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# Quasi-Monte Carlo simulation of coagulation-fragmentation

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**Abstract:** We extend a quasi-Monte Carlo scheme designed for coagulation to the simulation of the coagulation-fragmentation equation. A number $N$ of particles is used to approximate the mass distribution. After time discretization, three-dimensional quasi-random points decide at every time step whether the particles are undergoing coagulation or fragmentation. We prove that the scheme converges as the time step is small and $N$ is large. In a numerical test, we show that the computed solutions are in good agreement with the exact ones, and that the error of the algorithm is smaller than the error of a corresponding Monte Carlo scheme using the same discretization parameters.

**Keywords:** Quasi-Monte Carlo method, coagulation equation, fragmentation equation, stochastic particle method, low-discrepancy sequence

# 1 Introduction

Coagulation-fragmentation models have applications in many domains of science and technology: aerosol dynamics, polymerization, and combustion processes, for example [1]. M. von Smoluchowski modelled the coagulation of particles in a medium where only binary collisions occur: the numbers of particles of different sizes obey an infinite system of ordinary differential equations [16]. H. Müller rewrote the equations in terms of an integro-differential equation for the time evolution of the particle size density function [13]. S. K. Friedlander considered the result of binary collisions occuring simultaneously with splitting of single particles in two new particles [5]. This gives the coagulation-fragmentation equation for the density $c(x,t)$ of particles of size (or mass) $x$ at times $t$ (see [3]):

$$\begin{aligned}\frac{\partial c}{\partial t}(x,t) &= \frac{1}{2}\int_0^x K(x-y,y)c(x-y,t)c(y,t)dy \\ &\quad -\int_0^{+\infty} K(x,y)c(x,t)c(y,t)dy + \int_0^{+\infty} F(x,y)c(x+y,t)dy \\ &\quad -\frac{1}{2}\int_0^x F(x-y,y)c(x,t)dy, \quad x>0,\ t>0, \end{aligned} \tag{1}$$

with the initial condition $c(x,0) = c_0(x)$ $(x > 0)$, which is a given non-negative function. The coagulation kernel $K(x,y)$ describes the rate of formation of a particle of size $x+y$ by coagulation of two particles of size $x$ and $y$; the fragmentation kernel $F(x,y)$ is describing the rate of formation of particles of size $x$ and $y$ by fragmentation of a particle of size $x+y$. Both kernels are assumed to be non-negative and symmetric. For an integer $m \geq 0$, the moment of order $m$ is defined by: $M_m(t) := \int_0^{+\infty} x^m c(x,t)dx$. The total number of particles, $M_0(t)$ may decrease by coagulation or increase by fragmentation, while the total mass of particles, $M_1(t)$ remains constant if there is neither gelation (formation of particles of infinite size by coagulation: see [1]) nor shattering (formation of zero-size particles by fragmentation: see [12]). We denote $M_1 := M_1(0)$.

Existence, uniqueness, boundedness, and positiveness of the solutions of coagulation-fragmentation equation have been considered in [3]. Exact solutions are only known for pure coagulation ($F = 0$) or pure fragmentation ($K = 0$) equation and for specific kernels and initial data, so efforts are being pursued to obtain accurate numerical solutions. We focus on stochastic (or quasi-stochastic) strategies. Several Monte Carlo (MC) methods have been developed concurrently. In [4] the density is approximated using a particle system with a variable particle number. In contrast to this *direct simulation* scheme, where the particles represent the number density, a *mass flow* scheme was introduced in [2]: the particles represent the mass density so that the number of particles is kept constant throughout the simulation. The article [6] presents a mass flow scheme for the coagulation-fragmentation equation. In [17] *time-driven* MC are opposed to *event-driven* MC. In time-driven simulations a time step is chosen, and all events are implemented within that step; in event-driven MC, first an event is selected to occur, and time is advanced by an appropriate increment. It is found that event-driven methods generally provide better accuracy; but time-driven algorithms are more suitable in cases where the equation is to be solved within a larger process simulator that performs explicit integration in time; see [7] for recent numerical experiments.

The aim of the present work is to extend to the coagulation-fragmentation equation the time-driven constant-number method introduced in [8, 9] for pure coagulation. The method uses quasi-random numbers (low-discrepancy point sets) in place of pseudo-random numbers. The basic notations and concepts of quasi-Monte Carlo (QMC) methods are given in [15]. We denote $I := [0,1)$. For a dimension $s$, the Lebesgue measure is denoted $\lambda_s$. For $U = \{\mathbf{u}_0, \ldots, \mathbf{u}_{N-1}\} \subset I^s$ and for a Borel set $B \subset I^s$ the *local discrepancy* is

$$D_N(B,U) := \frac{1}{N}\sum_{0\leq k<N} 1_B(\mathbf{u}_k) - \lambda_s(B),$$

where $1_B$ denotes the indicator function of $B$. The *discrepancy* of $U$ is $D_N(U) := \sup_J |D_N(J,U)|$, where the supremum is taken over all subintervals $J \subset I^s$. The *star discrepancy* of $U$ is $D_N^\star(U) := \sup_{J^\star} |D_N(J^\star,U)|$, where $J^\star$ runs through all subintervals of $I^s$ with a vertex at the origin. QMC methods are versions of MC methods, where the pseudo-random samples are replaced with low-discrepancy point sets. Powerful methods

of constructing low-discrepancy point sets are based on the theory of $(t, m, s)$-*nets* and $(t, s)$-*sequences*. For an integer $b \geq 2$, an *elementary interval in base* $b$ is an interval of the form $\prod_{i=1}^{s}[a_i b^{-d_i}, (a_i + 1)b^{-d_i})$, with integers $d_i \geq 0$ and $0 \leq a_i < b^{d_i}$. If $0 \leq t \leq m$ are integers, a $(t, m, s)$-*net in base* $b$ is a point set $U$ of $N = b^m$ points in $I^s$ such that $D_N(J, U) = 0$ for every elementary interval $J$ in base $b$ with measure $b^{t-m}$. If $b \geq 2$ and $t \geq 0$ are integers, a sequence $\mathbf{u}_0, \mathbf{u}_1, \ldots$ of points in $I^s$ is a $(t, s)$-*sequence in base* $b$ if, for all integers $n \geq 0$ and $m > t$, the points $\mathbf{u}_p$ with $nb^m \leq p < (n + 1)b^m$ form a $(t, m, s)$-net in base $b$.

