \sum_{j=1}^{\infty} g_j c_j(\tau) = \frac{1}{2} \int_{\tau}^t \sum_{j=1}^{\infty} \sum_{k=1}^{\infty} (g_{j+k} - g_j - g_k) a_{j,k} c_j(s) c_k(s) ds, \quad (38)$$

where one assumes that  $0 \leq \tau \leq t$ , and  $(g_j)$  is a non-negative test sequence.

From this equality (or for a rigorous version of it) one extracts the *a priori* estimates needed for the proofs. For instance, from (38) one can infer that the only *a priori* estimate expected to hold in  $X_1^+$  is the boundedness of density (corresponding to the test sequence  $g_j = j$ ), and also that the stronger the growth rate of the coefficients, the harder to get the estimates one needs and the more likely it is to expect the need of some control via assumptions on the higher moments  $M_p(c) := \sum_{j=1}^{\infty} j^p c_j(t)$ ,  $p > 1$ . (Note that for non-negative sequences  $M_p(c) = \|c\|_p$ .)

Before proceeding it is necessary to be precise about what is meant by a solution [12, 123]:

**Definition 1** *Let  $T \in (0, +\infty]$  and  $c_0 = (c_{j0}) \in [0, +\infty)^{\mathbf{N}^+}$ . A solution  $c = (c_j)$  of (34) in  $[0, T)$  is a sequence of non-negative continuous functions satisfying  $\forall j \geq 1$  and  $\forall t \in (0, T)$ ,*

- (i)  $c_j \in C([0, T))$
- (ii)  $\sum_{j=1}^{\infty} a_{j,k} c_j \in L^1(0, t)$
- (iii)  $c_j(t) = c_{j0} + \int_0^t \left( \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k}(s) c_k(s) - \sum_{k=1}^{\infty} a_{j,k} c_j(s) c_k(s) \right) ds$

A problem that immediately arises is to know if solutions with non-negative [resp. positive] initial data remain non-negative [resp. positive] for all later times. This problem was first studied in the Becker-Döring case [13], and afterwards in the coagulation-fragmentation in [34, 36]. For the Smoluchowski coagulation system the following result was proved in [46]:

**Theorem 1** [46] *Let  $a_{j,k} > 0$  for all  $j, k$ . Take any  $c_0 \in X_1^+$  and let  $c$  be a solution of (34) in  $[0, T) \subset [0, +\infty)$ . For each  $t \in [0, T)$ , let  $\mathcal{J}(t)$  be the set of subscripts  $j$  for which  $c_j(t) > 0$ . Then  $\mathcal{J}(t) \equiv \mathcal{J}$  is independent of  $t$  and is given by  $\mathcal{J} = \text{span}_{\mathbf{N}_0}(\mathcal{J}(0)) := \left\{ j = \sum_i n_i p_i : p_i \in \mathcal{J}(0), n_i \in \mathbf{N}_0, \max_i n_i > 0 \right\}$ .*

It is easy to conclude from this result that if, for some subscript  $p$ , we have  $c_p(0) > 0$  then it is always true that  $c_p(t) > 0$  for all  $t > 0$ . On the other hand, the proof of the result implies that if  $c_p(0) = 0$  then, either  $c_p(t) = 0, \forall t > 0$  (if  $p \notin \text{span}_{\mathbf{N}_0}(\mathcal{J}(0))$ ), or it will be always positive (if  $p \in \text{span}_{\mathbf{N}_0}(\mathcal{J}(0))$ ).

The proof of the theorem uses in a fundamental way the following way to write Smoluchowski's equation:

$$c_j(t)E_j(t) = c_\tau E_j(\tau) + \int_\tau^t E_j(s)R_j(s)ds \quad (39)$$

where

$$E_j(t) := \exp \left( \int_0^t \sum_{k=1}^{\infty} a_{j,k} c_k(s) ds \right), \quad R_j(t) := \frac{1}{2} \sum_{k=1}^{j-1} a_{j-k,k} c_{j-k}(t) c_k(t),$$

and  $R_1(t) \equiv 0$ .

These positivity results are also relevant for the coagulation-fragmentation systems, for which this method (with the obvious modification in the definitions of  $E_j$  and  $R_j$ ) was first used in [34, 36], where it was also used to prove that, for all  $t > 0$ , all components  $j$  of  $(c_j(t))$  are strictly positive (i.e.,  $\mathcal{J} \equiv \mathbf{N}$ ) provided that, for all natural numbers  $k$ , the coefficients  $a_{1,k}$  and  $b_{1,k}$  are positive.

Naturally, the existence results depend on the hypothesis on the coagulation coefficients  $a_{j,k}$ . It is interesting to observe that not only the growth but also the structure

of the coefficients is crucial for the existence of solution, as it is clear in the following results. Let us start by considering the following coagulation coefficients, that will be called *multiplicative type coefficients*:

(H1) There exists non-negative sequences  $(r_j)$  and  $(\alpha_{j,k})$  such that

$$a_{j,k} = r_j r_k + \alpha_{j,k}, \quad (40)$$

and one of the two conditions is satisfied:

$$\lim_{j \rightarrow \infty} \frac{r_j}{j} = 0, \quad \lim_{j \rightarrow \infty} \frac{\alpha_{j,k}}{j} = 0, \quad \forall k \geq 1, \quad (41)$$

or

$$\inf_{j \geq 1} \frac{r_j}{j} = R > 0, \quad \alpha_{j,k} \leq K r_j r_k, \quad \forall j, k \geq 1, \quad (42)$$

for non-negative constants  $R$  and  $K$ .

For this kind of coefficients we have the following result due to Laurençot [123], and Leyvraz and Tschudi [147],

**Theorem 2** *Assuming (H1) and being  $c_0 \in X_1^+$ , there exists at least one solution  $c$  of (34) in  $[0, +\infty)$  such that, for all  $t \in [0, +\infty)$ , it holds true that  $c(t) \in X_1^+$  and  $\|c(t)\|_1 \leq \|c_0\|_1$ .*

*Sketch of proof:* The basic idea of the proof is, by taking limits as  $n \rightarrow \infty$  in the sequence of solutions of  $n$ -truncated systems, to obtain a non-negative continuous function and to prove that this function is, in fact, a solution to the Cauchy problem for Smoluchowski's equation (34).

Being a bit more precise, assume the coagulation coefficients satisfy (41). Then, the existence of a function  $c = (c_j)$  that is limit of the sequence of solution  $(c_j^N)$  of the truncated systems (for instance, using (35)) is a consequence of the Ascoli-Arzela theorem, due to the compact inclusion of  $X_1$  in  $X_{(r)} := \left\{ c = (c_j) \in \mathbf{R}^{N^+} : \|c\|_{(r)} < \infty \right\}$ , where  $\|c\|_{(r)} := \sum_{j=1}^{\infty} r_j |c_j|$ , and the equiboundedness and uniform equicontinuity of the sequence of solutions to the truncated systems. Another way to prove the existence of a limit as  $N \rightarrow \infty$  of the sequence of truncated solutions  $(c_j^N)$  is due to Ball and Carr [12] and consists in the application of Helly's theorem [113, pp. 370-371] to an equibounded sequence of uniformly bounded variation functions built from the solutions  $(c_j^N)$ .

By (41), the *a priori* uniform boundedness of the density of  $(c_j^N)$  is sufficiently strong to conclude the following estimate (uniform in  $N_k$ )

$$\sum_{i=M}^{N_k} r_i c_i^{N_k} \leq \sup_{i \geq M} \frac{r_i}{i} \sum_{i=M}^{N_k} i c_i^{N_k} \leq \|c_0\|_1 \sup_{i \geq M} \frac{r_i}{i}, \quad (43)$$

which allows us to control the infinite sum in the right-hand side of (35) and to obtain the pointwise limit

$$\lim_{k \rightarrow \infty} \left| \sum_{j=1}^{N_k} a_{i,j} c_j^{N_k} - \sum_{j=1}^{\infty} a_{i,j} c_j \right| = 0. \quad (44)$$

By the dominated convergence theorem, implies that we can take limits as  $N \rightarrow \infty$  and prove that  $c$  is a solution of (34).

If the coagulation coefficients satisfy (42) instead of (41) the problem is harder and the argument has to be modified. The difficulty of this case arises from the fact that the uniform bound on the density of  $(c_j^N)$  is not strong enough to control the terms in the second sum of the right-hand side of the truncated system. A way to overcome this problem, due to Laurençot [123], uses truncation (37).

The first part of the proof consists in getting the existence of a function  $c$  that is limit of the solutions  $(c_j^N)$  of the truncated systems. This is a consequence of the compact injection of  $H^1(0, T)$  in  $C(0, T)$ . We start by pointing out that the version of (38) for the solutions  $(c_j^N)$  of (37) is

$$\sum_{j=1}^{2N} g_j c_j^N(t) - \sum_{j=1}^{2N} g_j c_j^N(\tau) = \frac{1}{2} \int_{\tau}^t \sum_{j=1}^N \sum_{k=1}^N (g_{j+k} - g_j - g_k) a_{j,k} c_j^N(s) c_k^N(s) ds, \quad (45)$$

and the needed estimates are concluded by exploiting this  $g$ -moment propagation equation. Choosing in (45)  $g_j = j \mathbf{1}_{\{1, \dots, N\}}$ ,  $g_j = 1$ , and  $g_j = j^{1/2} \mathbf{1}_{\{1, \dots, N\}}$ , one gets

$$\sum_{j=1}^N j c_j^N(t) \leq \sum_{j=1}^N j c_j^N(\tau) \leq \sum_{j=1}^N j c_{j0} \quad (46)$$

$$\sum_{j=1}^{2N} c_j^N(t) + \frac{1}{2} \int_0^t \left| \sum_{j=1}^N r_j c_j^N(s) \right|^2 ds \leq \sum_{j=1}^{2N} c_{j0} \quad (47)$$

$$\int_{\tau}^t \left| \sum_{j=M}^N r_j c_j^N(s) \right|^2 ds \leq 4 \left( \sum_{j=1}^N j^{1/2} c_j^N(\tau) \right) M^{-1/2}. \quad (48)$$

These *a priori* estimates imply that, for every  $T \in (0, +\infty)$  and  $N \geq j$ ,  $c_j^N(t) \leq \|c_0\|_0$  in  $[0, T]$  and

$$\left\| \frac{dc_j^N}{dt} \right\|_{L^2(0, T)} \leq T^{1/2} \left( \sum_{i=1}^{j-1} a_{i, j-i} \right) \|c_0\|_0^2 + \sqrt{2}(1+K)r_j \|c_0\|_0^{3/2}. \quad (49)$$

Hence,  $(c_j^N)$  is bounded in  $H^1(0, T)$  and thus is relatively compact in  $C(0, T)$ . Using a diagonalization argument, one can conclude the existence of a subsequence  $(c_j^{N_k})$  converging in  $C(0, T)$  to some function  $c = (c_j)$  as  $N_k \rightarrow \infty$ .

This convergence, together with the estimates (46) and (48) imply the version of (44) for the present case,

$$\lim_{k \rightarrow \infty} \left\| \sum_{j=1}^{N_k} a_{i,j} c_j^{N_k} - \sum_{j=1}^{\infty} a_{i,j} c_j \right\|_{L^2(0,T)} = 0, \quad (50)$$

which is the main ingredient to pass to the limit in the  $i^{\text{th}}$  equation of (37), thus concluding the proof that  $c$  is a solution of (34). ■

Let us now consider *additive* coagulation coefficients, i.e., those satisfying the condition

(H2) There exist non-negative sequences  $(r_j)$  and  $(\alpha_{j,k})$  such that

$$a_{j,k} = r_j + r_k + \alpha_{j,k}, \quad (51)$$

and also  $0 \leq \alpha_{j,k} \leq K(j+k)$ , for some constant  $K \geq 0$ .

If the sequences  $(r_j)$  and  $\alpha_{j,k}$  are sublinear and satisfy (41) the argument of Leyvraz and Tschudy presented above can be adapted to obtain an existence proof also in this case. When  $r_j \leq (\text{const.})j$  it is necessary to modify those arguments: it is still possible to use Helly's theorem to prove the convergence of a subsequence of the sequence of solutions to truncated systems but the remaining proof needs to be changed using an identity like (38) for the evolution of the partial sums  $\sum_{j=m}^N g_j c_j^N(t)$ . This approach, due to Ball and Carr [12], is also applicable to the coagulation-fragmentation system, and so we leave a more detailed presentation to the next section. The result that is proved is the following:

**Theorem 3** *Let  $K > 0$  be a constant and assume  $a_{j,k} \leq K(j+k)$ . Let  $c_0 \in X_1^+$ . Then, there exists at least one solution  $c$  of (34) in  $[0, +\infty)$  such that  $c(t) \in X_1^+$  and  $\|c(t)\|_1 \leq \|c_0\|_1$ , for all  $t \in [0, +\infty)$ .*

An important distinction between systems with multiplicative and additive coefficients is that, in the last case, there are no solutions to the Cauchy problems (34) when the coefficients grow superlinearly, which certainly contrasts with what happen in the multiplicative case, as seen in Theorem 2.

This somewhat surprising non-existence result is a consequence of the following two theorems, to which we shall return in Section 4 on density conservation:

**Theorem 4** [12] *Assume (H2) and let  $c_0 \in X_1^+$ . Then, for every  $T > 0$ , all solution  $c$  of (34) in  $[0, T)$  conserve density,  $\|c(t)\|_1 = \|c_0\|_1$ .*

**Theorem 5** [35, 69] *Let  $T \in (0, +\infty]$ , and let  $C_L, C_U > 0$  and  $\beta \geq \alpha > 1$  be constants such that  $C_L(j^\alpha + k^\alpha) \leq a_{j,k} \leq C_U(jk)^\beta$ . Let  $c_0 \neq 0$  be an arbitrary element of  $X_1^+$ . Then, there are no solutions  $c$  of (34) in  $[0, T)$  that conserve density in  $[0, \tau)$ ,  $\forall \tau \leq T$ .*

Clearly, one can multiply the number of existence results indefinitely by making assumptions about the coefficients different from (H1) or (H2), but for these studies to be of any relevance it is necessary that the assumptions are either inordinately general, or of special interest for the applications. The case of the Becker-Döring type coefficients falls into this last class (cf. Section 1.6):

$$a_{j,k} = 0 \quad \text{if } j \wedge k > N,$$

where  $N \geq 1$  is a fixed constant. The classic case corresponds to  $N = 1$  [182] and is the only important one for the applications (cf., e.g., [16]). This means that the only non-zero coefficients are  $a_{j,1} (= a_{1,j})$  and the coagulation system is sometimes called the “addition model” (cf. [124]). The Cauchy problem (34) for these models with  $c_0 \in X_1^+$  has density conserving solutions in every interval  $[0, T)$  when  $a_{j,1} \sim \mathcal{O}(j)$ , [13], and, by arguments similar to those used with assumption (H2), do not have any solution in whatever non degenerate time interval if  $a_{j,1}$  is superlinear (satisfying somewhat technical but not very restrictive conditions) [124]. We shall return to these addition systems later in the chapter.

### 2.3 Discrete coagulation-fragmentation equations

Let us now turn our attention to the problem of existence of solutions to the initial value problem for discrete coagulation-fragmentation equations (16), that we now write as follows:

$$\begin{aligned} \dot{c}_j &= \frac{1}{2} \sum_{k=1}^{j-1} W_{j-k,k}(c) - \sum_{k=1}^{\infty} W_{j,k}(c), \\ c_j(0) &= c_{j0}, \end{aligned} \tag{52}$$

where  $W_{j,k}(c) := a_{j,k}c_jc_k - b_{j,k}c_{j+k}$ .

The approximation of these systems by finite dimensional truncations works as in the previous section. The systems are analogous to those then considered, (35)-(37), the main difference being the substitution of  $a_{p,q}c_p c_q$  by  $W_{p,q}(c)$ . With this minor change, and with the additional condition  $\sum_{j=1}^{\infty} b_{j,k}c_j \in L^1(0, t)$ , we obtain a definition of solution for coagulation-fragmentation systems of the same type as Definition 1.

As pointed out in Section 1.5, the first mathematical work on these equations was due to Spouge [206], who considered sublinear coagulation coefficients  $a_{j,k} \leq r_j r_k$  with  $r_j \sim \mathcal{O}(j)$  as  $j \rightarrow \infty$ , and somewhat restrictive conditions on the fragmentation coefficients.

More recent results, valid for much more general coefficients were proved by Ball and Carr [12], da Costa [45], Laurençot [126], and others, and it is to these that we will now turn our attention. The fundamental technique of these works is, as in the coagulation system considered in the previous section, the exploitation of the evolution of appropriate  $g$ -moments of the solutions  $(c^N)$  of the  $N$ -truncated systems as a way to obtain *a priori* estimates useful for taking limits as  $N \rightarrow \infty$ .

