Stochastic, analytic and numerical aspects of coagulation processes

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Abstract

In this paper we review recent results concerning stochastic models for coagulation processes and their relationship to deterministic equations. Open problems related to the gelation effect are discussed. Finally we present some new conjectures based on numerical experiments performed with stochastic algorithms.

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1. Introduction

The phenomenon of coagulation occurs in a wide range of applications, e.g., in physics (aggregation of colloidal particles, growth of gas bubbles), meteorology (merging of drops in atmospheric clouds, aerosol transport), chemistry (reacting polymers, soot formation) and astrophysics (formation of stars and planets). We refer to the survey papers [8] and [2] for more details. The time evolution of the average concentration of particles of a given size in some spatially homogeneous physical system is described by Smoluchowski's coagulation equation [36]

\[ \frac{\partial}{\partial t} c(t, x) = \]

\[ \frac{1}{2} \sum_{y=1}^{x-1} K(x - y, y) c(t, x - y) c(t, y) - \sum_{y=1}^{\infty} K(x, y) c(t, x) c(t, y), \]

where \( t \geq 0 \) and \( x = 1, 2, \ldots \). Concentration of particles of size \( x \) increases as a result of coagulation of particles of sizes \( x - y \) and \( y \). It decreases if particles of size

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$x$ merge with any other particles. The intensity of the process is governed by the (non-negative and symmetric) coagulation kernel $K$ representing properties of the physical medium.

The common stochastic model related to the coagulation equation (1) is a Markov jump process where two different clusters of size $x$ and $y$ merge into a single cluster of size $x + y$ with rate $K(x, y)$ (cf. [28], [15], [26]). The basic relationship between the stochastic model and the deterministic equation is given by the law of large numbers. Qualitative properties of the coagulation equation and its generalizations have been successfully studied using the stochastic approach. We refer to [20], [17], [21], [30], [9], [31], [11] concerning recent results, and especially to [2] as an excellent review. Stochastic particle systems play also an important role in the numerical treatment of equation (1). Many stochastic algorithms are based on the classical direct simulation process (cf. [16], [7], [34], [14], [18], [19], [10]). Various numerical methods for the coagulation equation are reviewed in [33], where also an extended list of references is given. Some stochastic algorithms (cf. [25], [24], [32], [23], [3]) contain an additional approximation parameter (time step), thus providing solutions to time discretized versions of equation (1).

The purpose of this paper is to review recent results concerning stochastic models for coagulation processes and their relationship to deterministic equations. We also discuss open problems related to the gelation phenomenon. Finally we present some new conjectures based on numerical experiments performed with stochastic algorithms.

2. Stochastic models and approximation

We start from considering the continuous coagulation-fragmentation equation

$$\frac{\partial}{\partial t} c(t, x) = \frac{1}{2} \int_0^\infty K(x - y, y) c(t, x - y) c(t, y) \, dy - \int_0^\infty K(x, y) c(t, x) c(t, y) \, dy$$

$$+ \int_0^\infty f(x, y) c(t, x + y) \, dy - \frac{1}{2} \int_0^\infty f(x, y) c(t, x) \, dy,$$

with initial condition $c(0, x) = \rho(x) \geq 0$, which describes the time evolution of the average concentration of particles of size $x > 0$. Here the non-negative and symmetric function $f(x, y)$ denotes the fragmentation rate of an $(x + y)$-cluster into clusters of size $x$ and $y$. In addition to the effect of coagulation, the concentration $c(x, t)$ increases by fragmentation of an $(x + y)$-cluster into clusters of size $x$ and $y$ (third term) and decreases by fragmentation of an $x$-cluster into clusters of size $y < x$ and $x - y$ (fourth term). The combined coagulation-fragmentation equation appeared in [29].