For many MC schemes, it is possible to develop corresponding QMC algorithms by replacing the pseudo-random numbers with quasi-random numbers. In [8, 9], it was shown that it is convenient to take special measures in order to benefit from the great uniformity of quasi-random points: as time advances, the simulation particles are renumbered according to size at every time step.

The present paper is restricted to the presentation and analysis of a QMC (deterministic) scheme, which is able to produce an approximation of the density $c(x, t)$ for any $(x, t)$: this may be of interest in some cases (we think of inverse problems). The remainder of the paper is organized as follows. In Section 2, the QMC algorithm is presented. We establish in Section 3 a convergence result for the discrepancy of the numerical particles relative to the exact mass distribution. Results of computational experiments are shown in Section 4.

## 2 The quasi-Monte Carlo scheme

The case of pure coagulation was analyzed in our previous paper [8]. If $F$ is not identically null, we assume that, for every $x > 0$, the function $y \in (0, x) \mapsto F(x - y, y)$ is not identically null. We multiply Equation (1) by $x/M_1$. If we write $x = x - y + y$ in the first integral, it can be split up into two integrals, which are equated by a change of variable; we apply the same transformation to the last integral. By introducing the mass density function $f(x, t) := xc(x, t)/M_1$ (which is a probability density function), we obtain the *mass-flow equation*: for $x > 0,\ t > 0$,

$$\begin{aligned}\frac{\partial f}{\partial t}(x,t) &= \int_0^x \widetilde{K}(x-y,y)f(x-y,t)f(y,t)dy \\ &\quad - \int_0^{+\infty} \widetilde{K}(x,y)f(x,t)f(y,t)dy \\ &\quad + \int_0^{+\infty} \widetilde{F}(x+y,x)f(x+y,t)dy - \int_0^x \widetilde{F}(x,y)f(x,t)dy, \end{aligned} \tag{2}$$

where $\widetilde{K}$ and $\widetilde{F}$ are the modified coagulation and fragmentation kernels respectively defined by: $\widetilde{K}(x, y) := M_1 K(x, y)/y$ (for $x, y > 0$), and $\widetilde{F}(x, y) := yF(x - y, y)/x$ (for $x > y > 0$). We denote $f_0(x) := xc_0(x)/M_1$ the initial data. We introduce a weak formulation of Equation (2), so we define a set of test functions. If $\mathbb{R}_+^* := (0, +\infty)$, then a function $\sigma : \mathbb{R}_+^* \to \mathbb{R}_+$ is said to be *simple* if its image is a finite pointset of $\mathbb{R}_+$; let $\mathcal{S}(\mathbb{R}_+^*)$ be the set of all measurable simple functions on $\mathbb{R}_+^*$. By multiplying Equation (2) by a simple function $\sigma \in \mathcal{S}(\mathbb{R}_+^*)$ and by integrating over $\mathbb{R}_+^*$, we obtain a first weak formulation of the mass-flow equation:

$$\begin{aligned}\frac{d}{dt}\int_0^{+\infty} f(x,t)\sigma(x)dx = \\ \int_0^{+\infty}\int_0^{+\infty} \widetilde{K}(x,y)f(x,t)f(y,t)(\sigma(x+y)-\sigma(x))dydx \\ + \int_0^{+\infty}\int_0^{x} \widetilde{F}(x,y)f(x,t)(\sigma(y)-\sigma(x))dydx\end{aligned}$$

(in the first integral we use new variables: $(x, z)$, where $z := x - y$, then rename $z \to y$; in the third integral we use new variables: $(x, z)$, where $z := x + y$, then rename $(z, x) \to (x, y)$). Before doing the numerical integration, we convert the integral over $[0, x]$ to an integral over $[0, 1]$. For $x \geq y > 0$, let

$$\mathcal{F}(x,y) := \int_0^y \widetilde{F}(x,w)dw, \quad \dot{\mathcal{F}}(x) := \mathcal{F}(x,x), \quad \widehat{\mathcal{F}}(x,y) := \frac{\mathcal{F}(x,y)}{\dot{\mathcal{F}}(x)}.$$

For $x > 0$, let $\widehat{\mathcal{F}}_x$ be the function $y \in [0,x] \to \widehat{\mathcal{F}}(x,y) \in [0,1]$, with $\widehat{\mathcal{F}}_x(0) := 0$. We assume that this function is strictly increasing; let $\widehat{\mathcal{F}}_x^{-1}$ be the inverse function. In the integral over $[0,x]$, we change variable $y$ to $u := \widehat{\mathcal{F}}_x(y)$, and we obtain another weak formulation of the mass-flow equation:

$$\begin{aligned}\frac{d}{dt}\int_0^{+\infty} f(x,t)\sigma(x)dx = \\ \int_0^{+\infty}\int_0^{+\infty} \widetilde{K}(x,y)f(x,t)f(y,t)(\sigma(x+y)-\sigma(x))dydx \\ + \int_0^{+\infty} \dot{\mathcal{F}}(x)f(x,t)\left(\int_0^1 \sigma\circ\widehat{\mathcal{F}}_x^{-1}(u)du - \sigma(x)\right)dx. \end{aligned} \tag{3}$$

We suppose that $\widetilde{K}$ and $\dot{\mathcal{F}}$ are bounded and we set $\widetilde{K}^\infty := \sup_{x,y>0}\widetilde{K}(x,y)$, $\dot{\mathcal{F}}^\infty := \sup_{x>0}\dot{\mathcal{F}}(x)$. For $b \geq 2$ and $m \geq 1$, we put $N := b^m$: this is the number of numerical particles used for the simulation. We need a low discrepancy sequence for the time evolution: $U = \{\mathbf{u}_0, \mathbf{u}_1, \ldots\} \subset I^3$. For $n \in \mathbb{N}$, we write: $U^n := \{\mathbf{u}_p : nN \leq p < (n+1)N\}$. We make the following assumptions: $U$ is a $(t,3)$-sequence in base $b$ (for some $t \geq 0$) and $\pi_2^3(U^n)$ is a $(0,m,2)$-net in base $b$ (where $\pi_2^3$ is the projection defined by $\pi_2^3(x_1,x_2,x_3) := (x_1,x_2)$).