The version of (38) for solutions  $(c_j^N)$  of the  $N$ -truncated maximal system that is useful in this study is

$$\sum_{j=m}^N g_j c_j^N(t) - \sum_{j=m}^N g_j c_j^N(\tau) = \int_{\tau}^t \left( \frac{1}{2} \sum_{T_{m,N}^1} (g_{j+k} - g_j - g_k) W_{j,k}(c) + \right.$$

$$+\frac{1}{2} \sum_{T_{m,N}^2} g_{j+k} W_{j,k}(c) + \sum_{T_{m,N}^3} (g_{j+k} - g_k) W_{j,k}(c) \Big) ds, \quad (53)$$

where  $T_{m,N}^p$  are the following subsets of  $\mathbf{N} \times \mathbf{N}$ :  $T_{m,N}^1 := \{j, k \geq m, j+k \leq N\}$ ,  $T_{m,N}^2 := \{j, k \leq m-1, m \geq j+k \leq N\}$ , and  $T_{m,N}^3 := \{j \leq m-1, k \geq m, j+k \leq N\}$ , with the sum defined to be zero if the corresponding set is empty.

Let us start with the case where the coagulation coefficients are of the type  $a_{j,k} \leq K(j+k)$ , for some positive constant  $K$ . This condition includes the case  $a_{j,k} \leq (\text{const.})(jk)^{1/2}$ , but not other important cases such as  $a_{j,k} \leq (\text{const.})jk$ , that will be considered afterwards.

**Theorem 6** [12, 126] *Let  $a_{j,k} \leq K(j+k)$ , where  $K$  is an arbitrary positive constant. Let  $c_0$  be any element of  $X_1^+$ . Then, there exists at least one solution  $c$  of (34) defined on  $[0, +\infty)$  and satisfying  $\|c(t)\|_1 = \|c_0\|_1$ .*

The original proof of this theorem is due to Ball and Carr [12, Theorems 2.4 and 2.5]. In what follows we present a simpler version by Laurençot [126] that is based on the adaptation and generalization of a classical result of de la Vallée Poussin [190, Theorem I.1.2-2] which, *grosso modo*, guarantees that every integrable function has an higher integrability property (cf. Lemma 1 below). It is this additional integrability that allows the deduction of an *a priori* estimate to pass to the limit  $N \rightarrow \infty$  in the sequence  $(c^N)$ .

A noteworthy aspect of these proofs is that no assumptions are made on the binary fragmentation coefficients (apart from the general ones of positivity and symmetry). The result of [126] is even applicable to equations with multiple fragmentation (10)-(11), but here we will particularize for the case of binary fragmentation (52).

*Sketch of proof:* To get a function  $c$  as limit of solutions  $(c^N)$  to the truncated systems we proceed as in the proof of Theorem 2, applying Helly's theorem to an auxiliary sequence [12]. The fundamental problem is to prove that the limit function is a solution to the Cauchy problem. It is on this problem that we will centre our attention.

Let  $\mathcal{K}_1$  be the subset of  $C^1([0, +\infty)) \cap W_{\text{loc}}^{2,\infty}(0, +\infty)$  whose elements are non-negative convex functions  $U$  such that  $U(0) = 0$ ,  $U'(0) \geq 0$ , and  $U'$  is concave. Let  $\mathcal{K}_{1,\infty} \subset \mathcal{K}_1$  be the set of those functions that, additionally, satisfy

$$\lim_{x \rightarrow +\infty} U'(x) = \lim_{x \rightarrow +\infty} \frac{U(x)}{x} = +\infty. \quad (54)$$

The following lemma is an extension of a result of de la Vallée Poussin that is useful in what follows:

**Lemma 1** [122, 140] *Let  $(\Omega, \mathcal{B}, \mu)$  be a measure space, and let  $w \in L^1(\Omega, \mathcal{B}, \mu)$ . Then, there exists a function  $U \in \mathcal{K}_{1,\infty}$  such that  $U(|w|) \in L^1(\Omega, \mathcal{B}, \mu)$ .*

In applying this lemma to our case  $\Omega = \mathbf{N}$ ,  $\mathcal{B} = 2^{\mathbf{N}}$ , and, for  $I \in \mathcal{B}$ , define  $\mu(I) := \sum_{i \in I} c_{0i}$ , where  $c_0 \in X^+$  is the initial condition of the Cauchy problem (34). Since  $c_0 \in X^+$  we have  $(x \mapsto x) \in L^1(\Omega, \mathcal{B}, \mu)$  and, by Lemma 1, there exists a function  $U_0 \in \mathcal{K}_{1,\infty}$  such that  $(x \mapsto U_0(x)) \in L^1(\Omega, \mathcal{B}, \mu)$ , and so

$$\sum_{i=1}^{\infty} U_0(i) c_{0i} < \infty. \quad (55)$$

Observe that, in the sense of (54),  $U_0$  grows faster at infinity than the identity and thus (55) provides a stronger decay of the initial condition  $c_0$ . As with the coagulation equations, the equation for the evolution of the  $U$ -moments of solutions  $(c^N)$  to the truncated systems is essential to obtain the needed estimates. For that we need to know that, for every  $U \in \mathcal{K}_1$ , there exists a positive constant  $m_U$  such that  $(i+j)(U(i+j) - U(i) - U(j)) \leq m_U(iU(j) + jU(i))$ , for all  $i, j \in \mathbf{N}$ . Using this inequality in (53) with  $m = 1$  e  $g = U_0$  we obtain, for every  $0 \leq t \leq T < +\infty$ ,

$$\sum_{j=1}^N U_0(j) c_j^N(t) \leq C(T), \quad (56)$$

$$0 \leq \int_0^T \sum_{i=1}^{N-1} i \sum_{j=i+1}^N \left( \frac{U_0(j)}{j} - \frac{U_0(i)}{i} \right) b_{i,j-i} c_j^N(s) ds \leq C(T), \quad (57)$$

where by  $C(T)$  we denote constants depending on  $T$ , and also of  $K$ ,  $c_0$  and  $U_0$ .

The same estimates are valid if in (56) we sum only up to  $i \leq M$  and in (57) only up to  $i \leq M-1$  and  $j \leq M$ , with  $M \leq N-1$ . Taking limits, first  $N \rightarrow +\infty$ , and then  $M \rightarrow \infty$ , we conclude that

$$\sum_{j=1}^{\infty} U_0(j) c_j(t) \leq C(T), \quad (58)$$

$$0 \leq \int_0^T \sum_{i=1}^{\infty} i \sum_{j=i+1}^{\infty} \left( \frac{U_0(j)}{j} - \frac{U_0(i)}{i} \right) b_{i,j-i} c_j(s) ds \leq C(T), \quad (59)$$

and from this it follows that  $\sum_{j=1}^{\infty} a_{i,j} c_j \in L^1(0, T)$ , and  $\sum_{j=i+1}^{\infty} b_{i,j-i} c_j \in L^1(0, T)$ . From (56) we deduce the following estimate, similar to (43),

$$\sum_{j=M}^{N-i} a_{i,j} c_j^N \leq 2iK \sup_{j \geq M} \frac{j}{U_0(j)} \sum_{j=M}^{N-i} U_0(j) c_i^N \leq C(i, T) \sup_{j \geq M} \frac{j}{U_0(j)}, \quad (60)$$

which, together with the analogous one obtained from (58) and with Lebesgue dominated convergence theorem, allow us to control the tails of the series corresponding to the coagulation terms and get

$$\lim_{N \rightarrow \infty} \left\| \sum_{j=1}^{N-i} a_{i,j} c_i^N c_j^N - \sum_{j=1}^{\infty} a_{i,j} c_i c_j \right\|_{L^1(0, T)} = 0. \quad (61)$$



The corresponding limit for the fragmentation terms, namely

$$\lim_{N \rightarrow \infty} \left\| \sum_{j=i+1}^N b_{i,j-i} c_j^N - \sum_{j=i+1}^{\infty} b_{i,j-i} c_j \right\|_{L^1(0,T)} = 0, \quad (62)$$

results from (57), (59) and the dominated convergence theorem.

This concludes the proof that the function  $c$  obtained as the limit of the truncations  $(c^N)$  when  $N \rightarrow \infty$ , is a solution of (52). To prove that the norm of  $c$  is equal to the norm of the initial condition we again use (56) and (58) in order to write, with  $N \geq M-1 \geq 2$  arbitrary,

$$\begin{aligned} \left| \|c(t)\|_1 - \|c_0\|_1 \right| &\leq \sum_{j=1}^{M-1} j |c_j^N(t) - c_j(t)| + \sum_{j=N+1}^{\infty} j c_{0j} + \sum_{j=M}^N j c_j^N(t) + \sum_{j=M}^{\infty} j c_j(t) \\ &\leq \sum_{j=1}^{M-1} j |c_j^N(t) - c_j(t)| + \sum_{j=N+1}^{\infty} j c_{0j} + 2C(T) \sup_{j \geq M} \frac{j}{U_0(j)}, \end{aligned} \quad (63)$$

which, by the arbitrariness of  $M$  and  $N$ , implies that  $\|c(t)\|_1 = \|c_0\|_1$ .  $\blacksquare$

If the coagulation coefficients do not satisfy the bound  $a_{j,k} \leq K(j+k)$ , but only the weaker condition  $a_{j,k} \leq K(jk)^\alpha$ , with  $\alpha \in [0, 1]$ , there are also several existence theorems for which it is also necessary to impose, in addition to the growth condition on the coefficients, some conditions on their structure, as well as restrictions upon the fragmentation coefficients. As we pointed out in page 30 a first result of this type, by Spouge [206], is the following (written with the hypothesis of binary fragmentation).

**Theorem 7** [206] *Let  $a_{j,k} \leq \sigma(j)\sigma(k)$ , as  $j, k \rightarrow +\infty$  where  $K$  is an arbitrary positive constant, and let  $b_{j,k}$  satisfy  $\sum_{k=1}^{j-1} b_{j,k} \leq Q$  and  $b_{j,k} \leq \sigma(k)$  when  $k \rightarrow +\infty$ , for  $j$  fixed, where  $Q > 0$  is a constant. Let  $c_0 \neq 0$  be an arbitrary sequence in  $X_1^+$ . Then, there exists at least one solution  $c$  of (52) defined in  $[0, +\infty)$ .*

The proof of this result, like the one of Theorem 2, uses Helly's and Ascoli-Arzelà theorems in order to obtain a solution of the Cauchy problem (52) by taking the limit  $N \rightarrow \infty$  in the sequence of solutions to truncated systems, [206].

Another existence result, obtained in [45] with a so called *strong fragmentation condition* on the fragmentation coefficients, is the following:

**Theorem 8** [45] *Let  $a_{j,k} \leq K_a(jk)^\alpha$ , with constants  $K_a > 0$  and  $\alpha \leq 1$ . Let  $b_{j,k}$  be such that  $\sum_{j=1}^{\lfloor \frac{r-1}{2} \rfloor} j^\mu b_{j,r-j} \geq K_f(\mu) r^{\gamma+\mu}$ , where  $\mu, \gamma$  and  $K_f(\mu)$  are non-negative constants, and  $\gamma > \alpha$ . Take any element  $c_0 \in X_1^+$ . Then, there exists at least one solution  $c$  of (52) defined on  $[0, +\infty)$ . The solutions of (52) obtained as limits of solutions of maximally truncated systems are unique and satisfy  $\|c(t)\|_1 = \|c_0\|_1, \forall t \geq 0$ .*

Observe that the strong fragmentation condition used in this theorem is satisfied by fragmentation coefficients of the type

$$b_{j,k} \sim (j+k)^\beta \text{ or } b_{j,k} \sim (jk)^\beta, \text{ with } \beta > -1. \quad (64)$$

*Sketch of proof:* The basic ingredient of the proof is the regularizing effect the strong fragmentation condition has on some higher moments, a fact that allows us to obtain the needed *a priori* estimates. This regularization result consists in the local integrability of moments  $\|c(\cdot)\|_{1+\gamma-\varepsilon}$ ,  $\forall \varepsilon$ , of functions  $c$  that are obtained as weak-\* limits of sequences of solutions to truncated systems. This idea had already been used by Carr [34] in the study of the asymptotic behaviour of solutions when the coagulation coefficients satisfy the conditions of Theorem 6. The main difference between the tools used in [34] and in [45] is that the differential inequality for the evolution of higher moments of solutions  $c^N$  to truncated systems is now

$$\frac{d}{dt} \|c^N\|_\mu \leq \mathcal{C}_0 + \mathcal{C}_1 \|c^N\|_\mu^{\alpha_1} - \mathcal{C}_2 \|c^N\|_\mu^{\alpha_2}, \quad (65)$$

where  $\mu \geq 1 + \alpha$ ,  $\alpha_1 = 1 + \frac{2\alpha-1}{\mu-1}$ ,  $\alpha_2 = 1 + \frac{\gamma}{\mu-1}$  and  $\mathcal{C}_j$  positive constants dependent only on  $\alpha, \gamma, \mu$  and  $\|c_0\|$ . In [34], the inequality akin to (65) has the right-hand side of the Bernoulli equation and thus can be explicitly solved by a standard change of variable. In the case of (65) the analysis is less direct but one can prove that  $\|c^N\|_\mu$  satisfies the inequality

$$\|c^N\|_\mu \leq [(v-1)At]^{-\frac{1}{v-1}}, \quad (66)$$

for every constant  $v \in (1, \alpha_2)$ , and for constants  $A = A(v, \alpha_1, \alpha_2, \mathcal{C}_0, \mathcal{C}_1, \mathcal{C}_2) > 0$  appropriately chosen. By taking the limit  $N \rightarrow \infty$  in (66) we obtain the local integrability of the  $\mu$ -moments ( $\mu < 1 + \gamma$ ) of functions obtained as weak-\* limits of solutions  $c^N$ . It is this local integrability of  $(1 + \gamma - \varepsilon)$ -moments that is the *a priori* estimate which, together with the dominated convergence theorem and Fatou's lemma, allows taking limits in the truncated equations, both in the coagulation and in the fragmentation terms. ■

To finish this section it is worth observing that the results hereby presented do not cover all possible conditions on the coefficients or definitions of solution. In particular, note that if the fragmentation coefficients decay more rapidly than what is determined by the estimates (64) the above results are not applicable to the case  $a_{j,k} \sim \mathcal{O}(jk)$  (Theorem 7, due to Spouge, requires  $a_{j,k} \sim \sigma(j)\sigma(k)$ ). The existence of solution in a case close to this critical growth case, where the coefficients have the structure  $a_{j,k} = j^\alpha k + k^\alpha j$ , with  $\alpha \in (0, 1)$ , was obtained in [81] for continuous coagulation-fragmentation systems, as a consequence of estimates proved for the study of the gelation problem. The analysis presented in that paper can also be applied to discrete equations and will be analysed later in Section 4.

## 2.4 On the uniqueness of solutions

As in the case of existence studies, the results about uniqueness have been obtained under several different assumptions about the rate coefficients. The approach used

by these studies consists, essentially, in assuming the existence of two distinct solutions  $c$  and  $d$  to the Cauchy problem (34) or (52), and then proving that some moment of the function  $|x| := |c - d|$  satisfies a differential inequality which implies  $x \equiv 0$  (cf. e.g. [12, 13, 45, 126]).

