
Preface

The International conference on *Multiscale problems in science and technology; Challenges to mathematical analysis and applications* brought together mathematicians working on multiscale techniques (homogenisation, singular perturbation) and specialists from applied sciences who use these techniques. Our idea was that mathematicians could contribute to solving problems in the emerging applied disciplines usually overlooked by them and that specialists from applied sciences could pose new challenges for multiscale problems.

Numerous problems in natural sciences contain multiple scales: flows in complex heterogeneous media, many particles systems, composite media, etc. Mathematically, we are led to study of singular homogenisation limits and the procedure is called upscaling or homogenisation. The processes to be up-scaled are usually described by differential equations. For simple cases, when the differential equation is linear and the heterogeneities are periodic some progress has been made. However, most natural phenomena are described by nonlinear differential equations in a random nonhomogeneous medium and, despite an intensive development in recent years, there are many open problems.

The objective of the conference was to bring together leading specialists from Europe and the United States and to discuss new challenges in this quickly developing field. Topics of the conference were Nonlinear Partial Differential Equations and Applied Analysis, with direct applications to the modeling in Material Sciences, Petroleum Engineering and Hydrodynamics.

The Conference was co-organised by the Department of Mathematics of University of Zagreb, and the Inter-University Centre (IUC) in Dubrovnik, Croatia, where it took place from September 3rd to 9th, 2000. We hope that these Proceedings reflect the variety of subjects discussed; in order to provide a better overall view, we have reprinted the abstracts of those talks whose authors did not provide a full written account. Unfortunately, any attempt to convey the inspiring atmosphere of regular evening scientific discussions, or the beautiful surroundings of the ancient town and its neighbourhood, would be futile. We can only thank Mr. Zoran Jurović, a wine producer of *plavac mali* red wines from Postup, Orebić, for the contribution of his cellar to the atmosphere.

We would like to thank the members of the Local Organising Committee:

Mladen Jurak, University of Zagreb
Eduard Marušić-Paloka, University of Zagreb
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Josip Tambača, University of Zagreb
Marko Vrdoljak, University of Zagreb

for all the work they did. The conference was organised, hotels booked, etc., while none of us was in Croatia. In Dubrovnik, the staff of IUC provided excellent support and warm reception. Their support was complemented by a number of graduate students who helped with the registration, provided directions for sightseeing and boat trip, etc. In particular, Martin Lazar was responsible for the local contacts in his home town, while Krešimir Burazin recorded Prof. Tartar's lectures.

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For these Proceedings, we would like to thank those authors who found time and energy to write their contributions. Professor Tartar's lectures were recorded, typed, and then the real work began. In spite of his other obligations (and no promises made), he found time to make several revisions, and a number of pages were added. Marko Vrdoljak generated all the figures for these lecture notes, and caught many misprints. Finally, Josip Tambača put all the contributions together, ironed the differences and provided the final version of the manuscript. To Springer, many thanks for constant encouragement and assistance.

Nenad Antonić was in charge of the Proceedings and other editors would like to thank him for his effort.

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Mesoscopic Models of Reaction-Diffusion Processes with Exclusion Mechanism

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Abstract. We study a class of Markov jump processes describing reaction-diffusion phenomena in a dynamic medium. The model consists basically of a bounded domain divided into cells, each of them being the collection of a given number of sites. These sites are in one of two possible phases of the medium, being either empty, or occupied by particles. The cells are considered as homogenous, the spatial structure of the sites being ignored. The model describes chemical reactions and interactions with the medium, while the individual particles can also perform jumps between neighbouring cells. We consider an exclusion mechanism which prohibits adsorption or jump of particles in a saturated cell. We analyze the macroscopic behaviour of this class of processes in the limit when the cell size tends to 0 and the number of sites per cell tends to infinity, giving conditions for convergence towards the solution of a deterministic reaction-diffusion system.