## 2.1 Initialization

We choose a set $X^0 = \{x_0^0, \ldots, x_{N-1}^0\} \subset \mathbb{R}_+^*$ of $N$ particles such that the initial mass probability $f_0(x)dx$ is approximated by the probability distribution:

$$f^0(x) := \frac{1}{N}\sum_{0\leq k<N}\delta(x-x_k^0),$$

where $\delta(x-\xi)$ is the Dirac measure at a point $\xi \in \mathbb{R}_+^*$. This reverts to generating $N$ samples from the density function $f_0$.

## 2.2 Time discretization

A fixed time step $\Delta t$ is chosen such that $\Delta t(\widetilde{K}^\infty + \dot{\mathcal{F}}^\infty) < 1$. We set $t_n := n\Delta t$ and we denote $f_n(x) := f(x,t_n)$. We suppose that a set $X^n = \{x_0^n, \ldots, x_{N-1}^n\} \subset \mathbb{R}_+^*$ of $N$ particles has been computed so that

$$f^n(x) := \frac{1}{N}\sum_{0\leq k<N}\delta(x-x_k^n)$$

approximates (in a sense to be made precise below) the exact mass probability $f_n(x)dx$. The approximation of the solution at time $t_{n+1}$ is calculated as follows.

*(i) Renumbering the particles.* Particles are relabeled at the beginning of the time step by increasing mass: $x_0^n \leq x_1^n \leq \cdots \leq x_{N-1}^n$. This sorting guarantees theoretical convergence: see Section 3 below.

*(ii) Coagulation/fragmentation.* We define an auxiliary probability measure $g^{n+1}$ by using an explicit Euler scheme in time for Equation (3):

$$\begin{aligned}\int_0^{+\infty} g^{n+1}(x)\sigma(x) = \\ \frac{1}{N}\sum_{0\leq k<N}\left(1 - \frac{\Delta t}{N}\sum_{0\leq \ell<N}\widetilde{K}(x_k^n,x_\ell^n) - \Delta t\dot{\mathcal{F}}(x_k^n)\right)\sigma(x_k^n) \\ + \frac{\Delta t}{N}\sum_{0\leq k<N}\left(\frac{1}{N}\sum_{0\leq \ell<N}\widetilde{K}(x_k^n,x_\ell^n)\sigma(x_k^n+x_\ell^n) + \dot{\mathcal{F}}(x_k^n)\int_0^1 \sigma\circ\widehat{\mathcal{F}}_{x_k^n}^{-1}(u)du\right).\end{aligned}$$

The measure $g^{n+1}$ approximates $f_{n+1}(x)dx$, but it is not a sum of Dirac measures. We recover this kind of approximation if we use a QMC quadrature rule. Let $R_{k,\ell} := [k/N,(k+1)/N)\times[\ell/N,(\ell+1)/N) \subset I^2$ be an elementary interval in base $b$: we denote by $1_{R_{k,\ell}}$ its indicator function. Let $\chi^n_{k,\ell}$ be the indicator function of $I^n_{k,\ell} := [0, \Delta t\widetilde{K}(x^n_k, x^n_\ell))$ and let $\varphi^n_k$ be the indicator function of $J^n_k := [1-\Delta t\dot{\mathcal{F}}(x^n_k), 1)$. To $\sigma \in \mathcal{S}(\mathbb{R}^*_+)$ corresponds the indicator function:

$$\begin{aligned}\Gamma^{n+1}_\sigma(\mathbf{u}) \quad := \quad & \sum_{0\le k,\ell<N} 1_{R_{k,\ell}}(u_1,u_2)\Bigg( \left(1-\chi^n_{k,\ell}(u_3)-\varphi^n_k(u_3)\right)\sigma(x^n_k) \\ & +\chi^n_{k,\ell}(u_3)\sigma(x^n_k+x^n_\ell) + \varphi^n_k(u_3)\sigma\circ\widehat{\mathcal{F}}^{-1}_{x^n_k}\left(\frac{1-u_3}{\Delta t\dot{\mathcal{F}}(x^n_k)}\right)\Bigg)\end{aligned}$$

(for $\mathbf{u} = (u_1,u_2,u_3) \in I^3$), which is such that

$$\int_0^{+\infty} g^{n+1}(x)\sigma(x) = \int_{I^3} \Gamma^{n+1}_\sigma(\mathbf{u})d\mathbf{u}.$$

We determine $f^{n+1}$ by performing a QMC quadrature in $I^3$:

$$\int_0^{+\infty} f^{n+1}(x)\sigma(x) = \frac{1}{N}\sum_{nN\le p<(n+1)N} \Gamma^{n+1}_\sigma(\mathbf{u}_p).$$

The calculation on a time step may be summarized as follows. If $u \in [0,1)$, let $k(u) := \lfloor Nu\rfloor$. Then, for every $p$ with $nN \le p < (n+1)N$, we have:

$$x^{n+1}_{k(u_{p,1})} = \begin{cases} x^n_{k(u_{p,1})} + x^n_{k(u_{p,2})} & \text{if } u_{p,3} < \Delta t\widetilde{K}\left(x^n_{k(u_{p,1})}, x^n_{k(u_{p,2})}\right), \\ \widehat{\mathcal{F}}^{-1}_{x^n_{k(u_{p,1})}}\left(\dfrac{1-u_{p,3}}{\Delta t\dot{\mathcal{F}}(x^n_{k(u_{p,1})})}\right) & \text{if } 1-\Delta t\dot{\mathcal{F}}\left(x^n_{k(u_{p,1})}\right) \le u_{p,3}, \\ x^n_{k(u_{p,1})} & \text{otherwise.}\end{cases}$$

For every $p$, the numbers $u_{p,1}$ and $u_{p,2}$ select particles; the particle $k(u_{p,1})$ has for coagulation partner the particle $k(u_{p,2})$ and the coagulation probability is $p_c := \Delta t\widetilde{K}(x^n_{k(u_{p,1})}, x^n_{k(u_{p,2})})$; the fragmentation probability of particle $k(u_{p,1})$ is $p_f := \Delta t\dot{\mathcal{F}}(x^n_{k(u_{p,1})})$. Then $u_{p,3}$ selects an event and in the case of fragmentation, it determines the size of the new particle. The algorithm is a slight modification of the method devised for pure coagulation and uses three-dimensional quasi-random numbers as well. This is interesting, because it is well known that QMC methods perform better in low dimensional spaces. Nevertheless, the extension of the convergence analysis is not straightforward, because fragmentation introduces new linear terms (the coagulation terms are quadratic in $c$).

## 3 Convergence analysis

We prove a convergence result for the QMC scheme previously described. First, we need to adapt the basic tools of QMC methods to the algorithm.