Multiplication with some test function $\varphi$ and integration with respect to the size
variable $x$ transform the right-hand side of equation (2) into

$$
\int_0^\infty \int_0^\infty \left[ \frac{1}{2} \varphi(x + y) - \varphi(x) \right] K(x, y) c(t, x) c(t, y) \, dy \, dx \\
+ \int_0^\infty \int_0^\infty \left[ \varphi(y) - \frac{1}{2} \varphi(x) \right] f(x - y, y) c(t, x) \, dy \, dx.
$$

(3)

Here the identity $\int_0^\infty \int_0^\infty \psi(x, y) \, dy \, dx = \int_0^\infty \int_0^\infty \psi(x - y, y) \, dy \, dx$ has been used. Having in mind (3), we introduce the weak version of the coagulation-fragmentation equation (2) (cf. [30], [11])

$$
\frac{d}{dt} \int_Z \varphi(x) \, P(t, dx) = \int_Z \int_Z \left[ \frac{1}{2} \varphi(x + y) - \varphi(x) \right] K(x, y) P(t, dy) \, P(t, dx) \\
+ \int_Z \int_Z \left[ \varphi(y) - \frac{1}{2} \varphi(x) \right] F(x, dy) \, P(t, dx).
$$

(4)

The size space is either $Z = (0, \infty)$, corresponding to equation (2), or $Z = \{1, 2, \ldots\}$, corresponding to equation (1). A continuous measure-valued function $P \in C([0, \infty), \mathcal{M}(Z))$ is called a solution if it satisfies equation (4), for all continuous function with compact support $\varphi \in C_c(Z)$, and

$$
\int_0^t \left[ \int_Z \int_Z K(x, y) P(s, dx) \, P(s, dy) + \int_Z F(x, Z) \, P(s, dx) \right] \, ds < \infty,
$$

(5)

for all $t \geq 0$.

The fragmentation measure $F$ is concentrated on $\{y \in Z : y < x\}$ and satisfies the symmetry assumption

$$
\int_0^x \varphi(y) \, F(x, dy) = \int_0^x \varphi(x - y) \, F(x, dy), \quad \forall x \in Z,
$$

(6)

for measurable bounded functions $\varphi$ on $(0, x)$. Note that (6), with $\varphi(x) = x$, implies

$$
\int_0^x y \, F(x, dy) = \frac{x^2}{2} F(x, Z).
$$

(7)

In particular, the choice

$$
F(x, dy) = \chi_{[0,x]}(y) f(x - y, y) \, dy,
$$

where $\chi_A$ denotes the indicator function of a set $A$, corresponds to expression (3).

The solution $P(t, dx)$ of equation (4) represents the flow of concentration in the size space $Z$. The total mass of the system is determined as $\int_Z x \, P(t, dx)$. We call the function

$$
\bar{P}(t, dx) = x \, P(t, dx), \quad t \geq 0,
$$

(8)

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the mass flow and consider the mass flow equation

$$\frac{d}{dt} \int_Z \varphi(x) \tilde{P}(t, dx) = \int_Z \int_Z \left[ \varphi(x + y) - \varphi(x) \right] \frac{K(x, y)}{y} \tilde{P}(t, dx) \tilde{P}(t, dy) \, ds$$

$$+ \int_Z \int_Z \left[ \varphi(y) - \varphi(x) \right] \frac{y}{x} F(x, dy) \tilde{P}(t, dx) . \quad (9)$$

A function $\tilde{P} \in C([0, \infty), M(Z))$ is called a solution if it satisfies (9) for all $\varphi \in C_c(Z)$ and

$$\int_0^t \left[ \int_Z \int_Z \frac{K(x, y)}{x y} \tilde{P}(s, dx) \tilde{P}(s, dy) + \int_Z \frac{F(x, Z)}{x} \tilde{P}(s, dx) \right] \, ds < \infty , \quad (10)$$

for all $t \geq 0$. Equation (9) was studied in [12] for the pure coagulation case. The discrete version of equation (9) has been used in [3] for constructing a discretized in time stochastic algorithm for equation (1).

If some measure-valued functions $P$ and $\tilde{P}$ satisfy (8), then $\tilde{P}$ is a solution of the mass flow equation (9) if and only if $P$ is a solution of the coagulation equation (4). Indeed, $\tilde{P} \in C([0, \infty), M(Z))$ iff $P \in C([0, \infty), M(Z))$ (vague topology on $M(Z)$), and (5) is satisfied iff (10) is satisfied. Moreover, using symmetry, one obtains

$$\int_Z \int_Z \left[ \frac{1}{2} \varphi(x + y) (x + y) - \varphi(x) x \right] K(x, y) P(t, dx) P(t, dy) =$$