Contents

1	Introduction	161
2	Construction of the stochastic model	162
3	Statement of the result	166
4	The proof	167
	References	173

1 Introduction

The model considered in this paper is closely related to a class of stochastic models for reaction-diffusion-type phenomena, known in the literature as 'density dependent models' or 'high density particle models'. Briefly, these models consist in the division of a bounded domain into cells by a regular mesh, while a large number of particles is considered inside each cell. Particles can perform random walks among neighbouring cells, or can react with a partner inside the same cell. In the works [1,7,2,3] the asymptotic properties of this class of Markov processes are analyzed, and convergence results towards a deterministic law (a reaction-diffusion system) are proved if the cell size tends to 0 and the number of particles tends to infinity. Of course, the results hold only under a suitable adjustment of the speed of convergence to 0 of the cell size and of the order of convergence to infinity of the number

of particles. In [5] an infinite-dimensional system of reaction-diffusion type is considered, namely the *coagulation-fragmentation equations*, and a convergence result of the same type as mentioned above is proved. In studying the macroscopic limits of this type of models, one can take advantage of probabilistic techniques, as well as of methods from the analysis. For this reason, this approach is often more convenient than an approach based on a lattice gas model, in which the particles perform random walks and where a reaction mechanism is present in the case when two particles meet on the same site. In the present work we consider a density-dependent particle model, with the modification that the number of particles in every cell is limited to a maximal number. However, in the limit it will tend to infinity. We consider particles which can perform random walks between the cells, while they also obey to a birth-death and a binary reaction mechanism, the only restriction being the limited maximal number of particles in each cell. The products of the binary reactions are not among the considered type of particles, and are eliminated instantaneously from the cell.

A further aspect is the modelling of a surface or a volume with crystalline structure as a *dynamic medium* which varies in time, exhibiting local phase changes caused by the mutual interaction between the medium and the particle system. The resolution of this model does not go down to the level of the atomic sites of the crystal surface, but considers the cells as being homogenous patches on a *mesoscopic scale*, in which the spatial distribution of the reactants and of the crystal phases is ignored. The motivation for this approach comes from modelling oxidation of **CO** on the crystalline surface of a catalyst, as treated e.g. in [8]. The simplified model presented in our approach is intended to be a first step in the rigorous mathematical modelling by stochastic processes of this type of phenomena.

2 Construction of the stochastic model

Let $\Omega \subset \mathbb{R}^d$, $d \leq 3$, be a bounded domain with smooth boundary, which we divide into cubic cells of size ε . Let \mathbb{G}_ε denote the collection of all cells which are *completely included* in Ω , and let $N_C = |\mathbb{G}_\varepsilon|$ denote their total number. Define

$$\Omega_\varepsilon := \bigcup_{G \in \mathbb{G}_\varepsilon} G \subset \Omega. \quad (1)$$

Consider particles which correspond to one of the reactants involved in the process. We associate to each cell a number of sites; each site can be either empty, or occupied by one particle. Moreover, each site is in one of the two possible phases of the medium. The sites are not a further subdivision of a single cell and their spatial structure is of no importance in this case. Due to this simplification, we call this type of model a *mesoscopic model*. Denote the set of all neighbours of a cell G which are contained in Ω_ε by $\mathbf{N}(G)$, and let $n(G) = |\mathbf{N}(G)|$. Let $n_G(t), m_G(t)$, denote respectively the number