Let $f$ be a probability density function on $\mathbb{R}^*_+$. If $z > 0$, then $\sigma_z$ is the indicator function of $(0,z)$. We define the *local discrepancy* of the set $X = \{x_k : 0 \le k < N\} \subset \mathbb{R}^*_+$ relative to $f$ by:

$$D_N(z,X;f) := \frac{1}{N}\sum_{0\le k<N}\sigma_z(x_k) - \int_0^{+\infty}\sigma_z(x)f(x)dx.$$

The *star discrepancy* of $X$ relative to $f$ is: $D^\star_N(X;f) := \sup_{z>0}|D_N(z,X;f)|$. The *error* of the scheme at time $t_n$ is defined to be $D^\star_N(X^n;f_n)$. The concept of variation of function in the sense of Hardy and Krause can be extended to a function $\phi$ defined on $\mathbb{R}^{*s}_+$ and is denoted by $V(\phi)$. The Koksma inequality can be generalized as follows.

**Proposition 1** *Let $f$ be a probability density function over $\mathbb{R}_+^*$. If $\phi$ has bounded variation $V(\phi)$ on $\mathbb{R}_+^*$, then for any $X = \{x_k : 0 \leq k < N\} \subset \mathbb{R}_+^*$,*

$$\left| \frac{1}{N} \sum_{0 \leq k < N} \phi(x_k) - \int_0^{+\infty} \phi(x) f(x) dx \right| \leq V(\phi) D_N^\star(X; f).$$

We introduce the following intermediate terms.

- The *local truncation error*:

$$\begin{aligned} \varepsilon_z^n &:= \frac{1}{\Delta t} \int_0^{+\infty} (f_{n+1}(x) - f_n(x)) \sigma_z(x) dx \\ &\quad - \int_0^{+\infty} \int_0^{+\infty} \widetilde{K}(x,y) f_n(x) f_n(y) (\sigma_z(x+y) - \sigma_z(x)) dy dx \\ &\quad - \int_0^{+\infty} \dot{\mathcal{F}}(x) f_n(x) \left( \int_0^1 \sigma_z \circ \widehat{\mathcal{F}}_x^{-1}(u) du - \sigma_z(x) \right) dx. \end{aligned}$$

- The *additional error*: $e_z^n = e_{z,K}^n + e_{z,F}^n$, where

$$\begin{aligned} e_{z,K}^n &:= \int_0^{+\infty} \int_0^{+\infty} \widetilde{K}(x,y) f^n(x) f^n(y) (\sigma_z(x+y) - \sigma_z(x)) \\ &\quad - \int_0^{+\infty} \int_0^{+\infty} \widetilde{K}(x,y) f_n(x) f_n(y) (\sigma_z(x+y) - \sigma_z(x)) dy dx, \\ e_{z,F}^n &:= \int_0^{+\infty} \dot{\mathcal{F}}(x) f^n(x) \left( \int_0^1 \sigma_z \circ \widehat{\mathcal{F}}_x^{-1}(u) du - \sigma_z(x) \right) \\ &\quad - \int_0^{+\infty} \dot{\mathcal{F}}(x) f_n(x) \left( \int_0^1 \sigma_z \circ \widehat{\mathcal{F}}_x^{-1}(u) du - \sigma_z(x) \right) dx. \end{aligned}$$

- The *QMC integration error*:

$$d_z^n := \frac{1}{N} \sum_{nN \leq p < (n+1)N} \Gamma_{\sigma_z}^{n+1}(\mathbf{u}_p) - \int_{I^3} \Gamma_{\sigma_z}^{n+1}(\mathbf{u}) d\mathbf{u}.$$

We have the recurrence formula:

$$D_N(z, X^{n+1}; f_{n+1}) = D_N(z, X^n; f_n) - \Delta t \varepsilon_z^n + \Delta t e_z^n + d_z^n.$$

The local truncation error is bounded as follows.

**Lemma 1** *If for every $x > 0$, the function $t \to f(x,t)$ is twice continuously differentiable over $(0,T)$ and if $f$, $\frac{\partial f}{\partial t}$, $\frac{\partial^2 f}{\partial t^2}$ are integrable over $\mathbb{R}_+^* \times (0,T)$, then, for $t_{n+1} \leq T$,*

$$|\varepsilon_z^n| \leq \int_0^{+\infty} \int_{t_n}^{t_{n+1}} \left| \frac{\partial^2 f}{\partial t^2}(x,t) \right| dt dx.$$

The part of the additional error associated to coagulation is bounded in our previous paper [8].

**Lemma 2** *If for every $y > 0$, the function $\widetilde{K}(\cdot, y) : x \in (0, +\infty) \mapsto \widetilde{K}(x,y)$ is of bounded variation $V(\widetilde{K}(\cdot, y))$ and $\sup_{y>0} V(\widetilde{K}(\cdot, y)) < +\infty$, and if for every $x > 0$, the function $\widetilde{K}(x, \cdot) : y \in (0, +\infty) \mapsto \widetilde{K}(x,y)$ is of bounded variation $V(\widetilde{K}(x, \cdot))$ and $\sup_{x>0} V(\widetilde{K}(x, \cdot)) < +\infty$, then*

$$|e_{z,K}^n| \leq \left( \sup_{y>0} V(\widetilde{K}(\cdot, y)) + \sup_{x>0} V(\widetilde{K}(x, \cdot)) + 3\widetilde{K}^\infty \right) D_N^\star(X^n; f_n).$$

We have the following bound for the part of the additional error associated to fragmentation.