In order to get the needed differential inequalities one needs to control the evolution of certain moments, which requires the imposition of restrictions, either on the class of solutions under consideration, or on the coefficients, that are usually more stringent than those required in order to prove existence. As an illustration we present the case, studied in [12], where all coagulation coefficients are bounded, which already contains the main ingredients used in more general cases:

**Theorem 9** [12] *Let  $a_{j,k} \leq K$  where  $K > 0$  is a constant. Take as initial condition any  $c_0 \in X_1^+$ . Then, there exists one and only one solution  $c$  to (52) defined in  $[0, +\infty)$  and satisfying  $\|c(t)\|_1 = \|c_0\|_1$  for all  $t \geq 0$ .*

*Sketch of proof:* Assuming there are two solutions of the initial value problem (52),  $c$  and  $d$ , define  $x := c - d$  and consider the function  $\psi_1(t) := \|x(t)\|_1$ . The version of (38) with  $g_j = j\mathbf{1}(j \leq n)$  and  $c$  substituted by  $|x|$  gives

$$\sum_{j=1}^n j|x_j(t)| = \int_0^t \left( U_n(s) + V_n(s) \right) ds, \quad (67)$$

where

$$U_n := \frac{1}{2} \sum_{T_{1,n}^1} (f_{j+k} - f_j - f_k)(W_{j,k}(c) - W_{j,k}(d)), \quad V_n := - \sum_{T_{1,n}^4} f_j(W_{j,k}(c) - W_{j,k}(d)),$$

with  $f_j := j \operatorname{sgn}(x_j)$ ,  $T_{1,n}^4 := \{1 \leq j \leq n, j+k \geq n+1\}$  and  $T_{1,n}^1$  was previously defined in page 31. Noting that  $W_{j,k}(c) - W_{j,k}(d) = (c_j x_k + d_k x_j) a_{j,k} - b_{j,k} x_{j+k}$ , and using  $a_{j,k} \leq K$ , we get  $\sum_{T_{1,n}^1} (f_{j+k} - f_j - f_k)(c_j x_k + d_k x_j) a_{j,k} \leq \operatorname{const.} \psi$ , and thus, because  $-(f_{j+k} - f_j - f_k)x_{j+k} \leq -((j+k) - j - k)|x_{j+k}| = 0$ , we have  $\int_0^t U_n(s) ds \leq (\operatorname{const.}) \psi(t)$ . The limit  $\int_0^t V_n(s) ds \rightarrow 0$  as  $n \rightarrow \infty$  is obtained using the condition on the coefficients and the hypothesis that  $c$  and  $d$  are density conserving, which is natural since these last conditions are equivalent to

$$\sum_{j=1}^n j c_j(t) - \sum_{j=1}^n j c_{j0} = - \int_0^t \sum_{T_{1,n}^4} j W_{j,k}(c(s)) ds \xrightarrow{n \rightarrow \infty} 0, \quad (68)$$

and similarly for  $d$ .

With these estimates we can write

$$\psi_1(t) \leq (\operatorname{const.}) \int_0^t \psi_1(s) ds, \quad (69)$$

and hence, by Gronwall's inequality,  $\psi_1 \equiv 0$ , implying uniqueness of density conserving solutions.  $\blacksquare$

Observe that Theorem 9 imposes a very strong boundedness condition on the coagulation coefficients but none on the fragmentation ones (apart from the basic ones of non-negativity and symmetry used to prove existence). Observe also that the theorem establishes uniqueness just in the family of density conserving solutions.

This type of restrictions occur also in other cases. In [45] it is proved that, under the hypothesis of Theorem 8, density conserving solutions are unique. The proof uses the same ideas as presented before but the estimates for  $U_n$  and  $V_n$  are now obtained using the following integrability result  $\|c(\cdot)\|_{1+\alpha} \in L^1(0, t)$ ,  $\forall t < \infty$ , of solutions  $c$  to (52). This additional regularity, similar to what was used in the proof of Theorem 8, has to be established for all solutions of (52), not only for those obtained by taking limits of solutions to truncated problems, and this is achieved by a kind of step-by-step argument first used by Carr in [34]. The estimates finally result in the following inequality, similar to (69),

$$\psi_1(t) \leq \text{const.} \int_0^t \varphi(s) \psi_1(s) ds, \quad (70)$$

where  $\varphi(s) = K_a \|c(s)\|_{1+\alpha} + K_a \|d(s)\|_{1+\alpha}$ .

Another uniqueness result, similar to the one in Theorem 9, is proved in [126] and complements the existence result whose proof was presented in Theorem 6. Under the condition  $a_{j,k} \leq A_j + A_k$ , where  $A_i \leq K_a i$ , it is shown, by a proof like the one above, that uniqueness holds in the class of density conserving solutions satisfying the integrability condition  $\sum_{j=1}^{\infty} j A_j c_j \in L^1(0, t)$ , for each  $t < \infty$ . There is a natural

problem that immediately comes to mind, which is the existence of solutions with this additional regularity, or, better still, to know what are the additional conditions (if any) that need to be imposed at  $t = 0$  that ensure this extra regularity at later times. The answer to this problem was given by Laurençot in [126] and generalizes previous similar results by Carr and da Costa [36]. Before presenting the result we need to introduce the following notation: we say that a function  $U$  is an element of  $\mathcal{K}_2$  if it is non-negative, convex, belongs to  $C^2([0, +\infty))$ , satisfies  $U(0) = U'(0) = 0$ , its derivative is a convex function and there exists a positive constant  $K_U$  such that  $U'(2x) \leq K_U U'(x)$  for all  $x \geq 0$ . (The functions  $x \mapsto x^m$  are in  $\mathcal{K}_2$  if  $m \geq 2$ .)

**Proposition 1** [126] *Let  $a_{j,k} \leq K(j+k)$ , where  $K > 0$  is a constant. Let  $c_0 \in X_1^+$  be such that there exists  $U \in \mathcal{K}_1 \cup \mathcal{K}_2$  with  $\sum_{j=1}^{\infty} U(j) c_{j0} < \infty$ . Then, there exists at least one solution  $c$  of (34) defined in  $[0, +\infty)$ , satisfying  $\|c(t)\|_1 = \|c_0\|_1$  and, for each  $t < \infty$ ,*

$$\sup_{s \in [0, t]} \sum_{j=1}^{\infty} U(j) c_j(s) < \infty.$$

This type of results, usually called “propagation of moments”, are very useful for the study of the long time behaviour of solutions and we shall return to them in the next section.

Imposing growth restrictions on the kinetic coefficients it is possible to prove uniqueness without further regularity restrictions on the initial data. An example, due to Ball and Carr [12], is the following:

**Theorem 10** [12] *Let  $K > 0$  and  $\alpha \in [0, \frac{1}{2}]$  be constants such that, for all natural numbers  $j, k, n_0$ , it holds  $a_{j,k} \leq K(jk)^\alpha$ ,  $\sum_{j=1}^{\lfloor (k+1)/2 \rfloor} j^{1-\alpha} b_{k-j,j} \leq Kk^{1-\alpha}$  and  $\sum_{j=n_0}^{\lfloor (r+1)/2 \rfloor} j^1 b_{r-j,j} \leq Kr$ , for  $r \geq 2n_0$ . Let  $c_0 \in X_1^+$  be arbitrary. Then, there exists only one solution  $c$  of (52) defined on  $[0, T)$ .*

Note that, due to  $(jk)^\alpha \leq (jk)^{1/2} \leq \frac{1}{2}(j+k)$ , Theorem 6 can be applied to this case and this means that, under these conditions, solutions to (52) are unique and conserve density. However, note that the present result is not a uniqueness theorem in the class of density conserving solutions (as in the result of [46] cited above) but in the universe of all solutions to the Cauchy problem (52) in the sense of Definition 1. The proof of this theorem uses the method presented above for Theorem 9. The only relevant difference is that now is more convenient to get estimates on  $\psi_{1-\alpha}(t) := \|x(t)\|_{1-\alpha}$  instead of  $\psi_1(t) = \|x(t)\|_1$ . The final result, from which uniqueness easily follows, is the inequality (69) with  $\psi_1$  substituted by  $\psi_{1-\alpha}$ .

A natural question at this point is to know to what extent the cases not covered by these uniqueness theorems correspond to real cases of non-uniqueness. Clearly, the complete elucidation of this problem means a complete characterization of uniqueness, something not yet achieved at present. However, there are examples of non-uniqueness that seem to provide evidence that this problem is not simple. To conclude we present one of these examples, due to Ball and Carr, [12]. Note that it is an example about the linear fragmentation system.

**Example 1** [12] *Let  $a_{j,k} = 0$  and  $b_{j,k} = 1$ . Then,  $c_j(t) = e^{-(j-1)t/2} (1 - e^{-t/2})^2$  is a solution of (52) with initial condition  $c_0 \equiv 0$ .*

Observe that the assumptions in Example 1 are included in those considered in Theorem 9. Hence, we know that density conserving solutions are unique. As a solution of (52) with  $c_0 \equiv 0$  is the identically zero solution, the above example implies that we have non-uniqueness. Due to the linearity of the system, we can obtain analogous non-uniqueness results for other initial conditions. For further discussion on these issues see [7].

### 3 Long-time behaviour of solutions to coagulation-fragmentation systems

We shall now review some of the most important mathematical aspects of the long time behaviour of solutions. The large number of results in the literature forces us to make some choices about the results we will cover. We will keep the approach at the level used in the previous section so as to give the reader not only a guide to the literature but also to the ideas and some details of the proofs of the existing results.

### 3.1 Convergence to equilibria and phase transitions

In this section we consider results about the convergence to equilibria of solutions to the discrete coagulation-fragmentation systems.

Since, as was pointed out in the Introduction, these equations can be seen as a mathematical model of an isolated chemical system, it is natural to expect solutions to converge to some equilibrium as  $t \rightarrow +\infty$ . In fact, in the usual chemical kinetics models this is exactly what normally happens. We shall see that in the infinite dimensional coagulation-fragmentation systems the asymptotic behaviour is much more interesting, even surprising, and a behaviour that is physically interpreted as a dynamical phase transition can take place under appropriate conditions.

#### 3.1.1 Strong fragmentation systems

Let us start with the *strong fragmentation* case. This case was studied by Carr [34] and by Fournier and Mischler [93] and these conditions on the fragmentation coefficients were also considered by da Costa in [45] for the study of existence of solutions discussed above (cf. Theorem 8). The technique used in [34, 93] is based on the fact, pointed out in the discussion of the proof of Theorem 8, that this assumption on the fragmentation coefficients imply the boundedness of some higher moments, which implies the solution is pre-compact in  $X_1^+$  for the norm topology. The existence of a Lyapunov function and the application of LaSalle's invariance principle were the tools that allowed Carr to prove the convergence of the solution to a unique equilibrium. Estimates based on the regularity of higher order moments were essential to obtain the exponential convergence to equilibria by Fournier and Mischler, valid for sufficiently small initial data. The result of [34] is the following:

**Theorem 11** [34] *Let  $K, K_f > 0$ ,  $\alpha \in [0, 1]$  and  $\gamma > \alpha$  be constants such that, for all natural numbers  $j, k$ , the following holds  $a_{j,k} \leq K(j^\alpha + k^\alpha)$ ,  $a_{1,k}, b_{1,k} > 0$  and  $\sum_{j=1}^{\lfloor \frac{r-1}{2} \rfloor} j^\mu b_{j,r-j} \geq K_f(\mu) r^{\gamma+\mu}$ . Assume that the detailed balance condition (15) is satisfied and that, for some  $q \geq 1$ , the partition function  $(M_j)$  satisfies  $\liminf_{j \rightarrow \infty} M_j^{1/j^q} > 0$ . Let  $\rho \geq 0$  be arbitrary. Then, there exists a time-independent solution  $c^\rho$  of the coagulation-fragmentation equations, with density  $\rho$ , such that, for all initial data  $c_0 \in X_1^+$  with density  $\|c_0\| = \rho$ , the unique solution  $c(\cdot)$  of (52) with constant density satisfies  $\|c(t) - c^\rho\|_m \xrightarrow{t \rightarrow \infty} 0$ , for all  $m \geq 1$ .*

*Sketch of proof:* With these assumptions on the coefficients the moments of solutions  $c^N$  to the truncated systems satisfy the following differential inequality, analogous to (65),

$$\frac{d}{dt} \|c^N\|_\mu \leq \mathcal{C}_0 \|c^N\|_\mu - \mathcal{C}_1 \|c^N\|_\mu^{\alpha_2}, \quad (71)$$

where  $\mu > 1$ ,  $\alpha_2 = 1 + \frac{\gamma}{m-1}$ ,  $\mathcal{C}_0$ , and  $\mathcal{C}_1$  are positive constants. The standard change of variables used to solve the Bernoulli ordinary differential equations,  $\|c^N\|_\mu \mapsto$

$u := \|c^N\|_\mu^{1-\alpha_2}$ , can be used to solve explicitly this differential inequality and taking  $N \rightarrow \infty$  we conclude the solutions of (52) that are obtained as limits of truncated system satisfy

$$\|c\|_\mu \leq A (1 - e^{-Bt})^{-\frac{\mu-1}{\gamma}}, \quad (72)$$

where  $\mu > 1$ ,  $A$  and  $B$  are positive constants.

The strong fragmentation condition also implies that the  $(1 + \gamma - \varepsilon)$ -moments of every density conserving solution to (52) are integrable, as stated above (cf. page 36), and this implies the uniqueness of density conserving solutions. This fact means that there exists a semi-group of operators  $T(\cdot)$  defined by  $T(\cdot)c_0 := c$ , where  $c$  is the unique density conserving solutions of (52). The inequality (72) implies that, for each  $\mu > 1$  and  $\tau > 0$ ,  $\cup_{t \geq \tau} T(t)c_0$  is a bounded set of  $X_\mu$ , and so, by the compact inclusion among the spaces  $X_\alpha$  (cf. page 24) it is a pre-compact subset of  $X_1^+$ . Hence, for each initial condition  $c_0 \in X_1^+$ , the solution  $T(t)c_0$  has a non-empty invariant  $\omega$ -limit set  $\omega(c_0) \subset X_\mu$ , for all  $\mu \geq 1$ . What remains to be proved is that  $\#\omega(c_0) = 1$  and its single element is an equilibrium, i.e., a time independent solution with density  $\rho = \|c_0\|$ , and that this equilibrium is independent of the initial condition, provided its density is  $\rho$ . It is at this point that the detailed balance condition is used, and the existence of a Lyapunov functions plays a central role.

For every  $d_1 \geq 0$  define the sequence  $d = (d_j)$  by

$$d_j := M_j(d_1)^j. \quad (73)$$

Clearly, the detailed balance condition implies that

$$W_{j,k}(d) = a_{j,k}d_jd_k - b_{j,k}d_{j+k} = a_{j,k}M_jM_k(d_1)^{j+k} - b_{j,k}M_{j+k}(d_1)^{j+k} = 0,$$

and so  $d$  is a stationary solution of (52) with  $c_0 = d$  if and only if  $d \in X_1^+$ . The positivity is obvious from (73), but the fact that  $d$  has finite density requires a little more care. In order to study the density of  $d = (d_j) = (M_j(d_1)^j)$  it is natural to consider the function  $z \mapsto F(z) : [0, +\infty) \rightarrow [0, +\infty]$  defined by

$$F(z) := \sum_{j=1}^{\infty} j M_j z^j. \quad (74)$$

Let  $z_s \in [0, +\infty]$  be the convergence radius of this series, and let  $\rho_s := \sup_{z \in [0, z_s)} F(z)$ . Clearly, there are three distinct cases for  $z_s$ : if  $z_0 = 0$  then  $\rho_s = 0$  and the only equilibrium solution is the zero solution; if  $z_s = +\infty$  then  $\rho_s = +\infty$  and for each  $\rho \geq 0$  there exists a unique equilibrium (73) with density  $\|d\|_1 = F(d_1)$ ; finally, if  $z_s \in (0, +\infty)$ , it can happen either  $\rho_s = +\infty$  or  $\rho_s < +\infty$ , and in this last case there are no equilibria with densities  $\rho > \rho_s$ . This case will be important in the next section but under the strong fragmentation conditions that we are currently considering it is possible to prove that  $\rho_s = +\infty$ , and so, for every  $\rho > 0$  there exists an equilibrium given by (73) with density  $\rho$ , [34]. We will denote this equilibrium by  $c^\rho$ .

The discovery of Lyapunov functions for coagulation-fragmentation systems, related with the free energy, or with the entropy, of the physical system, was first made

by Aizenman and Bak [1] for continuous systems with constant rate coefficients. For more general systems, the existence of a Lyapunov function seems to have been first identified, at a formal level, by Buhagiar in the Becker-Döring system (cf. ref. cit. [13]) and was first used in a mathematically rigorous way by Ball, Carr and Penrose in [13]. Our presentation will follow this paper closely, although the boundedness of higher moments due to our present strong fragmentation condition greatly simplifies it.

Let  $c \in X_1^+$  and consider the function

$$V(c) := \sum_{j=1}^{\infty} c_j \left( \log \frac{c_j}{M_j} - 1 \right), \quad (75)$$

where the term in the sum is defined to be zero if the corresponding  $c_j$  is zero. The continuity and minimization properties of this functional were established in [13] and will be presented next: let  $V(c) = G(c) - F_m(c)$ , with

$$G(c) = \sum_{j=1}^{\infty} c_j (\log c_j - 1), \quad F_m(c) = \sum_{j=1}^{\infty} j^m c_j \log M_j^{1/j^m}. \quad (76)$$

It is not hard to prove that  $G$  is finite and sequentially weak-\* continuous in  $X_1^+$ . As the radius of convergence of the series (74) is positive,  $\limsup_{j \rightarrow \infty} M_j^{1/j} < \infty$ , and we conclude that  $V$  is bounded below in

$$X_{1,\rho}^+ := \{c \in X_1^+ : \|c\|_1 = \rho\}, \quad (77)$$

$c^\rho$  is the only minimizer of  $V$  in  $X_{1,\rho}^+$ , and every minimizing sequence  $(c^{(j)})$  of  $V$  in  $X_{1,\rho}^+$  converges to  $c^\rho$  strongly in  $X_1$ . If  $\liminf_{j \rightarrow \infty} M_j^{1/j^q} > 0$ , for some  $q \geq 1$ , then  $V$  is bounded above in  $X_m \cap X_{1,\rho}^+$  and is continuous in this set if  $m \geq q$ .