of particles and the number of sites in a reference phase of the medium (denoted from now on as “phase 1”) considered in the cell G at time t . By multiplication with a scaling factor h , i.e. by defining $x_G^{h,\varepsilon}(t) = h \cdot n_G(t)$, we obtain the corresponding concentration. The factor h should be seen as the inverse of the ‘total number of sites’ in a cell. This implies that the concentration of the reactant cannot take values above 1. In a similar way we define $y_G^{h,\varepsilon}(t) = h \cdot m_G(t)$, which is the concentration of the sites in phase 1 in the cell G at time t . The pair $z_G^{h,\varepsilon}(t) := (x_G^{h,\varepsilon}(t), y_G^{h,\varepsilon}(t))$ is thus an element of the state space \mathbb{E} , which can be defined either as the finite-dimensional space of pairs of functions defined on Ω , piecewise constant on the cells in \mathbb{G}_ε , and zero in $\Omega \setminus \Omega_\varepsilon$, or as the space of $N_C \times 2$ matrices with real entries. In the first case, the values of the functions on the cell boundaries are of no importance, since we will work in an L^2 -setting. We will make selective use of these two viewpoints, depending on the context. For simplicity, where is no danger of confusion, the dependence of the state space on ε and h will be suppressed, since we will never consider two processes on two different state spaces at the same time. Denote further by E_G^x the $N_C \times 2$ matrix which has all elements equal to 0, except the element corresponding to the pair (G, x) which equals 1. Define further E_G^y in a similar way. The notation $\chi_A(\cdot)$ will stand as usually for the characteristic function of the set A . Consider some polynomial functions with positive coefficients $d(\cdot), a(\cdot)$ defined on $[0,1]$ and $q^1(\cdot, \cdot), q^2(\cdot, \cdot)$ defined on $[0,1]^2$. Let $z = (x, y) \in \mathbb{E}$ be the current state of the Markov process. The possible transitions are the following:

1. adsorption of new particles in an arbitrary cell G , at a rate proportional to the number of free sites, i.e.:

$$z \longrightarrow z + h \cdot E_G^x \text{ at rate } h^{-1} \cdot a(y_G) \cdot (1 - x_G) \tag{2}$$

2. disappearance of particles from an arbitrary cell G , i.e.:

$$z \longrightarrow z - h \cdot E_G^x \text{ at rate } h^{-1} \cdot d(y_G) \cdot x_G \tag{3}$$

This transition can occur as a consequence of desorption or reaction phenomena. Since we consider only one reactant and since the reaction products are leaving instantaneously the catalysator surface, we can describe the mentioned two processes with the same type of transition. Note that, as in the case of adsorption, the rates depend also on the medium, i.e. on the proportion of the two crystal phases inside the cell.

3. diffusion of particles inside the domain, i.e.:

$$z \longrightarrow z - h \cdot E_G^x + h \cdot E_{G'}^x \text{ at rate } \varepsilon^{-2} h^{-1} \cdot \chi_{[0,1]}(x_{G'}) \cdot x_G, \text{ where } G' \in \mathbf{N}(G). \tag{4}$$

The interpretation of this transition is the following: a particle jumps from the cell G to the neighbour cell G' with the given rate, under the

condition that there still are free sites available in the target cell. This is possible only if $x_{G'} < 1$. We have introduced in this way an *exclusion mechanism* into the model.

4. change of the crystal phase

$$\text{a) } z \longrightarrow z - h \cdot E_G^y \text{ at rate } h^{-1} \cdot q^1(x_G, y_G) \cdot y_G \quad (5)$$

$$\text{b) } z \longrightarrow z + h \cdot E_G^y \text{ at rate } h^{-1} \cdot q^2(x_G, y_G) \cdot (1 - y_G) \quad (6)$$

This transition models the change of phase of a site in the cell G (from the total number of h^{-1} sites in the cell), which can flip between phase 1 and phase 2. The terms $q^i(\cdot, \cdot)$, $i = 1, 2$, denote the flipping rates of one single site in phase i . The rates depend on both the concentration of the reactant and the proportion of the two phases. This model contains the possibility that only one site can flip at a time, but the results will hold also if we allow the flipping of a random number of sites, under the condition that the mean (expectation) of this random number is finite. However, for simplicity we will restrict ourselves only at the situation described above in 4.