**Lemma 3** *If for every $y > 0$, the map $\mathcal{F}(\cdot, y) : x \in [y, +\infty) \mapsto \mathcal{F}(x, y)$ is of bounded variation $V(\mathcal{F}(\cdot, y))$ and $\sup_{y>0} V(\mathcal{F}(\cdot, y)) < +\infty$, then*

$$|e^n_{z,F}| \leq \left( \sup_{y>0} V(\mathcal{F}(\cdot, y)) + \dot{\mathcal{F}}^{\infty} \right) D^{\star}_N(X^n; f_n).$$

**Proof.** We have

$$\begin{aligned} e^n_{z,F} &= \int_0^{+\infty} f^n(x) \int_0^x \widetilde{F}(x, y)(\sigma_z(y) - \sigma_z(x))dy \\ &\quad - \int_0^{+\infty} f_n(x) \int_0^x \widetilde{F}(x, y)(\sigma_z(y) - \sigma_z(x))dy. \end{aligned}$$

If we define a function: $\varphi(x) := \int_0^x \widetilde{F}(x, y)(\sigma_z(y) - \sigma_z(x))dy$ (for $x > 0$), then

$$e^n_{z,F} := \frac{1}{N} \sum_{0 \leq k < N} \varphi(x^n_k) - \int_0^{+\infty} \varphi(x) f_n(x) dx.$$

Using the generalized Koksma inequality leads to: $|e^n_{z,F}| \leq V(\varphi) D^{\star}_N(X^n; f_n)$. Since $V(\varphi) \leq \sup_{y>0} V(\mathcal{F}(\cdot, y)) + \dot{\mathcal{F}}^{\infty}$, the result of the lemma follows. □

For bounding the QMC integration error $d^n_z$, we use the following result (see Lemma 3.4 of [14]):

**Lemma 4** *Let $U$ be a $(t, m, s)$-net in base $b$. For every elementary interval $J' \subset I^{s-1}$ and for every $u_s \in I$, we have: $|D_{b^m}(J' \times (u_s, 1), U)| \leq b^{t-m}$.*

**Lemma 5** *If $\widetilde{K}$ is of bounded variation in the sense of Hardy and Krause, and if for every $y > 0$, the function $\mathcal{F}(\cdot, y) : x \in [y, +\infty) \mapsto \mathcal{F}(x, y)$ is of bounded variation $V(\mathcal{F}(\cdot, y))$ and $\sup_{y>0} V(\mathcal{F}(\cdot, y)) < +\infty$, then*

$$|d^n_z| \leq (2 + c_K \Delta t) b^{-\lfloor (m-t)/3 \rfloor} + (1 + c_F \Delta t)^{\lfloor (m-t)/2 \rfloor},$$

*where*

$$c_K := 4V(\widetilde{K}) + 3\widetilde{K}^{\infty} \quad \text{and} \quad c_F := \sup_{y>0} V(\mathcal{F}(\cdot, y)) + \sup_{x \geq y > 0} \mathcal{F}(x, y). \tag{4}$$

**Proof.** The function $\Gamma^{n+1}_{\sigma_z}$ is the indicator function of some subset $P^n_z$ of $I^3$, thus $d^n_z = D_N(P^n_z, U^n)$. We have the disjoint union: $P^n_z = (P^n_{z,0} \setminus \widetilde{P}^n_{z,1}) \cup P^n_{z,2} \cup \widetilde{P}^n_{z,F}$, where

$$\begin{aligned} P^n_{z,0} &:= \bigcup_{\substack{0 \leq k,\ell < N \\ x^n_k < z}} R_{k,\ell} \times I, \quad \widetilde{P}^n_{z,1} := \bigcup_{\substack{0 \leq k,\ell < N \\ x^n_k < z}} R_{k,\ell} \times (I^n_{k,\ell} \cup J^n_k), \\ P^n_{z,2} &:= \bigcup_{\substack{0 \leq k,\ell < N \\ x^n_k + x^n_\ell < z}} R_{k,\ell} \times I^n_{k,\ell}, \\ \widetilde{P}^n_{z,F} &:= \bigcup_{0 \leq k,\ell < N} R_{k,\ell} \times J^n_k \cap \left\{ u \in I : \widehat{\mathcal{F}}^{-1}_{x^n_k} \left( \frac{1-u}{\Delta t \dot{\mathcal{F}}(x^n_k)} \right) < z \right\}. \end{aligned}$$

We also have the disjoint union: $P^n_z = \left(P^n_{z,0} \setminus P^n_{z,1}\right) \cup P^n_{z,2} \cup P^n_{z,F}$, where

$$P^n_{z,1} := \bigcup_{\substack{0 \leq k,\ell < N \\ x^n_k < z}} R_{k,\ell} \times I^n_{k,\ell} \quad \text{and} \quad P^n_{z,F} := \bigcup_{\substack{0 \leq k,\ell < N \\ x^n_k \geq z}} R_{k,\ell} \times (1 - \Delta t \mathcal{F}(x^n_k, z), 1).$$

Consequently, $d_z^n$ can be split up: $d_z^n = d_{z,K}^n + D_N(P_{z,F}^n, U^n)$, where

$$d_{z,K}^n := D_N(P_{z,0}^n, U^n) - D_N(P_{z,1}^n, U^n) + D_N(P_{z,2}^n, U^n).$$

In [8] we have proved: $|d_{z,K}^n| \leq (2 + c_K \Delta t) b^{-\lfloor (m-t)/3 \rfloor}$. We now establish a bound for $|D_N(P_{z,F}^n, U^n)|$. By denoting $I_k := [k/N, (k+1)/N)$, we have:

$$P_{z,F}^n = \bigcup_{\substack{0 \leq k < N \\ x_k^n \geq z}} I_k \times I \times (1 - \Delta t \mathcal{F}(x_k^n, z), 1).$$

We set $\sigma_z^c := 1 - \sigma_z$ and we introduce the function:

$$\phi_z^n(u) := \sum_{0 \leq k < N} 1_{I_k}(u) \mathcal{F}(x_k^n, z) \sigma_z^c(x_k^n), \quad u \in I,$$

where $1_{I_k}$ is the indicator function of $I_k$. Then $P_{z,F}^n = \{\mathbf{u} \in I^3 : u_3 > 1 - \Delta t \phi_z^n(u_1)\}$. Let $d_1$ be an integer such that $d_1 \leq m - t$: its value is determined hereafter. For $a_1 \in \mathbb{N}$ with $0 \leq a_1 < b^{d_1}$, we define: $I'_{a_1} := [a_1 b^{-d_1}, (a_1 + 1) b^{-d_1})$ and

$$\begin{aligned}
\underline{P}_{z,F}^n &:= \bigcup_{0 \leq a_1 < b^{d_1}} I'_{a_1} \times I \times (1 - \Delta t \inf_{I'_{a_1}} \phi_z^n, 1), \\
\overline{P}_{z,F}^n &:= \bigcup_{0 \leq a_1 < b^{d_1}} I'_{a_1} \times I \times (1 - \Delta t \sup_{I'_{a_1}} \phi_z^n, 1), \\
\partial P_{z,F}^n &:= \bigcup_{0 \leq a_1 < b^{d_1}} I'_{a_1} \times I \times [1 - \Delta t \sup_{I'_{a_1}} \phi_z^n, 1 - \Delta t \inf_{I'_{a_1}} \phi_z^n].
\end{aligned}$$