For solutions  $(c^n)$  of the Cauchy problem for the maximally truncated coagulation-fragmentation system, the following holds:

$$V(c^n(t)) + \int_{\tau}^t D_n(c^n(s)) ds = V(c^n(\tau)) \quad (78)$$

where

$$D_N(c^n) := \frac{1}{2} \sum_{j+k \leq N} H_{j,k}(c^n) \quad (79)$$

and

$$\begin{aligned} H_{j,k}(c) &:= (a_{j,k} c_j c_k - b_{j,k} c_{j+k}) (\log(M_{j+k} c_j c_k) - \log(M_j M_k c_{j+k})) \\ &= (a_{j,k} c_j c_k - b_{j,k} c_{j+k}) (\log(a_{j,k} c_j c_k) - \log(b_{j,k} c_{j+k})) \geq 0, \end{aligned} \quad (80)$$



where the second equality comes from the detailed balance condition (15), and the positivity from the fact that solutions have all their components positive (cf. page 26) and  $\forall x, y > 0, (x - y)(\log x - \log y) \geq 0$ .

Since for every  $m \geq 1$  we have  $c^n \rightarrow c$  strongly in  $X_m^+$ , by the continuity of  $V$  we conclude that  $V(c^n(t)) \rightarrow V(c(t))$ , for every  $t \geq \tau > 0$ . Fixing a positive integer  $N$  we have  $D_n(c^n) \geq D_N(c^n)$  for  $n \geq N$ , and thus,

$$\liminf_{n \rightarrow \infty} \int_{\tau}^t D_n(c^n(s)) ds \geq \int_{\tau}^t D_N(c(s)) ds$$

which, letting  $N \rightarrow \infty$ , gives

$$V(c(t)) + \int_{\tau}^t D(c(s)) ds \leq V(c(\tau)), \quad (81)$$

with

$$D(c) := \frac{1}{2} \sum_{j,k \geq 1} H_{j,k}(c). \quad (82)$$

This concludes the proof that  $V$  is a Lyapunov function for (52).

With these ingredients is now easy to prove the existence of one, and only one, equilibrium  $c^\rho$  with density  $\rho$  (which has necessarily the form (73)), and to get the characterization of  $\omega(c_0)$ : since this set must consist of solutions along which the Lyapunov function is constant, this implies, by (81) and density conservation, that  $\omega(c_0) = \{c^\rho\}$ , with  $\rho = \|c_0\|$ , as we wanted to prove. ■

As the main ingredient of the above proof is, as already pointed out, the finiteness of higher order moments, Theorem 11 can be adapted, without further difficulties, to the conditions considered in [45], with the condition on the coagulation coefficients changed to  $a_{j,k} \leq K(jk)^\alpha$ , with  $\alpha \leq 1$ , [50].

To finish this section it is interesting to observe that the detailed balance condition is *not* a necessary condition to get convergence to equilibria. In fact, in [93], Fournier and Mischler proved that, if  $a_{j,k} \leq K(jk)^\alpha$  and  $L(j+k)^\gamma \leq b_{j,k} \leq K_f(j+k)^s$ , with  $\alpha \in [0, 1]$ ,  $\gamma > -2(1 - \alpha)$  and  $s, \gamma \in (-1, \infty)$ , then, for all  $\rho = \|c_0\|$  sufficiently small<sup>7</sup>, the solution  $T(t)c_0$  satisfies

$$\|T(t)c_0 - \hat{c}\|_2 \leq K e^{-\kappa t}, \quad \forall t \geq 1, \quad (83)$$

where the constants  $K, \kappa > 0$  depend only on  $\alpha, \gamma, K_c, L$ , and  $\rho$ , and  $\hat{c}$  is the only equilibrium of the system with density  $\rho$ . Note that this result establishes an exponential rate of convergence to equilibria.

The proof of this result is also based on the finiteness of higher order moments, which comes from the lower bound on the fragmentation coefficients. More specif-

<sup>7</sup> The precise technical condition used in [94], possibly not necessary, is that  $\rho$  satisfies the inequality

$$128 \frac{K_c \rho}{L} + 2 \left( \frac{32 K_c \rho}{L} \right)^{2 + \frac{1+2\alpha}{\gamma+2(1-\alpha)}} < 1.$$

ically, under the stated conditions one proves the following contraction property: there exists a  $T^*$  such that, for all  $t \geq T^*$ , and all solutions  $c$  and  $d$  of the coagulation-fragmentation system with initial data  $c_0$  and  $d_0$ , respectively, both with density  $\rho$ , the following inequality holds

$$\frac{d}{dt} \|c(t) - d(t)\|_2 \leq -\kappa \|c(t) - d(t)\|_2. \quad (84)$$

This differential inequality is obtained from

$$\frac{d}{dt} \|c - d\|_2 \leq \left( 2K \|c + d\|_3 - \frac{L}{16} \right) \|c - d\|_2,$$

and from an estimate on the third moment of  $c + d$  proving that one can estimate its value uniformly in time by a quantity smaller than the absolute value of the negative term, provided the density  $\rho$  of the initial condition (and hence of the solutions at later times, since they conserve density) is sufficiently small. It is likely that this restriction on the initial density can be improved, but so far this nice result is the best one available on rates of convergence with strong fragmentation conditions.

### 3.1.2 Weak-fragmentation systems

When fragmentation is weak (in a sense to be made precise soon) an extraordinarily interesting phenomenon occurs which is physically interpreted as corresponding to the existence of a dynamic phase transition in the system being modelled by (52). The phenomenon is the following: there exists a critical density  $\rho_s \in (0, \infty)$  such that

- (i) if the initial condition  $c_0$  has density  $\rho > \rho_s$ , then the solution  $c$  to (52) converges weak-\*, but not strongly, to the only equilibrium  $c^{\rho_s}$  with density  $\rho_s$  (supercritical case),
- (ii) if the initial condition  $c_0$  has density  $\rho \leq \rho_s$ , then the solution  $c$  to (52) converges strongly in  $X_1^+$  to the unique equilibrium  $c^\rho$  with density  $\rho$  (subcritical case).

Note that in case (i) the density of the  $\omega$ -limit solution,  $c^{\rho_s}$ , is strictly smaller than the density of the solution to (52) in every time instant  $t < \infty$ , whereas in case (ii) the density is also conserved in the limit.

Before going through a brief history of this result and analysing its proof, it is interesting to attend to a possible phase transition interpretation of this behaviour.

If we consider that each component  $c_j$  of the solution  $c = (c_j)$  represents the concentration of a microscopic  $j$ -cluster in a certain physical state, a gas say, and that  $\rho$  is the vapour density, the quantity  $\rho_s$  can be interpreted as the saturation density of the system. Thus, if the system is in a supersaturated state (i.e., a state with  $\rho > \rho_s$ ), there is no vapour equilibrium state with that density and the system will evolve to an equilibrium with density exactly equal to the saturated density. The excess density  $\rho - \rho_s$  disappears from the gaseous system via condensation, which

corresponds to the formation of another physical phase not modelled by none of the variables  $c_j$  but, heuristically, corresponding to a cluster incommensurably bigger than  $j$ , for every  $j$ ).

If the system is in a saturated or in a sub-saturated state, with density  $\rho \leq \rho_s$ , then its evolution proceeds to the unique equilibrium with that density, the density being conserved along the process and in the limit state.

The behaviour described above was first observed in the context of the Becker-Döring equations by Ball, Carr and Penrose in [13]. The (weak-\*) convergence to an equilibrium is proved using a Lyapunov function, as in the case of strong fragmentation described earlier (cf. page 40), however in the present case the finiteness of higher order moments is not valid, fact that increases the difficulty of the proof of orbit pre-compactity and the identification of the limit density in the subcritical case. In order to obtain these results, in [13] there was the need to impose the following extra decay condition on the initial data (typically, an exponential decay [13, Eq.(5.10)]):

$$\sum_{j=1}^{\infty} \frac{c_{0j}}{M_j z_s^j} < \infty, \text{ where } z_s \text{ is the only solution of } F(z_s) = \rho_s, \quad (85)$$

which provided the sought for control on the tail of the solution  $(c_j(t))$ . This restriction was later eliminated in [11] by noting that, for the variables  $x_n := \sum_{j=n}^{\infty} j c_j$ , it is possible to construct a supersolution independently of the decay behaviour of the initial condition  $x_0$ , and this implies pre-compactity of the orbit in  $X_1^+$  and thus strong convergence in  $X_1$ , which has as consequence that the limit equilibrium has the same density of the solution at finite times.

The extension of this result to coagulation-fragmentation systems (52) more general than the Becker-Döring was marred with several difficulties and was only truly achieved two decades later, with the work of Cañizo [30]. A first attempt was made by Carr and da Costa in [36] where, in order to prove strong convergence in the subcritical case, the following generalized Becker-Döring assumption was used:

$$a_{j,k} = b_{j,k} = 0 \quad \text{if } j \wedge k > N, \quad (86)$$

where  $N$  is a fixed positive integer. In the classic Becker-Döring case  $N = 1$  (cf. (18)). With this assumption, with the restriction (85) on the initial data, and with some technical assumptions on the kinetic coefficients, like the ones used in [13], it was possible to rigorously prove the behaviour described in (i) and (ii) above. A subsequent attempt to overcome the restrictions about the regularity of the initial condition was made by da Costa in [48] but was only partially successful: the result obtained, inspired in the method of [11], only allows to draw conclusions for initial data in  $X_1^+$  that, although has no extra decay requirement, needs to have its density  $\rho$  bounded above by a bound like  $\rho_N \sim \mathcal{O}(N^{-1})$  when  $N \rightarrow \infty$ . This certainly seems to point to the fact that the method is not only insufficient to deal with the generalized Becker-Döring, but it really is totally inadequate for the general coagulation-fragmentation (that corresponds, formally, to take  $N \rightarrow \infty$ ).

Notwithstanding these failures, the idea to construct supersolutions, introduced in [11], was a good one and could finally be carefully exploited by Cañizo in [29] to get rid of the restriction on the initial density imposed in [48]. With the assumptions on the coefficients used in previous works [36, 48], Cañizo used an argument similar to the one of Ball and Carr and proved [29, Proposition 3.3] that, if  $z < z_s$ ,  $\lambda \in (1, \frac{z_s}{z})$ , and if  $(\lambda_j)$  is a decreasing sequence such that

$$\frac{\lambda_{j-1} - \lambda_j}{\lambda_j - \lambda_{j+1}} < \lambda,$$

then, when the initial condition (in the  $x_n$  variables introduced above) satisfy  $x_n(0) \leq \lambda_n$  for all  $n$ , then there exist positive constants  $C$  and  $n_0$  such that  $x_n(t) \leq C\lambda_n$  for all  $n \geq n_0$  and  $t > 0$ . The proof of this pre-compactness result is based on the following differential inequality for  $H_j(\cdot) := (x_j(\cdot) - C\lambda_j)^+$ , where  $u^+ = u \vee 0$ ,

$$\frac{d}{dt} \sum_{j=n_0}^{\infty} H_j \leq (\text{const.}) \sum_{j=n_0}^{\infty} H_j,$$

and on the application of Gronwall's lemma.

In the remaining of this section we shall present a more general result, also due to Cañizo, [30], that overcomes the need to impose (86) and proves the phase transition behaviour for the general coagulation-fragmentation system and, in a sense, completes the work started by Ball, Carr and Penrose in 1986 with the Becker-Döring system. Cañizo work [30] imposes an additional decay to the initial data  $c_0 \in X_1^+$  but it is typically the existence of a moment of order smaller than two, and not an exponential decay, as in [13, 36]. This very mild restriction is more than compensated by the fact that the result is valid for the general coagulation-fragmentation equations, and not only to the restrictive Becker-Döring versions. At present it is not clear if this restriction is essential. The hypotheses considered in [30] are the following:

(H3) There exists constants  $K > 0$ ,  $\gamma \in \mathbf{R}$  and  $\lambda \in [0, 1)$  such that

$$a_{j,k}, b_{j,k} \leq K(j^\lambda + k^\lambda) \quad (87)$$

$$\sum_{j=1}^{i-1} b_{j,i-j} \leq Ki^\gamma, \quad \forall i \geq 1. \quad (88)$$

(H4) There exists a positive sequence  $(M_j)$  satisfying (15).

(H5)  $\lim M_j^{1/j} = z_s^{-1} \in (0, \infty)$  and  $\rho_s := F(z_s) \in (0, \infty]$ , where  $F$  is given by (74).

(H6) The sequence  $(M_j z_s^j)$  is monotone decreasing.

(H7) There exists a constant  $K_1 > 0$  such that

$$a_{j,1} \geq K_1 j^\lambda, \quad \forall j \geq 1 \quad (89)$$

(H8) The initial data are  $c_0 \in X_\mu^+$ , with  $\mu := \max\{2 - \lambda, 1 + \lambda, 1 + \gamma\}$ .

The main result, extending to coagulation-fragmentation the result of [13], is the following:

**Theorem 12** [30] *Assume (H3)–(H8). Let  $c$  be a solution of (52) with (constant) density  $\rho = \|c\|_1 = \|c_0\|_1$ . The following holds:*

- (i) *If  $\rho > \rho_s$ , then  $c(t) \xrightarrow{*} (M_j z_s^j)$  when  $t \rightarrow +\infty$ .*
- (ii) *If  $\rho \leq \rho_s$ , then  $c(t) \rightarrow c^{\text{eq}}$  strongly in  $X_1$ , when  $t \rightarrow +\infty$ , where  $c^{\text{eq}}$  is the only equilibrium solution with density  $\rho$ .*

*Sketch of proof:* The general strategy to prove this result was already used in [13] and in [29, 36] and consists of proving that, if a solution converges weak-\* to an equilibrium with density strictly smaller than the critical one  $\rho_s$ , then the convergence is actually strong in the norm topology of  $X_1$  and thus the limit density is equal to the initial one.

That all solutions of (52) converge weak-\* to equilibria had already been proved in [36]: assuming (H3)–(H8), solutions  $c$  to (52) satisfy  $c(t) \xrightarrow{*} c^\rho$  when  $t \rightarrow \infty$ , for some  $\rho \leq \min\{\|c_0\|, \rho_s\}$ , where  $c^\rho$  is the only equilibrium solution with density  $\rho$ . As in the case of strong fragmentation presented earlier, the proof of this result of weak-\* convergence to equilibria is based on the existence of a Lyapunov function. The proof in [30] is based on the Lyapunov function, on the control of the density by the  $(2 - \lambda)$ -moment, and in an estimate that implies that the growth of this moment is at most linear in  $t$ .

Cañizo also proved a result on the rate of convergence to equilibria using a slightly different Lyapunov function introduced by Jabin and Niethammer in the study of the rate of convergence to equilibria in Becker-Döring systems [109]: for  $c \in X_1^+$ , let  $V$  be defined by (75) and, for  $z \in (0, z_s]$ , define the *energy of  $c$  relative to the equilibrium  $(M_j z^j)$*  by the expression

$$\mathcal{V}_z(c) := V(c) - (\log z) \sum_{j=1}^{\infty} j c_j + \sum_{j=1}^{\infty} M_j z^j. \quad (90)$$

Observe that, if  $\rho_s < \infty$  and if we choose  $z$  so that  $c^{\text{eq}} = (M_j z^j)$  satisfies  $\|c^{\text{eq}}\| = \|c\|$ , then, it is easy to conclude that  $\mathcal{V}_z(c) = V(c) - V(c^{\text{eq}})$ , which justifies the name given to  $\mathcal{V}_z(c)$ .

The fundamental inequality used in the proof of Theorem 12, relating the density of a positive sequence  $c = (c_j)$  with its  $(2 - \lambda)$ -moment, is

$$\|c\| - \sum_{j=1}^{\infty} j M_j c_j^1 \leq C \sqrt{D} \sqrt{\|c\|_{2-\lambda}}, \quad (91)$$

where  $C$  is a positive constant and  $D := D(c)$  is given by

$$D(c) := \sum_{j=1}^{\infty} a_j M_j \left( \frac{c_1 c_j}{M_j} - \frac{c_{j+1}}{M_{j+1}} \right) \left( \log \frac{c_1 c_j}{M_j} - \log \frac{c_{j+1}}{M_{j+1}} \right), \quad (92)$$

where  $a_j = a_{j,1}$  if  $j \geq 2$ , and  $a_1 = \frac{1}{2}a_{1,1}$ . The inequality is valid under the assumptions (H3)–(H7) and provided  $c_1 \in (0, z_s)$  and  $c \in X_{2-\lambda}$ . The function  $D$  is called the *Becker-Döring free energy dissipation rate* in [30], which is a natural designation since it was proved in [13] that the time evolution of the Lyapunov function  $V$  along solutions  $c(t)$  of the Becker-Döring system satisfies

$$V(c(t)) = V(c(0)) - \int_0^t D(c(s))ds.$$

Observe that, as pointed out before,  $\forall x, y > 0$ ,  $(x - y)(\log x - \log y) \geq 0$ , and so the function  $D(c)$  is non-negative.