The infinitesimal generator of the process $z(t)$ is given by:

$$(A_{h,\varepsilon}g)(z) = \sum_{\tilde{z}} (g(\tilde{z}) - g(z)) r_{z \rightarrow \tilde{z}} \quad (7)$$

for any bounded, measurable function g defined on \mathbb{E} , where the value of $r_{z \rightarrow \tilde{z}}$ runs through all previously described transition rates from the state z to the new state \tilde{z} . The parameter of the exponentially distributed waiting time before jumping from the state z to the state \tilde{z} is given by:

$$\lambda_{h,\varepsilon}(z) = \sum_{\tilde{z}} r_{z \rightarrow \tilde{z}} \quad (8)$$

The observation that the number of possible states of the process is finite implies that the waiting time parameter function $\lambda_{h,\varepsilon}$ is bounded. Then, according to [4], pp.162-164 the jump process $z(t)$ is defined a.s. for all $t \geq 0$, i.e. the jumps do not accumulate. Define for all $r > 0$ and $z = (x, y) \in \mathbb{E}$:

$$p_{G,r}^x(z) = \beta_r(x_G) \quad (9)$$

$$p_{G,r}^y(z) = \beta_r(y_G) \quad (10)$$

where the functions β_r are defined on \mathbb{R} for all $r > 0$ by:

$$\beta_r(w) = \begin{cases} w & \text{for } |w| \leq r \\ 2r \cdot \text{sgn}(w) - r^2 \cdot w^{-1} & \text{for } |w| > r. \end{cases} \quad (11)$$

$p_{G,r}^x, p_{G,r}^y$ are thus bounded C^1 -functions. From our construction of the process we have ensured that $x_G, y_G \leq 1$, so the functions $p_{G,1}^x, p_{G,1}^y$ are nothing

else but the projection of the component x_G , respectively y_G . The application of Dynkin's formula ([4], p.162) for these functions implies:

$$x(t) = x(0) + \int_0^t \{ \Delta_\varepsilon^* x(s) - d(y(s)) \cdot x(s) + a(y(s)) \cdot (1 - x(s)) \} ds + M_x^{h,\varepsilon}(t) \quad (12)$$

$$y(t) = y(0) + \int_0^t \{ q^2(x(s), y(s)) \cdot (1 - y(s)) - q^1(x(s), y(s)) \cdot y(s) \} ds + M_y^{h,\varepsilon}(t) \quad (13)$$

where the terms $M_x^{h,\varepsilon}(t), M_y^{h,\varepsilon}(t)$ are martingales with the respect to the filtration generated by the process. The operator Δ_ε^* corresponds to the diffusion-type transitions of the reactant, under the exclusion mechanism taken into consideration. We have:

$$\begin{aligned} \Delta_\varepsilon^* x_G &= \varepsilon^{-2} \sum_{G' \in \mathbf{N}(G)} \{ \chi_{[0,1]}(x_G) \cdot x_{G'} - \chi_{[0,1]}(x_{G'}) \cdot x_G \} \\ &= \chi_G \Delta_\varepsilon x_G - \Delta_\varepsilon \chi_G x_G \end{aligned} \quad (14)$$

The operator denoted by Δ_ε , is defined by:

$$\Delta_\varepsilon x_G = \sum_{J \in \mathbb{G}_\varepsilon, J' \in \mathbf{N}(J)} (-h \cdot E_J^* + h \cdot E_{J'}^*)_G \cdot \varepsilon^{-2} h^{-1} \cdot x_J \quad (15)$$

where E_J^* denotes the characteristic function of the cell $J \in \mathbb{G}_\varepsilon$, and will turn out to be the finite-difference approximation of the Laplace operator on an ε -mesh corresponding to Neumann boundary conditions. Let us present some further notations and auxiliary results concerning this operator. Let H^ε denote the Hilbert subspace of $L^2(\Omega)$ consisting of step functions which are constant on the cells $G \in \mathbb{G}_\varepsilon$ and vanish on $\Omega \setminus \Omega_\varepsilon$. For any element $f \in H^\varepsilon$, its norm is given by the L^2 -norm:

$$\|f\|_{H^\varepsilon}^2 = \sum_{G \in \mathbb{G}_\varepsilon} |G| \cdot f_G^2 = \varepsilon^d \sum_{G \in \mathbb{G}_\varepsilon} f_G^2 = \|f\|_{L^2(\Omega)}^2 \quad (16)$$