We have $\underline{P}_{z,F}^n \subset P_{z,F}^n \subset \overline{P}_{z,F}^n$ and $\overline{P}_{z,F}^n \setminus \underline{P}_{z,F}^n \subset \partial P_{z,F}^n$. Consequently,

$$D_N(\underline{P}_{z,F}^n, U^n) - \lambda_3(\partial P_{z,F}^n) \leq D_N(P_{z,F}^n, U^n) \leq D_N(\overline{P}_{z,F}^n, U^n) + \lambda_3(\partial P_{z,F}^n).$$

Since the $I'_{a_1} \times I$ are disjoint elementary intervals in base $b$, an application of Lemma 4 yields: $\max\left(|D_N(\underline{P}_{z,F}^n, U^n)|, |D_N(\overline{P}_{z,F}^n, U^n)|\right) \leq b^{d_1 + t - m}$. Besides,

$$\lambda_3(\partial P_{z,F}^n) = \frac{\Delta t}{b^{d_1}} \sum_{0 \leq a_1 < b^{d_1}} \left( \sup_{I'_{a_1}} \phi_z^n - \inf_{I'_{a_1}} \phi_z^n \right).$$

If we introduce the function $\psi_z(x) := \mathcal{F}(x, z) \sigma_z^c(x)$ (for $x > 0$), then $\phi_z^n(u) = \psi_z(x_{k(u)}^n)$. We define the sets: $E_{a_1}^n := [x_{a_1 b^{m-d_1}}^n, x_{(a_1+1)b^{m-d_1}-1}^n]$. Since the particles are numbered by increasing size, we get: $u \in I'_{a_1} \Rightarrow x_{k(u)}^n \in E_{a_1}^n$. Consequently we have:

$$\sup_{I'_{a_1}} \phi_z^n - \inf_{I'_{a_1}} \phi_z^n \leq \sup_{E_{a_1}^n} \psi_z - \inf_{E_{a_1}^n} \psi_z.$$

Since

$$\sum_{0 \leq a_1 < b^{d_1}} \left( \sup_{E_{a_1}^n} \psi_z - \inf_{E_{a_1}^n} \psi_z \right) \leq V(\psi_z) \leq V(\mathcal{F}(\cdot, z)) + \sup_{x > 0} \mathcal{F}(x, z),$$

we obtain: $|D_N(P_{z,F}^n, X^n)| \leq b^{d_1 + t - m} + c_F \Delta t b^{-d_1}$. By choosing $d_1 = \lfloor (m-t)/2 \rfloor$, we get $|D_N(P_{z,F}^n, U^n)| \leq (1 + c_F \Delta t) b^{-\lfloor (m-t)/2 \rfloor}$. In conjunction with the bound for $d_{z,K}^n$, this gives the final result. □

By combining the results of Lemmas 1, 2, 3 and 5, we obtain an upper bound for the error $D_N^\star(X^n; f_n)$; it resembles the final result of [8] and is similarly established. Since $N = b^m$, we have $1/b^{\lfloor (m-t)/3 \rfloor} = \mathcal{O}(1/N^{1/3})$ and $1/b^{\lfloor (m-t)/2 \rfloor} = \mathcal{O}(1/N^{1/2})$.

**Proposition 2** *If*

*(1) for every $x > 0$, the function $t \to f(x,t)$ is twice continuously differentiable over $(0,T)$ and $f, \frac{\partial f}{\partial t}, \frac{\partial^2 f}{\partial t^2}$ are integrable over $\mathbb{R}_+^* \times (0,T)$,*

*(2) $\widetilde{K}$ is of bounded variation in the sense of Hardy and Krause,*

*(3) for every $y > 0$, the function $\mathcal{F}(\cdot, y) : x \in [y, +\infty) \mapsto \mathcal{F}(x,y)$ is of bounded variation $V(\mathcal{F}(\cdot,y))$ and $\sup_{y>0} V(\mathcal{F}(\cdot,y)) < +\infty$, then*

$$\begin{aligned} D_N^\star(X^n; f_n) \quad \leq \quad & \mathrm{e}^{ct_n} D_N^\star(X^0; f_0) + \Delta t \int_0^{+\infty} \int_0^{t_n} \mathrm{e}^{c(t_n - t)} \left| \frac{\partial^2 f}{\partial t^2}(x,t) \right| dt dx \\ & + \left( \left( \frac{2}{\Delta t} + c_K \right) \frac{1}{b^{\lfloor (m-t)/3 \rfloor}} + \left( \frac{1}{\Delta t} + c_F \right) \frac{1}{b^{\lfloor (m-t)/2 \rfloor}} \right) \frac{\mathrm{e}^{ct_n} - 1}{c} \end{aligned}$$

*where $t_n = n\Delta t$, $c_K$, $c_F$ are given by Equation (4) and*

$$c := \sup_{x>0} V(\widetilde{K}(x,.)) + \sup_{y>0} V(\widetilde{K}(.,y)) + 3\widetilde{K}^\infty + \sup_{y>0} V(\mathcal{F}(\cdot,y)) + \dot{\mathcal{F}}^\infty.$$

This upper bound does not converge as fast as we would like, but is nevertheless a worst-case deterministic bound. The $\mathcal{O}(N^{-1/3})$ convergence of the bound is due to QMC integration of indicator functions in 3D; the $\mathcal{O}(1/\Delta t)$ is due to the summation of integration errors at every time step. The same result would have been obtained if the same QMC pointset has been used at every time step (which leads in practice to a method where the error increases with the number of time steps). A more complicated error analysis would take into account that an infinite QMC sequence is used, but in this case, the QMC integration of indicator functions would be in 4D and leads to $\mathcal{O}(N^{-1/4})$ convergence. The present bound only guarantees that the error can be arbitrarily small, if $\Delta t$ is small enough and $N$ is large enough. It does not aim to provide an optimal time step for a given $N$. Numerical experiments show that the method converges faster than $\mathcal{O}(N^{-1/2})$ and the error decreases with $\Delta t$.