Its should be noted that (91) is a purely algebraic relation valid for certain sequences  $c$  and has nothing to do with these sequences being solutions of some differential equation. It is merely a consequence of the following estimate about the tail of the series  $\sum_j jM_j c_1^j$ , which is valid under the same assumptions,

$$\sum_{i=j+1}^{\infty} iM_i c_1^i \leq jM_{j+1} c_1^{j+1}.$$

Under the hypotheses of Theorem 12, taking a solution  $c = c(t)$  of (52) with initial condition  $c_0 \in X_{2-\lambda}$ , and using the approximation of  $c$  by solutions to the truncated systems, we can prove that  $\|c(t)\|_{2-\lambda}$  satisfies the differential inequality  $\frac{d}{dt}\|c(t)\|_{2-\lambda} \leq (\text{const.})\rho^2$ , and thus, for some constant  $C$  independent of  $t$ , it holds that

$$\|c(t)\|_{2-\lambda} \leq C(1+t) \quad (93)$$

As stated above the idea of the proof consists in showing that if a solution with initial density  $\rho_0$  converges weak-\* to an equilibrium with density  $\rho < \rho_s$ , then it converges strongly in  $X_1$  and thus  $\rho = \rho_0$ .

Let us assume that  $c(t) \xrightarrow{*} c^{\text{eq}}$  when  $t \rightarrow +\infty$ , where  $c^{\text{eq}} = (M_j z^j)$  and  $z < z_s$ . Thus  $c_1(t) \rightarrow z < z_s$  and  $c_1(t) \leq \frac{z+z_s}{2} < z_s$ , for all times  $t > t_0$ , where  $t_0$  is sufficiently large. Using (91) and (93) we know that  $\rho - \rho_1(t) \leq C\sqrt{D}\sqrt{1+t}$ , for  $t \geq t_0$ , where  $\rho_1(t) = \|(M_j c_1(t)^j)\|_1$ , and  $C$  is a constant. By continuity of (74) in the interior of its interval of convergence,  $\rho_1(t) \rightarrow \rho_z := \|c^{\text{eq}}\|$ , as  $t \rightarrow \infty$ .

Now, either  $\rho - \rho_1(t) > 0$  after some  $t_1$ , or there exists a sequence  $t_n \rightarrow \infty$  such that  $\rho - \rho_1(t_n) \leq 0$ . Let us start by the first possibility: if  $\rho - \rho_1(t) > 0$  for all  $t > t_1$  the previous inequality gives the estimate  $D(c(t)) \geq \frac{(\rho - \rho_1(t))^2}{1+t} C^{-2}$  and the evolution of  $V$  along solutions satisfies

$$V(t) = V(t_1) - \int_{t_1}^t D_{CF}(c(s))ds \quad (94)$$

$$\leq V(t_1) - \int_{t_1}^t D(c(s))ds \quad (95)$$

$$\leq V(t_1) - C^{-2} \int_{t_1}^t \frac{(\rho - \rho_1(s))^2}{1+s} ds, \quad (96)$$

where  $D_{CF}$  is the *coagulation-fragmentation free energy dissipation rate* defined by

$$D_{CF}(c) := \frac{1}{2} \sum_{i,j=1}^{\infty} a_{i,j} M_i M_j \left( \frac{c_i c_j}{M_i M_j} - \frac{c_{i+j}}{M_{i+j}} \right) \left( \log \frac{c_i c_j}{M_i M_j} - \log \frac{c_{i+j}}{M_{i+j}} \right). \quad (97)$$

It is worth calling the reader attention to the fact that, although the formal derivation of the evolution equation (94) is trivial, its rigorous proof is far from being simple [36, Theorem 5.2]. The inequality (95) is due to the obvious fact that  $D_{CF}(c) \geq D(c) > 0$ . The Lyapunov function  $V$  is bounded from below along solutions; using this result the integral in the right-hand side of (96) has to be bounded from above and, since  $\rho_1(t) \rightarrow \rho_z$ , we conclude that  $\rho_z = \rho$ . But then, since  $c(t) \xrightarrow{*} c^{\text{eq}}$  and  $\|c(t)\| = \rho = \rho_z = \|c^{\text{eq}}\|$ , [13, Lemma 3.3] implies that  $c(t) \rightarrow c^{\text{eq}}$  strongly in  $X_1$ .

It remains to consider the possibility of existence of a sequence  $t_n \rightarrow \infty$  such that  $\rho - \rho_1(t_n) \leq 0$ . In this case we would have  $\rho \leq \rho_1(t_n) \rightarrow \rho_z$  and thus  $\rho \leq \rho_z$ . By the lower semicontinuity of the norm of  $X_1$  with respect to weak-\* convergence, we have  $\rho_z \leq \rho$  and thus  $\rho_z = \rho$ . This concludes the proof. ■

Once the long-time limit of solutions in Theorem 12 has been proved, a natural problem to consider is to clarify the way the limit equilibrium solution is approached as  $t \rightarrow \infty$ . Recall that in the strong fragmentation case solutions converge to the limit equilibrium exponentially fast, at least for sufficiently small initial data (cf. page 41). Note that, for that type of coefficients, the critical density is infinite and so all solutions are subcritical.

Under weak fragmentation conditions the long-time behaviour of solutions is expected to be richer, and the cases of supercritical and subcritical densities are thought to exhibit distinct behaviours. However, at present, rigorous results about these aspects are restricted to the Becker-Döring case, and we shall briefly review them next.

For the Becker-Döring equations with subcritical initial density a recent paper by Cañizo and Lods [31] improved previous results by Jabin and Niethammer [109] and prove that, under appropriate assumptions that include exponentially decaying initial conditions, subcritical solutions converge exponentially fast to the limit equilibrium, and give an estimate for the convergence rate. The precise statement of the result requires the introduction of some hypotheses:

(H9) On the coagulation coefficients:

$$a_j = \mathcal{O}(j) \text{ as } j \rightarrow \infty, \quad \lim_{j \rightarrow \infty} \frac{a_{j+1}}{a_j} = 1, \quad \inf_j a_j > 0.$$

(H10) On the fragmentation coefficients:  $b_j = \mathcal{O}(j)$  as  $j \rightarrow \infty$ .

(H11) On the partition function:  $\lim_{j \rightarrow \infty} \frac{M_j}{M_{j+1}} =: z_s \in (0, +\infty)$

Under these conditions the following was proved:

**Theorem 13** [31] *Assume (H9)–(H11). Let  $c$  be a solution of (19) with initial condition  $M := \sum_{j=1}^{\infty} e^{vj} c_j(0) < +\infty$ , for some  $v > 0$ , and let  $z > 0$  be the monomer*

density of the corresponding limit equilibrium  $c^{\text{eq}} = (M_j z^j)$ . Then, there exists  $\bar{v} \in (0, v)$  and  $\lambda_* > 0$ , such that, for every  $\eta \in (0, \bar{v})$ , there is  $C > 0$ , depending only on  $\rho, \eta, M$ , and  $\sum_{j=1}^{\infty} e^{\eta j} M_j z^j$ , such that the following holds for all  $t \geq 0$ ,

$$\sum_{j=1}^{\infty} e^{\eta j} |c_j(t) - M_j z^j| \leq C e^{-\lambda_* t},$$

where, if  $\lim_{j \rightarrow \infty} a_j = +\infty$ , we can take  $\lambda_*^{-1} = \sup_k \left( \sum_{j=k+1}^{\infty} M_j z^j \right) \left( \sum_{j=1}^k \frac{1}{a_j M_j z^j} \right)$ .

The proof of this theorem is rather lengthy and involved and we will not delve with it here: the interested reader should consult the original paper [31]. We just point out that the main tool is an appropriate linearisation of the Becker-Döring equations around the equilibrium  $c^{\text{eq}}$  and the proof of an appropriate spectral gap in suitable sequence spaces.

Let us now briefly consider the case of supercritical solutions to the Becker-Döring system. As stated in Theorem 12(i), solutions (all of them conserve initial density  $\rho$  in finite times) converge to the limit equilibrium with critical density  $\rho_s < \rho$ . That, in general, this convergence can be a complicated dynamical process has been shown by Penrose in [180], where he proved that, for certain nonequilibrium initial conditions, each component of the solution of (19) remain exponentially close to the initial condition for a time that is exponentially long in  $(\rho - \rho_s)^{-1}$ , after which it converges to the critical equilibrium. This is a metastability behaviour that, in some sense, agrees with the physical fact that nucleation processes in supersaturated mixtures are exceedingly slow processes.

The occurrence of metastability behaviour is an interesting feature of the Becker-Döring system. Another very interesting and challenging problem is to know what happens to the solution for times so large that all possible metastability regimes have elapsed. So, the problem is to understand how do the excess density  $\rho - \rho_s$  spreads to larger and larger clusters once the solution “starts to move”. This problem of evolution of large clusters has been studied, in the small excess density regime, by several authors [130, 166, 167, 168] striving to get rigorous proofs to the pioneering work Penrose and collaborators [181, 183] who established the relation of the large time asymptotic of small excess density solutions with solutions to the Lipschitz-Slyozov-Wagner equation of Oswald ripening [149, 214]. The detailed presentation of the existing rigorous results on this very interesting hydrodynamic limit of the Becker-Döring (as yet with no parallel in more general coagulation-fragmentation models) would take too long. In what follows we will just attempt to give an heuristic idea of the approach.

A very crude formal computation suggests a possible connection: if one considers extremely large clusters sizes  $j$ , then  $j - 1$  and  $j$  are extremely close to each other and, considering  $j$  as a continuous real variable, the right-hand side of the equation for the  $j$ -cluster in the Becker-Döring system is roughly equal to  $-\partial_j J$ , so the equation itself is  $\partial_t c + \partial_j J = 0$ ; the assumptions about the reaction coefficients  $a_j$  and  $b_j$  determine the way the flux  $J$  depends on  $c(t, j)$ , and the monomer dynamics is determined by density conservation. To make more precise this idea, let us



take the following situation, considered in Penrose's approach to the modelling of first-order phase transitions:

- (i)  $a_j = j^\alpha$ , for some  $\alpha \in [0, 1)$ .
- (ii)  $b_j = a_j(z_s + qj^{-\gamma})$ , for constants  $z_s, q > 0$ , and  $\gamma \in (0, 1)$ .

The more precise heuristic argument is roughly the following [166, 180, 202]: since we are considering large times, consider a new time scale  $\tau := \varepsilon^{1+\gamma-\alpha}t$ , with a small positive parameter  $\varepsilon \rightarrow 0$ . Adequately choosing a separation  $j_* = j_*(\varepsilon) \rightarrow +\infty$  between small and large clusters, we consider the large cluster sizes  $j > j_*$  as a continuous variable  $x = \varepsilon j$  and introduce the rescaled functions  $v(\tau, x)$ ,  $u(\tau, x)$ , and  $u(\tau)$ , such that  $c_j(t) = \varepsilon^2 v(\tau, x)$ ,  $J_j(c) = \varepsilon^{2+\alpha-\gamma} v(\tau, x)$ , and  $c_1 = z_s + \varepsilon^\gamma u(\tau)$ , respectively. The equation for large cluster sizes becomes, in the limit  $\varepsilon \rightarrow 0$ ,

$$\partial_\tau v + \partial_x (x^\alpha (u - qx^{-\gamma}) v) = \sigma(1). \quad (98)$$

In the other hand, density conservation and a judicious choice of  $j_*$  lead to, in the limit  $\varepsilon \rightarrow 0$ ,

$$\int_0^\infty x v(\tau, x) dx = \rho - \rho_s + \sigma(1), \quad (99)$$

which is equivalent to

$$u(\tau) = \frac{q \int_0^\infty x^{\alpha-\gamma} v(\tau, x) dx}{\int_0^\infty x^\alpha v(\tau, x) dx},$$

at least for some relation between  $\alpha$  and  $\gamma$  [166].

System (98)-(99) is the classic Lipschitz-Slyozov-Wagner model. We direct the interested reader to the references above for the precise statement and proof of this result.

### 3.2 Self-similar behaviour of solutions

Contrasting to what happens with the coagulation-fragmentation system, in Smoluchowski's coagulation equations it is easy to prove that the identically zero sequence is the only non-negative equilibrium, and the proof that every solutions converges, in the weak-\* sense, to this equilibrium as  $t \rightarrow \infty$  is also elementary (cf. below, and Subsection 3.2.1).

The zero sequence has density equal to zero and, being this density value the largest for which there is an equilibrium, it can be considered the critical density of the system and so, in a sense, all non-zero solutions are supercritical. So, the problem considered in the closing part of last subsection, namely how does the excess density is spread to larger and larger cluster sizes as  $t \rightarrow \infty$ , is also relevant in the Smoluchowski's equation setting, where its concretization has taken the form of the investigation of self-similar behaviour of solutions, i.e., the existence of a function

(or family of functions) for which, after an appropriate rescaling of the variables, all solutions converge when  $t \rightarrow \infty$ . This problem, of clear scientific importance, has received a good deal of attention in the mathematical modelling community (cf., e.g. [71, 72, 96, 144, 145, 146, 150, 217] and ref. cit.) but important progresses in its rigorous analysis are much more recent. In what follows we will review some of these.

### 3.2.1 Similarity behaviour in Smoluchowski's coagulation equations

Let us start by justifying the sentence above about the triviality of the convergence to equilibria in the Smoluchowski system:

**Theorem 14** [36] *Let  $a_{j,j} > 0$  for all  $j$ . Let  $c$  be a solution of (34) in  $[0, \infty)$  with  $c_0 \in X_1^+$ . Then  $c(t) \xrightarrow{*} 0$  as  $t \rightarrow +\infty$ .*

*Sketch of proof:* The heuristic idea is clear enough: since the coagulation process entails the increase of the clusters' mean size (cf. Section 1.2) we expect the total density of clusters with sizes below any arbitrarily fixed value to decrease with time. The convergence proof is based in this monotonicity property.

Let  $c$  be a solution of (34) and, for each  $n \in \mathbb{N}$ , let us consider

$$p_n(t) := \sum_{j=1}^n j c_j(t). \quad (100)$$

This function measures the total density at time  $t$  of clusters with size not larger than  $n$ . By (100) and the definition of solution we get, for all  $t, \tau \geq 0$ ,

$$p_n(t + \tau) - p_n(t) = - \int_t^{t+\tau} \sum_{T_{1,n}^4} j a_{j,k} c_j(s) c_k(s) ds \leq 0, \quad (101)$$

where  $T_{1,n}^4$  was defined in page 35. As  $c_n(t)$  and  $p_n(t)$  are non-negative functions, there exists a non-decreasing positive sequence  $(\bar{p}_n)$  such that  $p_n(t) \rightarrow \bar{p}_n$  when  $t \rightarrow +\infty$ . Since  $c_n(t) = \frac{p_n(t) - p_{n-1}(t)}{n}$ , these functions  $c_n(t)$  also converge to some constants,  $\bar{c}_n := \frac{\bar{p}_n - \bar{p}_{n-1}}{n} \geq 0$ , as  $t \rightarrow +\infty$ . By induction in the coagulation equation integrated in  $[t, t + \tau]$  the conclusion that  $\bar{c}_n \equiv 0$  is easily reached [36]. ■

We are now interested in knowing whether or not solutions to (34) converge to the zero solution in a self-similar way. In a slightly more precise manner: under what conditions there exists a function  $\Phi$  such that, for a large class of initial conditions, the corresponding solutions to (34) satisfy

$$c_j(t) \approx \zeta(t)^{-a} \Phi(j \zeta(t)^{-b}), \quad \text{as } t \rightarrow +\infty \text{ and } j \rightarrow +\infty, \quad (102)$$

where  $\zeta(\cdot)$  is a positive increasing function, and  $a$  and  $b$  are positive constants?

Not much is rigorously known about the problem in this general setting. What we present next are answers for certain particular coefficients  $a_{j,k}$  (constant, additive,

product) for which rigorous and fairly complete answers have been obtained, and then point to very recent rigorous analysis for systems with more general classes of rate coefficients.