$$\int_Z \int_Z \left[ \varphi(x + y) x - \varphi(x) x \right] \frac{K(x, y)}{x y} \tilde{P}(t, dx) \tilde{P}(t, dy)$$

$$= \int_Z \int_Z \left[ \varphi(x + y) - \varphi(x) \right] \frac{K(x, y)}{y} \tilde{P}(t, dx) \tilde{P}(t, dy) ,$$

for any $\varphi \in C_c(Z)$. Finally, using (7), one obtains

$$\int_Z \int_Z \left[ y \varphi(y) - \frac{1}{2} x \varphi(x) \right] F(x, dy) P(t, dx) =$$

$$\int_Z \int_Z \left[ \frac{y}{x} \varphi(y) - \frac{1}{2} \varphi(x) \right] F(x, dy) P(t, dx)$$

$$= \int_Z \int_Z \left[ \varphi(y) - \varphi(x) \right] \frac{y}{x} F(x, dy) \tilde{P}(t, dx) ,$$

and the equivalence follows.

We want to approximate the solution of equation (4) by a sequence of particle systems. For every $n \in \mathbb{N}$, define

$$\mathbb{S}^n = \left\{ p = \frac{1}{n} \sum_{i=1}^N \delta_{x_i}, \quad x_i \in \mathbb{Z}, \quad N = 1, 2, \ldots \right\} ,$$

and consider an $\mathbb{S}^n$-valued jump process $U^{(n)}(t)$ with generator $K + F$, where

$$K \Phi(p) = \frac{1}{2n} \sum_{1 \leq i, j \leq N} \left[ \Phi(J_K(p, i, j)) - \Phi(p) \right] K(x_i, x_j) , \quad (11)$$
with
\[ J_K(p, i, j) = p + \frac{1}{n}(\delta_{x_i + x_j} - \delta_{x_i} - \delta_{x_j}), \] (12)

is the coagulation operator, and
\[
\mathbb{F} \Phi(p) = \frac{1}{2} \sum_{i=1}^{N} \int_{\mathbb{Z}} \left[ \Phi(J_F(p, i, y)) - \Phi(p) \right] F(x_i, dy),
\]

with
\[
J_F(p, i, y) = p + \frac{1}{n}(\delta_y + \delta_{x_i - y} - \delta_{x_i}),
\]
is the fragmentation operator. For
\[
\Phi(p) = \int_{\mathbb{Z}} \varphi(z) p(dz), \quad p \in \mathbb{S}^n, \quad \varphi \in C_c(\mathbb{Z}),
\]
one obtains
\[
\int_{\mathbb{Z}} \varphi(z) U^n(t, dz) =
\int_{\mathbb{Z}} \varphi(z) U^n(0, dz) + \int_0^t \mathbb{[K} \Phi + \mathbb{F} \Phi](U^n(s)) \, ds + M^\varphi(t),
\]
where \( M^\varphi \) is some martingale term. The representations
\[
\mathbb{K} \Phi(p) = \int_{\mathbb{Z}} \int_{\mathbb{Z}} K(x, y) \left[ \frac{1}{2} \varphi(x + y) - \varphi(x) \right] p(dx) \, p(dy)
\]
and (cf. (6))
\[
\mathbb{F} \Phi(p) = \int_{\mathbb{Z}} \int_{\mathbb{Z}} \left[ \varphi(y) - \frac{1}{2} \varphi(x) \right] F(x, dy) \, p(dx)
\]
indicate the connection between (14) and equation (4), suggesting the property
\[
\lim_{n \to \infty} U^{[n]}(t) = P(t).
\]
Rigorous results concerning this transition can be found, e.g. in [20], [30], [11]. Note that the proofs of these results provide existence theorems for the deterministic limiting equation as a by-product.