## 4 Numerical results

The efficiency of the QMC scheme is tested empirically. To do this, approximate solutions are computed in a case where analytical solutions for the moments are available. A quantity of interest when simulating liquid-liquid extraction problems is the *Sauter mean diameter* of drops, which is calculated with the second and third moments. The QMC solutions are also compared with those given by the MC scheme adapted from the algorithm described in [2]. We consider the case where coagulation and fragmentation occur simultaneously with the following kernels: $K(x,y) = 1$ and $F(x,y) = 2/(x+y)$, which is considered in [6]. Then Equation (1) becomes:

$$\begin{aligned} \frac{\partial c}{\partial t}(x,t) \quad = \quad & \frac{1}{2} \int_0^x c(x-y,t) c(y,t) dy - c(x,t) \int_0^{+\infty} c(y,t) dy \\ & + 2 \int_x^{+\infty} \frac{1}{y} c(y,t) dy - c(x,t), \quad x > 0,\ t > 0. \end{aligned} \tag{5}$$

We restrict ourselves to monodisperse initial condition: $c(x,0) = \delta(x-1)$. By multiplying Equation (5) by $x^m$ and using integration over $(0, +\infty)$, we get differential equations for the moments $M_m(t)$, which can be solved to obtain:

$$M_2(t) = 3 - 2\mathrm{e}^{-t/3} \quad \text{and} \quad M_3(t) = 18 - 36\mathrm{e}^{-t/3} + 19\mathrm{e}^{-t/2}.$$

We compute the solution up to time $T = 1.0$ with $N$ particles ($N$ varying from $2^7$ to $2^{20}$) and $P$ time steps ($P$ varying from $2^0 \times 100$ to $2^4 \times 100$). All QMC computations use a $(1,3)$-sequence in base 2 constructed by H. Niederreiter [15]. The moment $M_m(t_n)$ at time $t_n$ can be approximated, using the MC or QMC scheme, by:

$$M_{m,N,P,n} := \frac{1}{N} \sum_{0 \leq k < N} (x_k^n)^{m-1}. \tag{6}$$

First we compute the absolute error in the moment estimation at time $\alpha T$, for $\alpha = 1/100, 1/10$ and 1:

$$E^{\alpha}_{m,N,P} := |M_m(\alpha T) - M_{m,N,P,\alpha P}|.$$

Figure 1 represents the log-log plots (base 2) of these absolute errors, for different values of $N$ and $P$. In all cases, the QMC simulations give better results (for the same discretization parameters) than those given by the Monte Carlo scheme. The error seems stable on the time interval $[1/100, 1/10]$, so that it makes sense to compare the methods on the whole time interval $[0, 1]$: the *mean absolute error* in the moment estimation is

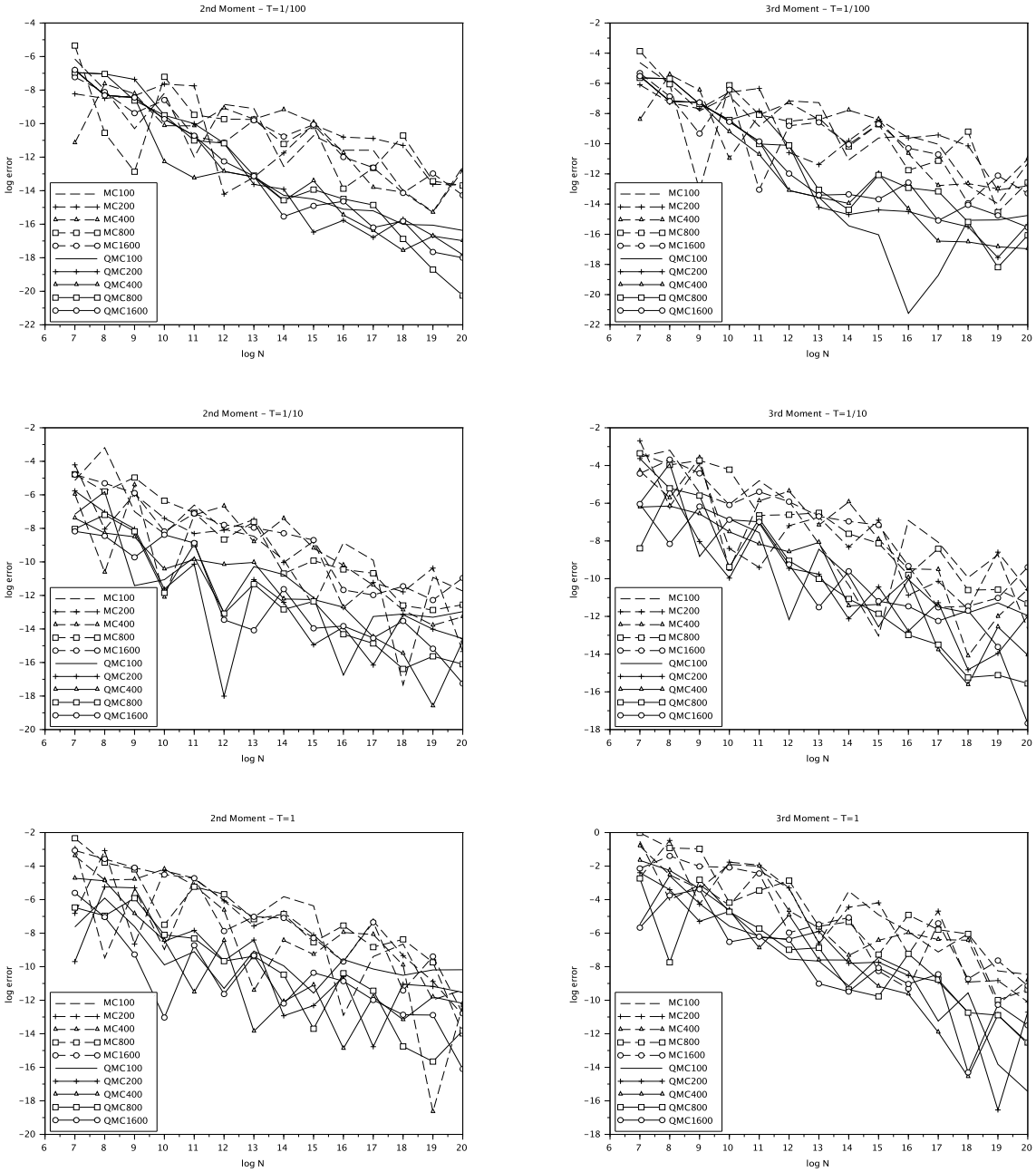


**Figure 1: Error in the second (left) and third (right) moment estimation at time $1/100$ (top) $1/10$ (middle) and $1$ (bottom), as a function of $N$ (number of particles varying from $2^7$ to $2^{20}$), for $P$ (number of time steps) between $100$ and $1\,600$. Log-log plots of the MC (dashed line) versus QMC (solid lines) results.**

calculated as follows:

$$E_{m,N,P} := \frac{1}{100}\sum_{h=1}^{100} |M_m(hT/100) - M_{m,N,P,hP/100}|.$$

Figure 2 shows the log-log plots (base 2) of the mean absolute error in the estimation of the moments of order 2 and 3, for different values of $N$ and $P$. In both cases, the QMC simulations show faster convergence than the Monte Carlo scheme.