We start by the constant coefficient case<sup>8</sup>,  $a_{j,k} = 2$ . This case was studied by Kreer and Penrose [117] and by da Costa [47] using an idea first introduced by Lushnikov [150] which is based in exploiting the generating function

$$\varphi(z, t) := \sum_{j=1}^{\infty} c_j(t) z^j, \quad |z| \leq 1. \quad (103)$$

Observe that (103) is the discrete Laplace transform  $\sum_{j=1}^{\infty} c_j(t) e^{-jw}$ , ( $\text{Re}(w) \geq 0$ ), of the solution  $c$  of (34). It is easy to prove that  $\varphi$  is solution of the initial value problem

$$\begin{cases} \frac{d\|c\|_0}{dt} = -\|c\|_0^2 \\ \frac{\partial \varphi}{\partial t} = \varphi^2 - 2\|c\|_0 \varphi \end{cases} \quad (104)$$

with  $\|c(0)\|_0 = N_0 := \|c_0\|_0$  and  $\varphi(z, 0) = \phi(z) := \sum_{j=1}^{\infty} c_{0j} z^j$ , from which we immediately conclude that

$$\varphi(z, t) = t^{-2} \frac{1}{N_0 + t^{-1}} \frac{\phi(z)}{N_0 + t^{-1} - \phi(z)}. \quad (105)$$

Since  $\varphi(\cdot, t)$  is an analytic function on the unit open ball  $B_1 \subset \mathbf{C}$ , we use Cauchy's integral formula to write

$$t^2 c_j(t) = \frac{1}{2\pi i} \frac{1}{N_0 + t^{-1}} \oint_{\gamma_0} \frac{1}{z^{j+1}} \frac{\phi(z)}{N_0 + t^{-1} - \phi(z)} dz, \quad (106)$$

where  $\gamma_0 = \{z \in \mathbf{C} : |z| = r_0 < 1\}$ . In order to conclude something about the long-time behaviour of the right-hand side of (106) we need to know the behaviour of the zeros of  $F(z, \tau) := N_0 + \tau - \phi(z)$  as  $\tau \rightarrow 0$ . With the additional hypothesis of an exponentially decaying initial condition  $c_{0j} \leq A(1 + \Delta)^{-j}$ , for some constants  $A \geq 0$  and  $\Delta \in (0, 1)$ , it can be proved that, for all sufficiently small  $\tau$  there exists  $q$  simple zeros of  $F(z, \tau)$ ,  $z_k(\tau)$ , satisfying  $|z_k(\tau)| > 1$  and  $z_k(\tau) = \omega_q^k \left(1 + \frac{1}{\|c_0\|_1} \tau + \mathcal{O}(\tau^2)\right)$ , when  $\tau \rightarrow 0$ , where  $\omega_q$  is the  $q^{\text{th}}$  root of unity,  $e^{2\pi i/q}$ ; all the remaining roots of  $F(z, \tau)$  are in the exterior of  $B_1$  and keep a distance uniformly positive from  $B_1$  when  $\tau \rightarrow 0$ . The positive integer constant  $q$  is given by  $q = \gcd \mathcal{J}(0)$ , where  $\mathcal{J}(0)$  is the set of subscripts  $j$  for which  $c_{0j} > 0$  (cf. statement of Theorem 1). With this result and the representation formula (105) it is not difficult to prove the following:

**Theorem 15** [47, 117] *Let  $a_{j,k} \equiv 2$  and consider a non-negative exponentially decreasing initial condition  $c_0 \in X_1^+$ . Let  $q$  and  $\mathcal{J}(0)$  be as above. Thus, the solution  $c$  of (34) has the following self-similar behaviour:*

<sup>8</sup> The value of the constant is irrelevant for the result since it can always be transformed into another value by a time rescaling. The choice we make simplifies the computations a bit.

$$\lim_{\substack{j,t \rightarrow +\infty \\ \xi = j/t \text{ fixed} \\ j \in \text{span}_{\mathbb{N}_0} \mathcal{J}(0)}} t^2 c_j(t) = \frac{q}{\|c_0\|_1} e^{-\xi/\|c_0\|_1}. \quad (107)$$

A version of this result valid for the continuous Smoluchowski's equations, was also proved by Kreer and Penrose in [117], also for exponentially decaying initial data. The same behaviour occurs with the obvious changes of notation:

$$\lim_{\substack{j,t \rightarrow +\infty \\ \xi = j/t \text{ fixed}}} t^2 c(x,t) = \frac{1}{\|c(x,0)\|_1} e^{-\xi/\|c(x,0)\|_1}. \quad (108)$$

If the coefficients  $a_{j,k}$  (or  $a(x,y)$ ) are not constants, or if the initial condition does not decay exponentially, the above technique is not applicable. In these cases, only recently the rigorous analysis of the self-similar behaviour of solutions was achieved, in a number of notable papers by Menon and Pego [159, 160, 161] (see also [179]). These works use a more general notion of solution, in terms of measures, allowing for the simultaneous consideration of the discrete and the continuous cases of the Smoluchowski's equation. Using the modified Laplace transform  $\varphi(z,t) := \int_0^\infty (1 - e^{-zx}) v_t(dx)$ , where  $v_t(dx) = c(x,t)dx$  is the finite measure on  $(0,\infty)$  that describes the cluster size distribution, Menon and Pego proved the following:

**Theorem 16** [179] *Let  $a(x,y) \equiv 2$ , and  $t_0 = 1$  and consider initial data satisfying  $\int_0^\infty v_1(dx) = 1$ . Let  $v_t(dx)$ , be a finite measure solution of (5)-(6), and let  $F_t$  be the probability distribution function*

$$F_t(x) := \frac{\int_0^x v_t(dy)}{\int_0^\infty v_t(dy)} = t \int_0^x c(t,y) dy. \quad (109)$$

1. *Suppose there exists a  $\lambda(t) \rightarrow \infty$  and a probability distribution  $F_*$  such that  $F_*(x) < 1$  for some  $x > 0$ , and*

$$F_t(\lambda(t)x) \rightarrow F_*(x), \text{ as } t \rightarrow \infty, \quad (110)$$

*in its points of continuity. Then*

$$\int_0^x y v_1(dy) \sim x^{1-\rho} L(x), \text{ as } x \rightarrow \infty, \quad (111)$$

*for some constant  $\rho \in (0,1]$  and some function  $L$  slowly varying at infinity [87, pp. 275-9].*

2. *Reciprocally, suppose (111) is true. Then (110) holds, with*

$$F_*(x) := F_\rho(x) = \sum_{k=1}^{\infty} \frac{(-1)^{k+1} x^{\rho k}}{\Gamma(1 + \rho k)}, \quad (112)$$

*being a Mittag-Leffler distribution [87, page 453], whose Laplace transform is  $\int_0^\infty e^{-zx} F_*(dx) = \frac{1}{1+q^\rho}$ .*

Observe that Theorem 16 provides a classification of all possible self-similar solutions of the Smoluchowski's equation with constant coefficients, which are solutions of the type

$$c(t, x) = t^{-1-1/\rho} n_\rho(t^{-1/\rho} x), \quad \rho \in (0, 1], \quad (113)$$

where  $n_\rho(\cdot) = F'_\rho(\cdot)$  is the Mittag-Leffler distribution density (112).

Using the Laplace transform, the self-similar *ansatz* like (102) for the transform, and separation of variables, it is easy to check that the functions (113) are in fact solutions to (5)-(6).

It is interesting to observe that if  $v_1(dx)$  has finite density, then  $\rho = 1$  and  $F_1(x) = 1 - e^{-x}$ , corresponding to the solution  $c(t, x) = \frac{1}{t^2} e^{-x/t}$ . Compare this with (108).

This result is similar to the Central Limit Theorem. The distributions  $F_\rho$  with  $\rho \in (0, 1)$  have infinite density and correspond to Lévy's stable distributions.

Menon and Pego also proved that the convergence to the self-similar limit is uniform in the similarity variable  $\frac{x}{t}$ :

**Theorem 17** [160] *Let  $c(1, x) > 0$ , be an initial condition satisfying  $\int_0^\infty c(1, x) dx = \int_0^\infty xc(1, x) dx = 1$ . Suppose the Fourier transform of  $xc(1, x)$  is integrable. Then*

$$\lim_{t \rightarrow +\infty} \sup_{\frac{x}{t} > 0} \frac{x}{t} \left| t^2 c(t, x) - e^{-x/t} \right| = 0.$$

An improved version, establishing convergence in a stronger topology and providing an upper bound  $Kt^{-\delta}$  for the rate of this convergence to zero was obtained by Cañizo, Mischler and Mouhou on [33]. The result of this theorem is also valid, *mutatis mutandis*, for the solutions of the discrete system (cf. [160, Theorem 2.2]).

Results similar to those above were also proved by Menon and Pego [159] for the two solvable types of coefficients (additive  $a(x, y) = x + y$ , and product  $a(x, y) = xy$ ) for which they also obtained a complete characterization of the self-similar attractor [161]. For these kernels the rate of the convergence stated in the theorem was proved to be exponential by Ravi Srinivasan [207] also using methods based on the Laplace transform in an approach that is analogous, in spirit, to the Berry-Esséen theorem when one considers the result in theorems 16 and 17 as a Central Limit Theorem for the clusters distributions.

The first rigorous proofs of existence of self-similar solutions for non-solvable kernels are by Fournier and Laurençot [90] and Escobedo and collaborators [82], who proved the existence of (but not the convergence to) self-similar solutions to the continuous system (5)-(6). Their approach is different from those presented above, not resorting to Laplace transforms, and returning to an idea commonly used in the mathematical modelling and physics literatures [72, 144], which is to use the *ansatz* (102) directly in (5)-(6) in order to obtain an integro-differential equation for the self-similar profile  $\Phi(\cdot)$ , and to prove that, for certain types of coefficients, this equation has a non identically zero weak solution. This is an extremely natural approach from a mathematical perspective and was certainly attempted before; the fact that only in [82, 90] this idea could have been set on a firm basis attests to

the very demanding technical difficulties its concretization involves. We will now present a very brief description of the result and approach of [90].

Write

$$c(t, x) = \varsigma(t)^{-2} \Phi(\varsigma(t)^{-1} x) \quad (114)$$

and assume the coagulation coefficients satisfy the homogeneity condition

$$a(ux, uy) = u^\lambda a(x, y), \quad \forall u, x, y \in \mathbf{R}^+, \quad (115)$$

for some real constant  $\lambda$ .

Substituting (114) into (5)-(6) and using (115) the following equation arises

$$\gamma \frac{d}{dx} (x^2 \Phi(x)) + x Q_c(\Phi)(x) = 0 \quad (116)$$

$$\int_0^\infty x \Phi(x) dx = \rho, \quad (117)$$

for the unknown function  $\Phi$  and the real positive unknowns  $(\gamma, \rho)$ .

It is easy to conclude that, if  $(\Phi, \gamma, \rho)$  is a solution of (116)-(117), then each of the elements of the two parameter family  $(a\Phi(bx), a\gamma b^{-1-\gamma}, a\rho b^{-2})$  is also a solution of (116)-(117). This means that, without loss of generality, we can consider  $\gamma = \frac{1}{1-\lambda}, \rho = 1$ .

A non-negative function  $\Phi \in L^1(0, \infty, x dx)$  is a weak solution of (116) if  $\Phi \in L^1(0, \infty, x^2 dx)$ , if  $(x, y) \mapsto xya(x, y)\Phi(x)\Phi(y) \in L^1(\mathbf{R}^+ \times \mathbf{R}^+)$ , and if

$$\gamma z^2 \Phi(z) = \int_0^z \int_{z-x}^\infty a(x, y) x \Phi(x) \Phi(y) dy dx, \quad (118)$$

for almost all  $z \in \mathbf{R}^+$ . Thus, if  $\Phi$  is a weak solution of (116) we have, for every  $\phi \in C_b^1([0, \infty))$ ,

$$\gamma \int_0^\infty x^2 \Phi(x) \phi'(x) dx = \int_0^\infty \int_0^\infty xa(x, y) (\phi(x+y) - \phi(x)) \Phi(x) \Phi(y) dy dx. \quad (119)$$

From a technical viewpoint, it is more suitable to consider the unknown function  $\tilde{\Phi}(x) = x\Phi(x)$  instead of  $\Phi(x)$ , and, instead of (119), to write the weak version of (116) as

$$\gamma \int_0^\infty x \tilde{\Phi}(x) \phi'(x) dx = \int_0^\infty \int_0^\infty \frac{a(x, y)}{y} (\phi(x+y) - \phi(x)) \tilde{\Phi}(x) \tilde{\Phi}(y) dy dx. \quad (120)$$

The main result of [90] is the following:

**Theorem 18** [90] *Consider coagulation coefficients satisfying any one of the following conditions:*

- (i)  $a(x, y) = (x^\alpha + y^\alpha)(x^{-\beta} + y^{-\beta})$ ,  $\alpha \in [0, 1], \beta \in \mathbf{R}^+, \lambda = \alpha - \beta \in (-\infty, 1)$
- (ii)  $a(x, y) = (x^\alpha + y^\alpha)^\beta$ ,  $\alpha \in [0, \infty), \beta \in \mathbf{R}^+, \lambda = \alpha\beta \in [0, 1)$
- (iii)  $a(x, y) = x^\alpha y^\beta + x^\beta y^\alpha$ ,  $\alpha \in (0, 1), \beta \in (0, 1), \lambda = \alpha + \beta \in (0, 1)$

Let  $\gamma = \frac{1}{1-\lambda}$ ,  $\rho = 1$ . Then, there exists a positive weak solution  $\Phi$  of (116)-(117) and the function  $c_s(x, t) := t^{-2\gamma}\Phi(xt^{-\gamma})$ , with  $x, t > 0$ , is a (self-similar) weak solution of (5)-(6) with unit density for all  $t > 0$ .

The proof by Fournier and Laurençot starts by the following discretization of (120),

$$-\frac{\gamma}{n} (i\mathbf{1}_{1 \leq i \leq n^2-1} f_{i+1} - (i-1)f_i) = \sum_{j=1}^{i-1} \frac{1}{j} a\left(\frac{i-j}{n}, \frac{j}{n}\right) f_{i-j} f_j - \sum_{j=1}^{n^2-i} \frac{1}{j} a\left(\frac{i}{n}, \frac{j}{n}\right) f_i f_j.$$

Considering the solutions of this system of  $n^2$  equations as the stationary solutions of an appropriate system of ordinary differential equations, one concludes the existence of a non-negative solution  $f = \tilde{f}^n$  satisfying

$$\sum_{i=1}^{n^2} \tilde{f}_i^n = 1, \quad \forall n.$$

Using the solutions  $\tilde{f}^n$ , the following sequence of probability measures indexed by  $n$  is constructed

$$\tilde{\Phi}^n(dx) = \sum_{i=1}^{n^2} \tilde{f}_i^n \delta_{i/n}(dx), \quad (121)$$

and the *a priori* estimate

$$\sup_{n \geq 1} \int_0^\infty x^\sigma \tilde{\Phi}^n(dx) < \infty, \quad (122)$$

is proved, where the domain of the parameter  $\sigma$  depend of the type of coefficient, (i), (ii) or (iii), considered. From (121) and (122) one deduces the tightness of the sequence  $(\tilde{\Phi}^n(dx))$  and thus the existence of a probability measure  $\tilde{\Phi}(dx)$  and a subsequence  $(\tilde{\Phi}^{n_k}(dx))$  such that, for all functions  $\phi \in C_b^1([0, \infty))$ , it holds that

$$\lim_{k \rightarrow \infty} \int_0^\infty \phi(x) \tilde{\Phi}^{n_k}(dx) = \int_0^\infty \phi(x) \tilde{\Phi}(dx).$$

The last step in the proof is to establish that  $\tilde{\Phi}$  is a weak solution of (120) and thus  $\Phi(x) = \frac{\tilde{\Phi}(x)}{x}$  is a weak solution of (116)-(117).

The problem of self-similar dynamic behaviour in (5)-(6), i.e., the convergence of a “generic” solution of (5)-(6) to the self-similar ones, whose existence was proved in Theorem 18, is still largely open.

A natural approach to study this stability problem would be to consider a transformation like the following one, more general than (114),

$$c(t, x) = \zeta(t)^{-2} \varphi(\log \zeta(t), \zeta(t)^{-1} x),$$

and, substituting in (5)-(6), to obtain an evolution equation for  $\varphi$  that would allow to prove that, for an appropriate notion of convergence,  $\varphi(\log \zeta(t), \cdot) \longrightarrow \Phi(\cdot)$ ,

when  $t \rightarrow +\infty$ . This idea was successfully implemented, in the framework of weak convergence in  $L^1$ , in the case of constant coefficients  $a(x, y) \equiv 1$ , using Lyapunov functions whose construction were strongly dependent of the known form of the limit  $\Phi$ , [134], which turns the potentially promising method useless if the form of  $\Phi$  is not known. The idea was also applied with success in the prove of existence and stability of self-similar solutions in the Oort-Hulst-Safronov equations with constant [127], with additive [9], and with multiplicative [128] coefficients.