Next we construct a particle approximation to the solution of the mass flow equation (9), which, due to the equivalence of the equations, provides an alternative approximation to the solution of equation (4). We introduce the generator \( \mathbb{K} + \mathbb{F} \), where
\[
\mathbb{K} \Phi(p) = \frac{1}{n} \sum_{1 \leq i, j \leq N} \left[ \Phi(J_K(p, i, j)) - \Phi(p) \right] \frac{K(x_i, x_j)}{x_j},
\]
(15)
with
\[
\tilde{J}_K(p, i, j) = p + \frac{1}{n} (\delta_{x_i + x_j} - \delta_{x_i}),
\]
(16)
is the modified coagulation operator, and
\[
\tilde{\Phi}(p) = \frac{1}{y} \int_{x_i} \left[ \Phi(J_F(p, i, y)) - \Phi(p) \right] F(x, dy),
\]
with
\[
J_F(p, i, y) = p + \frac{1}{n} (\delta_y - \delta_{x_i}),
\]
is the modified fragmentation operator. Taking into account the martingale representation (14), and considering test functions (13), one obtains the expressions
\[
\tilde{K}(p) = \int \int \frac{K(x, y)}{y} p(dx) p(dy)
\]
and
\[
\tilde{\Phi}(p) = \int \int \frac{\varphi(y) - \varphi(x)}{y} F(x, dy) p(dx)
\]
indicating the connection with equation (9). A rigorous transition has been carried out in [12] for the pure coagulation case.

Note the following intuitive derivation of the approximating particle system for the mass flow equation. Consider a system of independent particles determined by the generator
\[
Q(p) = \sum_{i=1}^{n} \int \left[ \Phi(J_Q(p, i, y)) - \Phi(p) \right] Q(x, dy),
\]
(17)
with
\[
J_Q(p, i, y) = p + \frac{1}{n} (\delta_{x_i + y} - \delta_{x_i}),
\]
where \( Q \) denotes some jump measure. Obviously, the corresponding limiting equation is
\[
\frac{d}{dt} \int_{x_i} \varphi(x) P(t, dx) = \int \int \left[ \varphi(x + y) - \varphi(x) \right] Q(x, dy) P(t, dx).
\]
(18)
Note that the mass flow equation (9) has the form (18) with appropriately chosen \( Q \). In particular, the coagulation term corresponds to
\[
Q(x, dy) = \frac{K(x, y)}{y} P(t, dy).
\]
After replacing formally \( \tilde{P} \) by the empirical measure of the particle system, the generator (17) takes the form (15). This connection suggests how to construct a non-linear Markov process related to the mass flow equation (9). Such a process is determined by a stochastic equation with coefficients depending on the law of the solution. This approach has been carried out in [5], [6] for the pure coagulation case, and in [22] for the case including fragmentation.
3. Numerical algorithms

Note that a basic element of the convergence proofs mentioned in the previous section is uniqueness of the solution to the limiting equation. However this uniqueness has been established [30] only up to the gelation point

$$t_{gel} = \inf \left\{ t \geq 0 : m_1(t) < m_1(0) \right\}, \quad \text{where} \quad m_1(t) = \sum_{x=1}^{\infty} x c(t, x),$$

(19)

which is finite for sufficiently fast increasing coagulation kernels. Thus the problem of convergence after that point is open for general kernels, and numerical observations concerning the behaviour of the stochastic processes are of special interest.

In this section we discuss the problem how to determine the time evolution of the mass $m_1$ (cf. (19)) for gelling kernels, using stochastic algorithms. Here we restrict our considerations to the case of pure coagulation.

Kernels satisfying

$$K(Cx, Cy) = C^\varepsilon K(x, y), \quad \forall C, x, y > 0,$$

(20)

are called homogeneous, with exponent of homogeneity $\varepsilon$. The following properties have been proved in particular cases and are widely expected to be true in general (cf. [20], [21], and the recent paper [13]). If $\varepsilon \leq 1$ then there is no gelation, while $\varepsilon > 1$ implies gelation, i.e. $t_{gel} < \infty$. Moreover, if $\varepsilon > 2$ then there is instantaneous gelation, i.e. $t_{gel} = 0$. The special case of the multiplicative kernel

$$K(x, y) = x y$$

(21)

has been intensively studied in the framework of random graph theory (see [2, Section 4.4]). In particular, it has been proved in [4] that the standard stochastic model (cf. (11), (12)) converges to a deterministic limit, which coincides with the solution of the Smoluchowski equation (1) before the gelation point, but is different from the solution after that point. Moreover, it has been established that the asymptotic behaviour of the maximal component $M^{(n)}_1(t)$ in the system changes qualitatively at the gelation point, namely it becomes of order $n$ after $t_{gel}$ while it is of lower order before that point. Thus, for the kernel (21), one obtains

$$\lim_{n \to \infty} \frac{M^{(n)}_1(t)}{n} > 0, \quad \forall t > t_{gel},$$

(22)

but the property (cf. (19))

$$m_1(t) = 1 - \lim_{n \to \infty} \frac{M^{(n)}_1(t)}{n}, \quad \forall t \geq 0,$$

(23)

does not hold.