Where it is no confusion, by $\|\cdot\|_{L^2}$ we will mean always the norm in $L^2(\Omega)$. Define also the projections $\pi_\varepsilon : L^2(\Omega) \rightarrow H^\varepsilon$ by:

$$(\pi_\varepsilon f)_G = \varepsilon^{-d} \int_G f(x) dx \quad (17)$$

for any $G \in \mathbb{G}_\varepsilon$. It is easy to see that π_ε are continuous linear operators with norm 1, which implies that the internal approximation of $L^2(\Omega)$ by the spaces H^ε is stable. It can be easily seen that the approximation is also convergent, i.e.

$$\|\pi_\varepsilon f - f\|_{L^2} \rightarrow 0 \quad (18)$$

as $\varepsilon \rightarrow 0$, for any $f \in L^2(\Omega)$. Let Δ denote the L^2 -realization of the Laplace operator under Neumann boundary conditions, i.e. with the domain

$$H = \left\{ u \in H^2(\Omega) : \frac{\partial u}{\partial \nu} = 0 \text{ on } \partial\Omega \right\} \quad (19)$$

On the space H we have the norm:

$$\|u\|_H = \|u\|_{L^2(\Omega)} + \|\Delta u\|_{L^2(\Omega)}, \quad (20)$$

which is equivalent with the H^2 -norm (see [5]).

3 Statement of the result

The main result of this paper is the convergence of the family of stochastic processes to the solution of a deterministic reaction-diffusion system. Let Ω be a bounded domain in \mathbb{R}^d , $d \leq 3$ with smooth boundary and consider the polynomial functions with positive coefficients $d(\cdot), a(\cdot)$ defined on $[0, 1]$ and $q^1(\cdot, \cdot), q^2(\cdot, \cdot)$ defined on $[0, 1]^2$. Denote by $x(t, \cdot), y(t, \cdot)$ the solution of the following deterministic system:

$$\begin{aligned} \frac{dx}{dt} &= \Delta x(t) - d(y(t)) \cdot x(t) + a(y(t)) \cdot (1 - x(t)) \\ \frac{dy}{dt} &= -q^1(x(t), y(t)) \cdot y(t) + q^2(x(t), y(t)) \cdot (1 - y(t)) \text{ in } \Omega \\ \frac{\partial x}{\partial \nu} &= 0 \text{ on } \partial\Omega \end{aligned} \quad (21)$$

The initial conditions $x(0)$ and $y(0)$ are assumed to be sufficiently regular functions on Ω , positive and bounded strictly from above by 1. Global existence and uniqueness results of the above system are standard (see e.g. [9]). However, for our purpose it suffices to show the existence and uniqueness of a *mild solution* of the system. This will be pointed out subsequently. The regularity is only needed in order to apply a comparison principle for showing that the components of the solution stay in the range $[0, 1]$, if the initial conditions are in this range (in the case of our stochastic process, this is already ensured by construction). Moreover, due to the dissipativity of the diffusion operator, the component $x(t)$ will stay uniformly bounded away from 1.

Theorem 1. *If $h\varepsilon^{-2-d} \rightarrow 0$ as $h, \varepsilon \rightarrow 0$ and if*

$$\|x^{h,\varepsilon}(0) - x(0)\|_{L^\infty} + \|y^{h,\varepsilon}(0) - y(0)\|_{L^\infty} \rightarrow 0$$

in probability for $h, \varepsilon \rightarrow 0$, then the family of stochastic processes $z^{h,\varepsilon}(t) = (x^{h,\varepsilon}(t), y^{h,\varepsilon}(t))$ satisfies the convergence property:

$$\sup_{t \leq T} [\|x^{h,\varepsilon}(t) - x(t)\|_{L^2} + \|y^{h,\varepsilon}(t) - y(t)\|_{L^2}] \rightarrow 0$$

in probability as $h, \varepsilon \rightarrow 0$, for any $T > 0$ fixed.

4 The proof

For a better overview, we will divide the proof into several lemmata.