A number of recent studies have greatly enhanced our understanding of the self-similar behaviour of Smoluchowski's equation with non-solvable kernels. It was established by Fournier and Laurençot in [91] that, for sum type kernels  $a(x, y) = x^\lambda + y^\lambda$ , with  $\lambda \in (0, 1)$ , the self-similar profile  $\eta \mapsto \Phi(\eta)$  proved to exist in [82, 90] is continuously differentiable in  $\mathbf{R}^+$  decay exponentially fast as  $\eta \rightarrow \infty$  and is singular at  $\eta \rightarrow 0$ . Further improved results on the behaviour of the profile  $\Phi(\eta)$  when  $\eta \rightarrow 0$  were obtained in [32, 170] and when  $\eta \rightarrow \infty$  in [173]. A very interesting, albeit formal, study of the behaviour of the density conserving self-similar profiles in the limit  $\eta \rightarrow 0$  was published in [157].

The regularity of self-similar profiles in [91] was greatly improved by Cañizo and Mishler [32] who proved that, if  $a(x, y) = x^\alpha y^\beta + x^\beta y^\alpha$ , with  $-1 < \alpha \leq \beta < 1$  and  $\alpha + \beta \in (-1, 1)$ , then the profiles are  $C^\infty(\mathbf{R}^+)$ . Some results on the uniqueness of self-similar profiles have also been established [32, 172].

The existence of self-similar fat tail solutions (i.e., non-exponentially decaying profiles, as exists for solvable kernel equations) has been proved by Niethammer and Velázquez for diagonal kernels, in [169], and more recently, in [171], for homogeneous kernels satisfying  $a(x, y) \leq C(x^\lambda + y^\lambda)$ , with  $\lambda \in [0, 1)$ .

Most of these papers use an *ansatz* like (114) in order to get an equation for the profile  $\Phi$  that is then exploited recurring to a variety of means, comprising among other tools, rather delicate estimates and adequately carved fixed point theorems. We direct the reader to the original papers for the statement of the results and to fully appreciate the beauty and difficulty of their proofs.

With the exception of the exact case  $a(x, y) = xy$ , studied by Menon, Pego, and Srinivasan [159, 161, 207], all the above results correspond to systems for which solutions conserve density. The general problem of conservation, or non-conservation, of density will be treated in Section 4. Here we just briefly refer to a recent work by Breschi and Fontelos [25] which is the first rigorous proof of existence of self-similar solutions for a non-exact kernel for which solutions do not conserve density for all times, namely  $a(x, y) = (xy)^{1-\varepsilon}$ , with  $\varepsilon \ll 1$ . With these coefficients there exists a time  $T_g$  before which all solutions conserve density, but density decrease afterwards (cf. Theorem 21 in Section 4). In [25] the authors study the existence of self-similar solutions for  $t < T_g$ ; in particular they prove, among other things, that, with these coefficients, the Laplace transform of Smoluchowski equation results in a nonlocal Burgers' equation  $\omega(\lambda, t) = \frac{1}{2} \partial_\lambda (D_\lambda^{-\varepsilon} \omega(\lambda, t))^2$ , where  $\omega(\lambda, t) := -\int_0^\infty (e^{-\lambda x} - 1) x c(x, t) dx$ , and  $D_\lambda^{-\varepsilon}$  is a nonlocal operator. If one proves that this Burgers' equation has self-similar solutions of the form  $\omega(\lambda, t) = (T_g - t)^\alpha \psi(\xi)$ , where  $\xi := \lambda (T_g - t)^{-\beta}$ , one can use the inverse Laplace transform to prove that the original Smoluchowski equation has a corresponding self-similar solution



$\Phi(\eta) = \frac{1}{2\pi i} \frac{1}{\eta^2} \int_{-i\infty}^{i\infty} e^{\xi\eta} \psi'(\xi) d\xi$ . The equation satisfied by  $\psi$  is the following ordinary differential equation

$$-((1-2\varepsilon)\beta - 1)\psi(\xi) + \beta\xi\psi'(\xi) = \frac{1}{2} \frac{d}{d\xi} \left( D_{\xi}^{-\varepsilon} \psi(\xi) \right)^2,$$

the (rather non-trivial) analysis of which, using perturbative functional analytic techniques, is one of the accomplishments of [25].

### 3.2.2 Similarity behaviour in addition models with input of monomers

Another system for which the self-similar behaviour of solutions has been studied is the “addition model”, referred to in page 30, with input of monomers. Its kinetics consists of the Smoluchowski’s coagulation equation where the only coefficients that are eventually non-zero are those corresponding to reactions between clusters and monomers:  $a_{j,k} = 0$  if  $j \wedge k > 1$ . This kind of models are extensively used in studies of the early stages of submonolayer epitaxial growth, where a thin layer of a material is built on the surface of a crystal by bombarding it with monomers (see, for example, [10, 16, 101]). This is an extremely important technological process and has been theoretically modelled by a variety of techniques.

Using the notation adopted for the Becker-Döring system (cf. page 12) the mean field approach to these processes by coagulation equations consists in the following addition model

$$\begin{cases} \dot{c}_1 = J_1(t) - a_1 c_1^2 - c_1 \sum_{j=1}^{\infty} a_j c_j \\ \dot{c}_j = a_{j-1} c_1 c_{j-1} - a_j c_1 c_j, \quad j \geq 2, \end{cases} \quad (123)$$

where  $J_1(t)$  is a function describing the input rate of monomers.

The first rigorous studies of the self-similar behaviour of solutions to systems like (123) are relatively recent [54, 57, 58, 62, 195] and only for the case of constant rate coefficients  $a_j \equiv 1$ , and for monomer input rate of polynomial-like type  $J_1(t) = (1 + \varepsilon(t))\alpha t^\omega$ , where  $\alpha > 0$  and  $\omega$  are real constants and  $\varepsilon(\cdot)$  is a continuous function converging to zero at infinity. The methods used in these works are distinct from those presented above and are based, in an essential way, in the fact that by defining the auxiliary variable

$$c_0 := \sum_{j=1}^{\infty} c_j, \quad (124)$$

the equation (123) can be written as

$$\begin{cases} \dot{c}_0 = (1 + \varepsilon(t))\alpha t^\omega - c_0 c_1, \\ \dot{c}_1 = (1 + \varepsilon(t))\alpha t^\omega - c_0 c_1 - c_1^2, \\ \dot{c}_j = c_1 c_{j-1} - c_1 c_j, \quad j \geq 2. \end{cases} \quad (125)$$

The essential observation is that (125) can be studied by decoupling the system with the first two equations, for the variables  $(c_0, c_1)$ , from the remaining infinite dimensional system for the variables  $c_j(t)$  with  $j \geq 2$ . Furthermore, considering in this last system the change of time scale defining by  $t \mapsto \varsigma(t) := \int_{t_0}^t c_1(s)ds$ , the infinite system is transformed in the lower triangular linear system

$$\tilde{c}_j' = \tilde{c}_{j-1} - \tilde{c}_j, \quad j \geq 2, \quad (126)$$

where  $\tilde{c}_j(\varsigma) := c_j(t(\varsigma))$ . Clearly, (126) can be explicitly solved by the variation of constants formula to get

$$\tilde{c}_j(\varsigma) = e^{-\varsigma} \sum_{k=2}^j \frac{\varsigma^{j-k}}{(j-k)!} c_k(0) + \frac{1}{(j-2)!} \int_0^\varsigma \tilde{c}_1(\varsigma-s) s^{j-2} e^{-s} ds. \quad (127)$$

Hence, to study the self-similar behaviour of solutions to (123) we can exploit the representation formula (127) if the needed information about the behaviour of the component  $\tilde{c}_1(\varsigma)$  of the solution is known, and this can in principle be obtained from the study of the long-time behaviour of solutions to the two-dimensional system

$$\begin{cases} \dot{c}_0 = (1 + \varepsilon(t))\alpha t^\omega - c_0 c_1 \\ \dot{c}_1 = (1 + \varepsilon(t))\alpha t^\omega - c_0 c_1 - c_1^2. \end{cases} \quad (128)$$

The result obtained in [54, 58] using this approach is the following:

**Theorem 19** [54, 58] *Let  $a_j \equiv 1$  and  $J_1(t) = (1 + \varepsilon(t))\alpha t^\omega$ , with  $\alpha > 0$ ,  $\omega > -\frac{1}{2}$ , and  $\varepsilon(t)$  a continuous function such that  $\varepsilon(t) \rightarrow 0$  when  $t \rightarrow +\infty$ . For  $r_0 := \frac{1-\omega}{2+\omega}$ , define  $Q_0(\omega) := \left(\frac{3}{(1+2\omega)\alpha}\right)^{\frac{1}{2+\omega}} \left(\frac{2+\omega}{3}\right)^{r_0}$ . Let  $(c_j)$  be a solution to (123) with initial condition  $c_j(0) \in X_0$ , and let  $\varsigma(t)$  and  $\tilde{c}_j(\varsigma)$  be as above. Then*

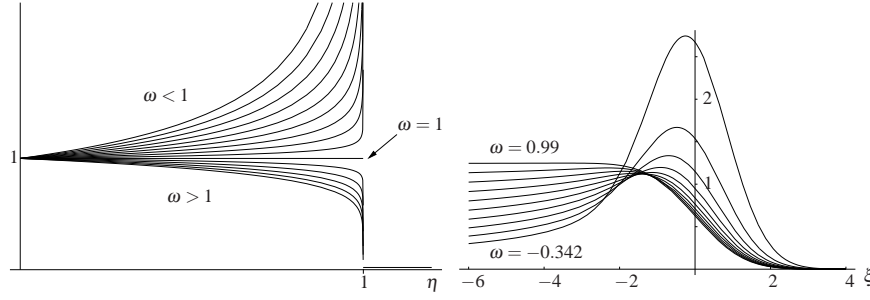
$$(i) \quad \lim_{\substack{j, \varsigma \rightarrow +\infty \\ \eta = j/\varsigma \text{ fixed} \\ \eta \neq 1}} Q_0(\omega) \varsigma^{r_0} \tilde{c}_j(\varsigma) = \Phi_{1,\omega}(\eta) := \begin{cases} (1-\eta)^{-r_0}, & \text{if } 0 < \eta < 1 \\ 0, & \text{if } \eta > 1, \end{cases}$$

(ii) *Furthermore, if  $c_j(0) = 0$  when  $j \geq 2$ , then*

$$\lim_{\substack{j, \varsigma \rightarrow +\infty \\ \xi = \frac{j-\varsigma}{\sqrt{\varsigma}} \text{ fixed} \\ \xi \in \mathbf{R}}} \left(\frac{\pi}{2}\right)^{\frac{1}{2}} Q_0(\omega) \varsigma^{\frac{1}{2}r_0} \tilde{c}_j(\varsigma) = \Phi_{2,\omega}(\xi) := e^{-\frac{1}{2}\xi^2} \int_0^{+\infty} y^{1-2r_0} e^{-\xi y^2 - \frac{1}{2}y^4} dy.$$

In Figure 9 we present the graphs of some similarity profiles  $\Phi_{1,\omega}$  and  $\Phi_{2,\omega}$  for several values of the parameter  $\omega$ . It is interesting to observe that the functions  $\Phi_{2,\omega}$  can be thought of as something like an inner expansion for the jump discontinuity that exists in the functions  $\Phi_{1,\omega}$  at  $\eta = 1$  when  $\omega \leq 1$ .

*Sketch of proof:* In order to use (127) to get the seeked for conclusions we need to know not only the limit of  $\tilde{c}_1(\varsigma)$  when  $\varsigma \rightarrow +\infty$ , but also its rate of convergence.



**Fig. 9** Graphs of the similarity limits in Theorem 19.

On the left:  $\Phi_{1,\omega}$  for values of  $\omega$  below and above 1 in steps of 0.1;

On the right:  $\Phi_{2,\omega}$  with  $\omega$  from  $-0.342$  to  $0.99$  in steps of  $0.148$ .

In order to obtain this information a detailed study of the long-time behaviour of the solutions of (128) is needed. In the autonomous case ( $\omega = 0$  and  $\varepsilon(t) \equiv 0$ ), this study can be done using invariant regions for the dynamics of (128), a change of variables suggested by Poincaré's compactification and the use of central manifolds techniques [57], or else using only elementary (but somewhat more elaborate) arguments based on invariant regions and monotonicity [52]. For the non-autonomous case all of these approaches do not seem to be applicable and the study uses an *ansatz* for a non-autonomous change of variables that is suggested by Wattis in [218]. In the new variables system (128) takes the form

$$\begin{cases} x' = (1 + \varepsilon(\tau) - xy) - A\tau^{-\frac{1}{2}}x^2 + B\tau^{-1}x \\ y' = (1 + \varepsilon(\tau) - xy) \cdot A\tau^{-\frac{1}{2}} - A^2\tau^{-1}y, \end{cases} \quad (129)$$

where  $\tau$  is the new time, related with the variable  $t$  used in (128) by  $\frac{d\tau}{dt} = \left(\frac{3\alpha^2}{1+2\omega}\right)^{1/3} t^{\frac{1+2\omega}{3}}$ , the constants  $A$  and  $B$  are defined by  $A := \left(\frac{1+2\omega}{4+2\omega}\right)^{\frac{1}{2}}$ , and  $B := \frac{1-\omega}{4+2\omega}$ , respectively, and the vector  $(x, y)$  is obtained from  $(c_1, c_0)$  by the same non-autonomous change of variables. Exploiting some differential inequalities, the behaviour of the auxiliary functions  $h := xy$  and  $b := y - A\tau^{-\frac{1}{2}}x$  along solutions, and methods from the qualitative theory of ordinary differential equations, leads to the following [58, 54]:

$$\left(\frac{3}{\alpha(1+2\omega)}\right)^{\frac{1}{3}} t^{\frac{1-\omega}{3}} c_1(t) \xrightarrow{t \rightarrow +\infty} 1. \quad (130)$$

Using (130) and the change of variables  $t \mapsto \zeta$  we conclude that

$$Q_0(\omega) \zeta^{r_0} \tilde{c}_1(\zeta) \xrightarrow{\zeta \rightarrow +\infty} 1, \quad (131)$$

and using this result and appropriate estimates for the sum and the integral of (127) we obtain the similarity limits stated in the theorem. ■

What happens if the monomers input rate is slower than the stated in Theorem 19 is studied by Sasportes in [195], where he concludes that if  $\omega = -\frac{1}{2}$  there exists a self-similar profile for the variable  $\eta$ , correspondent to (i) in Theorem 19, and the limit:

$$\lim_{\substack{j, \varsigma \rightarrow +\infty \\ \eta = j/\varsigma \text{ fixed} \\ \eta \neq 1}} (1/2)^{1/3} (3/\alpha)^{2/3} \varsigma (\log \varsigma)^{2/3} \tilde{c}_j(\varsigma) = \Phi_{1,-1/2}(\eta).$$

However, the limit in (ii), for that similarity variable, does not exist.

The kind of self-similar behaviour presented in Theorem 19(i) seems to be valid also when  $a_j = j^p$  with  $p < 1$ . Although a rigorous proof is lacking, non-rigorous formal computations [51] seem to suggest that, for certain functions  $Q_p(\omega)$  and  $A(\omega, p)$ , for  $r_p = \frac{1-\omega(1-p)}{(2+\omega)(1-p)}$ , and for the time scale  $\tau = \left( \frac{(3-2p)A(\omega, p)}{2+\omega} \right)^{\frac{1}{1-p}} t^{\frac{2+\omega}{3-2p}}$ , it could be true that

$$\lim_{\substack{j, \varsigma \rightarrow +\infty \\ \eta = j/\varsigma \text{ fixed} \\ \eta \neq 1}} Q_p(\omega) \varsigma^{r_p} \tilde{c}_j(\varsigma) = \Phi_{1,\omega,p}(\eta) := \begin{cases} \eta^{-p} (1 - \eta^{1-p})^{-r_p}, & \text{if } \eta < 1 \\ 0, & \text{if } \eta > 1. \end{cases}$$

An important feature of many physical systems that is definitely not considered in the simple model (123) is the existence of a critical cluster size below which clusters are very unstable and do not exist. The first rigorous approach to the modelling of this phenomenon in the framework we are considering was proposed recently by Costin and co-workers in [62], where the following system, analogous to (123) but with constant input of monomers, constant reaction rates, and a critical cluster size  $n > 2$ , was considered:

$$\begin{cases} \dot{c}_1 = \alpha - nc_1^n - c_1 \sum_{j=n}^{\infty} c_j \\ \dot{c}_n = c_1^n - c_1 c_n \\ \dot{c}_j = c_1 c_{j-1} - c_1 c_j, \quad j > n. \end{cases} \quad (132)$$

Again, as in (123), the definition of an auxiliary variable  $X(t) := \sum_{j=n}^{\infty} c_j(t)$  allows for the decoupling of (132) into a two-dimensional and an infinite dimensional that can be solved recursively. The qualitative methods used in [52, 57] for the determination of the exact long-time convergence rates of solutions of (123) do not seem to work in this case. However, a careful rigorous asymptotic analysis was possible to implement fully and the following self-similar behaviour was obtained in [62]:

$$\lim_{\substack{j, \varsigma \rightarrow +\infty \\ \eta = j/\varsigma \text{ fixed} \\ \eta \neq 1}} (n/\alpha)^{(n-1)/n} \varsigma^{(n-1)/n} \tilde{c}_j(\varsigma) = \begin{cases} (1 - \eta)^{-(n-1)/n}, & \text{if } 0 < \eta < 1 \\ 0, & \text{if } \eta > 1. \end{cases}$$

Observe that if  $n = 2$  the result of Theorem 19(i) is recovered.