The common stochastic algorithm for the coagulation equation (4) is based on the process with the generator (11), (12). Due to its physical interpretation, we call
this simulation procedure **direct simulation algorithm**. Note that, if the current state of the process is represented by particles \(x_1(t), \ldots, x_{N(t)}(t)\), then functionals are approximated by

\[
\int_{\mathbb{R}} \varphi(x) P(t, dx) \sim \frac{1}{n} \sum_{i=1}^{N(t)} \varphi(x_i(t)), \quad t \geq 0, \tag{24}
\]

for some test function \(\varphi\). Obviously, this algorithm is mass-preserving. However, property (22) suggests to study the gelation problem for general coagulation kernels using the maximal component in the system, namely measuring the quantity

\[
\frac{M_{i}^{(n)}(t)}{n}, \quad t \geq 0, \tag{25}
\]

for different values of \(n\).

The simulation procedure based on the process with the generator (15), (16) is called **mass flow algorithm**. If the current state of the process is represented by particles \(\tilde{x}_1(t), \ldots, \tilde{x}_{\tilde{N}(t)}(t)\), then functionals of the solution are approximated by (contrast this with (24))

\[
\int_{\mathbb{R}} \varphi(x) P(t, dx) \sim \frac{1}{n} \sum_{i=1}^{\tilde{N}(t)} \varphi(\tilde{x}_i(t)) \tilde{x}_i(t), \quad t \geq 0. \tag{26}
\]

An interesting aspect of the mass flow model is the emergence of infinite clusters in finite time for gelling kernels. It has been conjectured in [12] that the (random) explosion times \(\tau_{\infty}^n\) of the stochastic system converge (as \(n \to \infty\)) to the gelation time \(t_{gel}\). This would connect the gelation effect (a property of the limiting equation) with the explosion phenomenon of a stochastic process, thus representing the physical interpretation of gelation. In this respect we refer to recently announced results [27] concerning explosion behaviour of some appropriately scaled tagged particle in the direct simulation process.

In order to be able to perform calculations beyond the gelation point, we introduce some truncation parameter \(b_n\) such that particles with size bigger than \(b_n\) are removed from the system. This corresponds to a modification of the coagulation jump (16) by

\[
p - \frac{1}{n} \delta_{x_j} + \chi(a_{i,b_n})(x_i + x_j) \frac{1}{n} \delta_{x_i+x_j}.
\]

Since the truncated mass flow algorithm is not mass-preserving, (26) suggests a natural approximation of the mass, namely

\[
\frac{\tilde{N}^{(n)}(t)}{n}, \quad t \geq 0. \tag{27}
\]

It has been conjectured in [12] that

\[
m_i(t) = \lim_{n \to \infty} \frac{\tilde{N}^{(n)}(t)}{n}, \quad \forall t \geq 0.
\]
The numerical algorithms consist in generating trajectories of the corresponding process and calculating estimators (25) and (27), respectively. The results in the figures below are averaged over ten independent runs. Both algorithms are initialized according to the monodisperse initial condition

$$a_0(1) = 1, \quad a_0(x) = 0, \quad x = 2, 3, \ldots$$

For an efficient generation of trajectories the coagulation kernel is replaced by some majorant kernel, and fictitious jumps are introduced. This leads to an easy calculation of the time step and to an independent generation of the collision partners. For more details of this common numerical approach we refer to [10].

As test examples, we consider the two classes of kernels

$$K^{(1)}_{\alpha,\beta}(x, y) = \frac{1}{2} \left[ x^\alpha y^\beta + x^\beta y^\alpha \right], \quad 0 \leq \beta \leq \alpha \leq 1, \quad (28)$$

and [37]

$$K^{(2)}_{\gamma}(x, y) = \frac{2(x y)^\gamma}{(x + y)^{\gamma} - x^\gamma - y^\gamma}, \quad 1 < \gamma \leq 2. \quad (29)$$

The exponents of homogeneity (cf. (20)) are $\varepsilon = \alpha + \beta$ for the kernel (28) and $\varepsilon = \gamma$ for the kernel (29). Note that the multiplicative kernel (21) is contained in both classes (28) ($\alpha = \beta = 1$) and (29) ($\gamma = 2$). For kernels (29) it has been proved in [1] that the moment of order $\gamma$ explodes at $t = 1$, “strongly suggesting” $t_{\varepsilon=1} = 1$ for all $\gamma$.