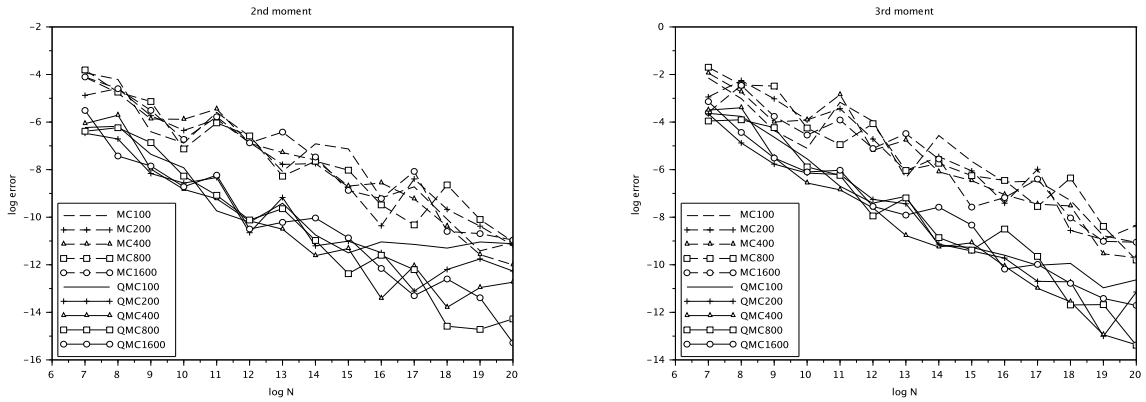


**Figure 2: Mean error in the second (left) and third (right) moment estimation as a function of $N$ (number of particles varying from $2^7$ to $2^{20}$), for $P$ (number of time steps) between 100 and 1 600. Log-log plots of the MC (dashed line) versus QMC (solid lines) results.**

## 5 Conclusion

In this paper, we have presented and analyzed a new algorithm for the approximation of the continuous coagulation-fragmentation equation. A sample of $N$ numerical particles is used to simulate the behavior of the system as a whole. Time is discretized into $P$ time steps and the mass density is approximated by a sum of $N$ Dirac measures. Three-dimensional quasi-random points are used to change the particles masses according to the dynamics of the equation.

A deterministic error bound is established. The accuracy of the algorithm is assessed through comparison with exact values, in a test case where analytical results are available. The numerical approximations given by the QMC scheme converge to the exact results when $P$ and $N$ are large enough. The errors given by the QMC simulation are always smaller than the errors given by the MC method using the same $P$ and $N$.

As announced, we have restricted the study to a deterministic scheme. Randomized QMC would be an interesting alternative (see [10, 11]), but requires a different analysis: this will be the subject of a forthcoming paper.

## References

[1] D. J. Aldous, Deterministic and stochastic models for coalescence (aggregation and coagulation): a review of the mean-field theory for probabilists, Bernoulli 5 (1999) 3–48.

[2] H. Babovsky, On a Monte Carlo scheme for Smoluchowski's coagulation equation, Monte Carlo Methods Appl. 5 (1999) 1–18.

[3] P. B. Dubovskiĭ, I. W. Stewart, Existence, uniqueness and mass conservation for the coagulation-fragmentation equation, Math. Methods Appl. Sci. 19 (1996) 571–591.

[4] A. Eibeck, W. Wagner, Approximative solution of the coagulation-fragmentation equation by stochastic particle systems, Stochastic Anal. Appl. 18 (2000) 921–948.

[5] S. K. Friedlander, On the particle size spectrum of a condensing vapor, Phys. Fluids 3 (1960) 693–696.

[6] M. Goodson, M. Kraft, Simulation of coalescence and breakage: an assessment of two stochastic methods suitable for simulating liquid-liquid extraction, Chem. Eng. Sci. 59 (2004) 3865–3881.

[7] G. Kotalczyk, J. Devi, F.E. Kruis, A time-driven constant-number Monte Carlo method for the GPU-simulation of particle breakage based on weighted simulation particles, Powder Technol. 317 (2017) 417–429.

[8] C. Lécot, A. Tarhini, A quasi-Monte Carlo method for the coagulation equation, in: G. Larcher, F. Pillichshammer, A. Winterhof, C. Xing (Eds.) Applied Algebra and Number Theory, Cambridge University Press, 2014, pp. 216–234.

[9] C. Lécot, W. Wagner, A quasi-Monte Carlo scheme for Smoluchowski's coagulation equation, Math. Comput. 73 (2004) 1953–1966.

[10] P. L'Ecuyer, C. Lécot, B. Tuffin, A randomized quasi-Monte Carlo simulation method for Markov chains, Operations Research 56 (2008) 958–975.

[11] P. L'Ecuyer, D. Munger, C. Lécot, B. Tuffin, Sorting methods and convergence rates for Array-RQMC: Some empirical comparisons, Math. Comput. Simulation 143 (2018) 191–201.

[12] E. D. McGrady, R. M. Ziff, "Shattering" transition in fragmentation, Phys. Rev. Lett. 58 (1987) 892–895.

[13] H. Müller, Zur allgemeinen Theorie der raschen Koagulation, Kolloidchemische Beihefte 27 (1928) 223–250.

[14] H. Niederreiter, Point sets and sequences with small discrepancy, Monatsh. Math. 104 (1987) 273–337.

[15] H. Niederreiter, Random Number Generation and Quasi-Monte Carlo Methods, SIAM, Philadelphia, 1992.

[16] M. v. Smoluchowski, Versuch einer mathematischen Theorie der Koagulationskinetik kolloider Lösungen, Z. f. physik. Chemie 92 (1916) 129–168.

[17] H. Zhao, A. Maisels, T. Matsoukas, C. Zheng, Analysis of four Monte Carlo methods for the solution of population balances in dispersed systems, Powder Technol. 173 (2007) 38–50.