## 4 Density conservation and gelation

More than once in this chapter we referred to problems and results related to the conservation, or non conservation, of the solution density through time evolution. The problem of characterizing the rate coefficients and the initial data for which there is conservation of density, or lack thereof, has been one of the main open problems in the mathematics of coagulation-fragmentation for many decades. Only from the late 1990s was real significant progress made, first by Jeon [110], using probabilistic methods, and afterwards by Escobedo, Mischler and Perthame [81] and by these authors together with Laurençot [80], using purely analytic methods.

Seeing the coagulation-fragmentation equations as a model from chemical kinetics it is all too natural to expect the density of solutions to be a time invariant, due to mass conservation in each elementary reaction. In fact, proceeding in a formal way, if we substitute (4), (14) and (16) into  $\sum_{j=1}^{\infty} j \dot{c}_j$  we obtain  $\sum_{j=1}^{\infty} j \dot{c}_j = 0$ , with an analogous formal result being valid for the continuous version of the equations.

The attempts to turn these formal computations rigorous were faced, from the beginning, with mathematical difficulties, perhaps unexpected, which resulted in that, for many years, the results available in the literature were formal studies (cf. e.g. [77, 83, 106, 227]), rigorous studies of particular cases [26, 27], and examples of solutions that did not conserve density [147, 148]. All these studies were for Smoluchowski's equation with product type coagulation kernels growing fast with the cluster size. We will start by a brief review of the first mathematically rigorous studies.

The first results on the existence of solution to Smoluchowski's coagulation system, proved by McLeod in the beginning of the 1960s [154, 155, 156] considered coagulation coefficients  $a_{j,k} = r_j r_k$  and an initial condition  $c_{0j} = \delta_{j,1}$ . The condition considered in those studies on the finiteness of the solutions' second moment  $\|c(t)\|_2 < \infty$  implied that, when  $r_j = j$ , the maximal interval of existence is  $[0, 1)$ , when  $r_j \leq j$  it contains  $[0, e^{-1}]$ , and when  $r_j = jq_j$ , with  $q_j \rightarrow +\infty$ , there is no solution in any non degenerate time interval. Observe that, when  $a_{j,k} \leq jk$ , the requirement of finite second moment easily implies density conservation of the solution (cf. (68)) and, in fact, waving this condition Leyvraz and Tschudi proved [147] that for  $r_j = j$  McLeod's solution can be continued for  $t > 1$ , using to that end the generating function

$$G(t, z) := \sum_{j=1}^{\infty} \varphi_j(t) z^j, \text{ where } \varphi_j(t) := j c_j(t) e^{j \int_0^t \|c(s)\|_1 ds}.$$

This generating function is a solution of the equation

$$\frac{\partial G}{\partial t} = zG \frac{\partial G}{\partial z}, \quad z \in (0, 1), \quad t > 0,$$

with  $G(0, z) = z$ . As this problem can be integrated by the method of characteristics to obtain an explicit expression for  $G$ , from which we then obtain  $\varphi_j$  and hence  $c_j$ .

The final result is Leyvraz-Tschudi's solution:

$$c_j(t) = \begin{cases} \frac{j^{j-2}}{j!} t^{j-1} e^{-jt}, & \text{if } 0 \leq t \leq 1 \\ \frac{j^{j-2} e^{-j}}{j!} \frac{1}{t}, & \text{if } t > 1, \end{cases} \quad (133)$$

for which an easy computation results in

$$\|c(t)\|_1 = \begin{cases} 1, & \text{if } 0 \leq t \leq 1 \\ t^{-1}, & \text{if } t > 1. \end{cases}$$

Thus, Leyvraz-Tschudi's solution does not conserve density for  $t > 1$ . This same result was later re-derived by Slemrod [200] without recourse to generating functions.

It is very interesting to observe that the nonconservation of density happens at a finite time (in Leyvraz-Tschudi's solution, at  $t = 1$ ) and not at the long-time limit,  $t \rightarrow +\infty$ , as in the Becker-Döring and the coagulation-fragmentation equations with weak fragmentation. The physical interpretation is analogous: the missing density  $\|c(0)\|_1 - \|c(t)\|_1$  corresponds to the runaway of part of the density to entities whose sizes are not described by subscripts  $j \in \mathbf{N}$  (or  $x \in \mathbf{R}^+$  in the continuous version), which means that they are, from the physical point of view, incommensurably larger than every  $j$ . This “infinite cluster” is interpreted in the physics literature as a different macroscopic phase, called a *gel*, and its occurrence is called the sol-gel transition or gelification. The smallest time  $T_g \geq 0$  after which density conservation no longer holds is called the geling time<sup>9</sup>.

The above interpretation for the finite time break down of density conservation also suggests that such a phenomenon occurs when the coagulation coefficients grow fast with the cluster sizes. In fact, Leyvraz showed in [148] that, if  $r_j = j^\alpha$ , with  $\alpha > \frac{1}{2}$ , then there exists a solution of (34) with a specially chosen initial condition, for which  $T_g = 0$ . This result, as well as those above are examples of non density conserving solutions with very particular initial data; nevertheless they are historically important because for many years they were essentially the only rigorous examples known of solutions exhibiting a behaviour that was generally believed to hold for all non zero solutions to (34) when  $a_{j,k} \geq (jk)^\alpha$  with  $\alpha > \frac{1}{2}$ .

The case  $\alpha > 1$  was first studied by van Dongen [69] and rigorously proved by Carr and da Costa [35]:

**Theorem 20** [35] *Let  $C_L(j^\alpha + k^\alpha) \leq a_{j,k} \leq C_U(jk)^\beta$ , with constants  $C_L, C_U > 0$  and  $\beta > \alpha > 1$ . Let  $c$  be a solution of (34) in  $[0, T)$  with  $c_0 \neq 0$ . Then,  $c$  does not conserve density in any time interval  $[0, t_\infty)$ ,  $\forall t_\infty \leq T$ .*

*Sketch of proof:* The basic idea of the proof is an argument by contradiction using higher moments: assuming that  $c$  is a density conserving solution in an interval

<sup>9</sup> There is an analogous behaviour of loss of mass in the pure fragmentation system, called “shattering” and interpreted as a loss of mass to infinitesimal clusters. We shall not consider this behaviour here, directing the interested reader to the literature, e.g., [7, 89, 152].

$[0, t_\infty)$  and using the lower bound for the coefficients it is possible to prove that, for all  $p > 1$ ,

$$\sum_{j=m}^{\infty} j^p c_j(t) \leq \|c_0\|_1 \sum_{j=m}^{\infty} j^{p-1} e^{-C_L \|c_0\|_1 j^{1-\alpha}(\varepsilon-t)/2}, \quad (134)$$

where  $0 < t < \varepsilon < t_\infty$  and  $m$  is sufficiently large.

Obviously (134) implies that all moments  $\|c(t)\|_p$  are finite in  $(0, t_\infty)$  and this is the result that is at the centre of the contradiction because the lower bound on the coefficients, the hypothesis of density conservation, and Hölder's inequality imply that,  $\forall \delta, t, \tau \in (0, t_\infty)$  with  $\delta < t \leq \tau$ ,

$$\|c(t)\|_p - \|c(\delta)\|_p \geq p C_L \|c_0\|^{1-\frac{\alpha-1}{p-1}} \int_{\delta}^t \|c(s)\|_p^{1+\frac{\alpha-1}{p-1}} ds,$$

from which we get a blowing up time for  $\|c(t)\|_p$ ,  $T^{(p)}$ , with  $\lim_{p \rightarrow +\infty} T^{(p)} \leq \delta$ . ■

For many decades the only rigorous results for the case  $\alpha \in (\frac{1}{2}, 1]$  were the particular solutions in [147, 148], already referred to. An attempt by da Costa [49] to prove that the same behaviour would occur for all solutions, based in a dynamical systems approach, resulted in the identification of a larger family of gelling solutions but did not solve the problem, although it had some use in the numerical analysis of the gelling phenomenon [8]. For the continuous system Laurençot [125] considered  $a(x, y) = r(x)r(y) + \alpha(x, y)$  with  $\alpha(x, y) \leq Ar(x)r(y)$  and  $r(x) \geq Rx$ , and proved that all solution exhibit gelation and obtained some results about the density decay and the gelification time.

In other works fragmentation was also included. Recall that, in [46] it was proved that, with coagulation coefficients for which gelation was expected to occur, a sufficiently strong fragmentation prevents that to happen at least for solutions obtained as limits of truncated systems (cf. Theorem 8). This is also in line with the interpretation of gelation as a loss of density to an infinite size entity, because it is natural to expect that a high rate of fragmentation of big clusters inhibits the accumulation of density in larger and larger clusters, thus preventing the runaway phenomenon causing the emergence of gelation.

The rigorous analytic elucidation of gelation was achieved in 2002 by Escobedo, Mischler and Perthame in [81], and in 2003 by the same authors with Laurençot in [80]. In what follows we briefly describe those results.

Let us start by observing that the results in [80, 81] are proved for the continuous version of the coagulation-fragmentation equations but, naturally, they are also valid for the discrete case. We shall concentrate our attention in the coagulation equation:

**Theorem 21** [81] *Let  $a(x, y) = \frac{1}{2} (x^\alpha y^\beta + x^\beta y^\alpha)$ , with  $0 \leq \alpha \leq \beta \leq 1$  and  $\lambda := \alpha + \beta > 1$ . Let  $c$  be an arbitrary weak solution of (5)-(6) with a non zero initial condition<sup>10</sup>  $c_0 \in Y_1$ . Then, there exists a positive constant  $C_* = C_*(M_1(0), M_0(0), \lambda)$  such that, for all  $t \geq 0$ , it holds*

<sup>10</sup> The Banach space  $Y_1$  was defined in page 24.

$$M_1(t) \leq \frac{C_*}{(1+t)^{1/\lambda}} \quad (135)$$

and so the gelling time is finite and satisfies the upper bound

$$T_g \leq T_* := \left( \frac{C_*}{M_1(0)} \right)^\lambda. \quad (136)$$

In this statement the notation  $M_k(t) := \|c(t, \cdot)\|_{L^1(\mathbf{R}^+, y^k dy)}$  was used.

*Sketch of proof:* The proof is based on some estimates for the weak solutions using carefully constructed test functions, from which it is possible to conclude that, for all  $\tau \geq 0$ ,

$$\int_\tau^\infty M_1(t)^2 dt \leq C_\lambda M_0(0)^{\lambda-1} M_1(\tau)^{2-\lambda}, \quad (137)$$

from which we obviously see that the density cannot be constant and, with a bit more extra work, obtain (135). Let us look at this argument in more detail. The definition of weak solution of (5)-(6) is like the one in the discrete case: it is any function  $c \in C([0, \infty); L^1) \cap L^\infty(0, T; Y_1)$ ,  $\forall T > 0$ , satisfying  $M_1(t) \leq M_1(0)$ ,  $\forall t \geq 0$ , such that, for all  $t \geq \tau \geq 0$  and  $g \in L^\infty(0, \infty)$ , the following holds

$$\begin{aligned} & \int_0^\infty g(x)c(t, x)dx - \int_0^\infty g(x)c(\tau, x)dx = \\ &= \frac{1}{2} \int_\tau^t \int \int_{\mathbf{R}^+ \times \mathbf{R}^+} (g(x+y) - g(x) - g(y))a(x, y)c(s, x)c(s, y)dx dy ds. \end{aligned} \quad (138)$$

Let  $c$  be a weak solution of (5)-(6) and in (138) consider the test function  $g(x) = g_A(x) := x \wedge A \in L^\infty(0, \infty)$ . As  $g_A(x+y) - g_A(x) - g_A(y) \leq 0$  in  $\mathbf{R}^+ \times \mathbf{R}^+$  it is possible to estimate the right-hand side of (138) keeping only the contribution due to the integration on  $[A, \infty)^2$ , which immediately results in

$$\int_\tau^t \left( \int_A^\infty y^{\lambda/2} c(s, x) dx \right)^2 ds \leq \frac{2M_1(\tau)}{A}. \quad (139)$$

Consider now a function  $\Phi : [0, \infty) \rightarrow [0, \infty)$  which is monotonic increasing, differentiable a.e., with  $\Phi(0) = 0$ , and  $C_\Phi := \|\Phi'\|_{L^1(\mathbf{R}^+, y^{-1/2} dy)} < \infty$ . Writing  $\Phi(x) = \int_0^x \Phi'(A) dA$ , using Fubini's theorem, Cauchy-Schwarz inequality and (139) we concluded that

$$\int_\tau^t \left( \int_0^\infty x^{\lambda/2} \Phi(x) c(s, x) dx \right)^2 ds \leq 2C_\Phi^2 M_1(\tau).$$

Taking limits as  $t \rightarrow \infty$  and considering  $\Phi(x) := (x^{1-\lambda/2} - (R/2)^{1-\lambda/2})^+$ , where  $R > 0$  is an arbitrary constant, we conclude that

$$\int_\tau^\infty \left( \int_R^\infty xc(s, x) dx \right)^2 ds \leq CR^{1-\lambda} M_1(\tau). \quad (140)$$



Finally, (137) is obtained by using  $M_1(t)^2 \leq 2\left(\int_0^R xc(t,x)dx\right)^2 + 2\left(\int_R^\infty xc(t,x)dx\right)^2$ , and  $\left(\int_0^R xc(t,x)dx\right)^2 \leq R^{2-\lambda} M_{\lambda/2}(\tau)^2$ , applying (140) and (138) with the test function  $g \equiv 1$ , and taking  $R = M_1(\tau)/M_0(\tau)$ . ■

Reference [81] also contains an extensive study of several properties of the density of weak solutions of (5)-(6), including the behaviour of the solutions at gelling time  $T_g$ .

The same method was used in [80, 81] for the continuous system with fragmentation, establishing the following result:

**Theorem 22** [80, 81] *Let  $a(x,y) = \frac{1}{2}(x^\alpha y^\beta + x^\beta y^\alpha)$ , with  $0 \leq \alpha \leq \beta \leq 1$  and  $\lambda := \alpha + \beta$ . Let  $b(x,y) = (1+x+y)^\gamma$ , with  $\gamma \in \mathbf{R}$ . Then, the following hold:*

- (i) *if  $\lambda \leq 1$  or if  $\gamma > \lambda - 2$ , there exists a density conserving weak solution (17)*
- (ii) *if  $\lambda > 1$  and  $\gamma < \lambda - 2$ , there exists a critical density  $\rho^* > 0$  such that, when  $c_0 \in Y_1$  satisfies  $\|c_0\|_{L^1(\mathbf{R}^+, ydy)} > \rho^*$ , every weak solution of (17) with initial condition  $c_0$  exhibits gelation.*

Note that when the condition on  $\gamma$  is (i) the behaviour of solutions is the same that occurred for the discrete system in the strong fragmentation case (cf. page 33), that is: even with conditions on the coagulation coefficients for which solutions to the purely coagulating dynamics have break down of density conservation, a sufficiently strong fragmentation forces density to remain constant through time evolution.

Case (ii) leaves unanswered what happens for sufficiently small densities. Formal arguments presented in [80] lead to the conjecture that, if  $\gamma \in ((\lambda - 3)/2, \lambda - 2)$ , there are weak solutions, with low density initial conditions, for which density is conserved, whereas if  $\gamma < (\lambda - 3)/2$ , all non zero solutions have gelation.

As stated above, Theorems 21 and 22 are fundamental contributions to the problem of density conservation in coagulation-fragmentation systems, although, as stated in [80, 81], several relevant problems still wait for a proof.

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