Spouge’s conjecture [34] says that for kernels (28) with $\alpha = \beta \in (0, 1)$ the mass of the gel is approximated by the maximal cluster, i.e., (23) and, in particular, (22) hold. On the contrary, there is Aldous’ conjecture [2, Section 5.2] that $M^{(r)}_i(t) = o(n)$ after the gelation point, i.e., (22) does not hold, for homogeneous kernels (20) with exponent $1 < \varepsilon < 2$.

Numerical experiments in [10], [12] have indicated that for the kernel (29) property (22) holds, but (23) does not. On the other hand, for the kernel $(x y)^\alpha, \alpha < 1$, numerical observations did not provide any suggestion concerning the asymptotic behaviour. Similar slow convergence has been observed in [19] for the case $\alpha > 1$, related to instantaneous gelation [21].

An additional parameter characterizing growth properties of the coagulation kernel was used in [35], namely a coefficient $\kappa$ such that

$$\lim \frac{1}{y^{\varepsilon - \kappa}} \lim_{x \to \infty} \frac{K(x, y)}{x^\kappa} \in (0, \infty), \quad \varepsilon / 2 \leq \kappa \leq \varepsilon.$$ 

An easy computation implies

$$\lim_{x \to \infty} \frac{K^{(2)}_{\gamma}(x, y)}{x} = \frac{1}{\gamma} y^{\gamma - 1}$$

so that $\kappa = 1$ for the kernel (29). Note that $\kappa = \alpha$ for the kernel (28). The numerical observations mentioned above indicate that the cases $\kappa = 1$ and $\kappa < 1$
are qualitatively different, when \( \varepsilon \in (1, 2) \). We conjecture that in the case \( \kappa = 1 \) property (22) holds, but (23) does not, in analogy with the case of the multiplicative kernel.

This conjecture is supported by numerical results for the kernel (28) with \( \alpha = 1 \) and \( \beta = 0.5 \) displayed in Figure 1. On the left-hand side curves for the estimator (25) are given for \( n = 10^3, 10^6, 10^7 \). On the right-hand side curves for the estimator (27) are given for \( n = 10^3, 10^4, 10^5 \) and \( b_n = 100n \).

![Figure 1: Estimators \( \frac{M^{(n)(u)}}{n} \) (left) and \( \frac{\hat{N}^{(n)(u)}}{n} \) (right) for kernel \( \frac{1}{2} [x y^{0.5} + x^{0.5} y] \)](image)

Next we compare the kernel (cf. (28))

\[
\frac{1}{2\beta - 1} \ K^{(1)}_{1+\beta}(x, y) = \frac{1}{2(2\beta - 1)} \ [x y^\beta + x^\beta y]
\]

(30)

and the kernel \( K^{(2)}_{1+\beta} \) (cf. (29)). Note that for these kernels both \( \varepsilon \) and \( \kappa \), as well as the value at \( x = y = 1 \), are the same. Numerical results are displayed in Figure 2 for \( \beta = 0.5 \) (left) and \( \beta = 0.8 \) (right). They were obtained using the estimator (27) with \( n = 10^3 \) and \( b_n = 10n \). The dashed lines correspond to the kernel (30), and the solid lines to the corresponding kernel (29). The curves for both kernels are rather similar to each other. In particular, using a scaling argument and \( t_{\text{gel}} = 1 \) for kernel (29), the quantity \( 1/(2\beta - 1) \) provides a surprisingly good approximation for the gelation time corresponding to kernel (28) with \( \alpha = 1 \) and different values of \( \beta \).

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Figure 2: Estimator \( \hat{K}^{(n)}(t, \beta) \) for kernels \( \frac{1}{2^\beta - 1} K_{1,\beta}^{(1)} \) (dashed) and \( K_{1,\beta}^{(2)} \) (solid) with \( \beta = 0.5 \) (left) and \( \beta = 0.8 \) (right).

References