Lemma 1. *Let $S(t)$ be the contraction semigroup generated by the operator Δ with domain H on $L^2(\Omega)$. We have then: (i) If $u \in L^\infty(\Omega)$, then $S(t)u \in L^\infty(\Omega)$ and $\|S(t)u\|_{L^\infty} \leq \|u\|_{L^\infty}$. (ii) The operator Δ_ε generates a contraction semigroup S_ε on H^ε in the L^p -norm, for all $p \in [2, \infty]$. Moreover, if $p < \infty$, the semigroup is analytic. (iii) The semigroup $S_\varepsilon(t)$ on the space H^ε (considered as a subspace of $L^2(\Omega)$) satisfies the approximation property:*

$$\|S_\varepsilon(t)\pi_\varepsilon f - S(t)f\|_{L^2(\Omega)} \rightarrow 0 \quad \text{as } \varepsilon \rightarrow 0$$

uniformly in t on bounded intervals and for any $f \in L^2(\Omega)$. (iv) If $q(\cdot, \cdot)$ is a polynomial and u^1, u^2, v^1, v^2 are L^∞ -functions defined on Ω with norms ≤ 1 , then

$$\|q(u_1, u_2) - q(v_1, v_2)\|_{L^2} \leq M (\|u_1 - v_1\|_{L^2} + \|u_2 - v_2\|_{L^2})$$

where the constant M depends only on the coefficients and of the degree of the polynomial q .

Proof. For (i)-(iii) see [5], Lemma 2.2 and Lemma 3.1. (iv) The proof is a simple computation, which we will perform only in the case of monomials. The extension to general polynomials is straightforward. We have:

$$\begin{aligned} \|u_1^i u_2^j - v_1^i v_2^j\|_{L^2} &\leq \|u_1^i u_2^j - v_1^i u_2^j\|_{L^2} + \|v_1^i u_2^j - v_1^i v_2^j\|_{L^2} \\ &\leq \|u_1^i - v_1^i\|_{L^2} \|u_2^j\|_{L^\infty} + \|u_2^j - v_2^j\|_{L^2} \|v_1^i\|_{L^\infty} \\ &\leq M_1 (\|u_1 - v_1\|_{L^2} + \|u_2 - v_2\|_{L^2}) \end{aligned} \quad (22)$$

Since all L^∞ -norms of the considered functions are bounded from above by 1, the constant M_1 depends only on i and j . In the case of general polynomials, we simply sum up the results of the above computations and get the statement of the lemma. \square

Fix some time moment $T > 0$. Consider the Banach space $Z_T := \{z = (x(t), y(t)) \in L^\infty(\Omega) \times L^\infty(\Omega)\}$ with the norm

$$\|z\|_{Z_T} = \left\| \|x(t)\|_{L^\infty(\Omega)} + \|y(t)\|_{L^\infty(\Omega)} \right\|_{L^\infty[0, T]}$$

Define on Z_T the operator A by $(Az)(t) := ((Az)^x(t), (Az)^y(t))$, where

$$\begin{aligned} (Az)^x(t) &= S(t)x(0) + \int_0^t S(t-s) [-d(y(s))x(s) + a(y(s))(1-x(s))] ds \\ (Az)^y(t) &= y(0) + \int_0^t [-q^1(x(s), y(s))y(s) + q^2(x(s), y(s))(1-y(s))] ds \end{aligned} \quad (23)$$

Lemma 2. *For $r \leq 1$, the operator A defined above has a unique fixed point in the open ball $B_r(z(0))$ of the space Z_{T_1} , provided $T_1 = K^{-1}(1 + \|z(0)\|_{Z_{T_1}})^{-1}r$. K is a sufficiently large constant, depending only on the reaction functions and on the domain Ω . This is the mild (local) solution on the time interval $[0, T_1]$ of the system (21).*

Proof. The result follows from a straightforward application of Banach’s fixed point theorem, making use also of Lemma 1 (i). It is similar to [5] Theorem 1.1. The existence result is only local, since we did not impose positivity of the initial conditions. \square

Lemma 3. *(L^2 -estimate of the martingales) For a given $T > 0$ and for small ε , the martingales introduced in (12) and (13) satisfy the estimates:*

$$E_{z(0)} [\|M_x^{h,\varepsilon}(t)\|_{H^\varepsilon}^2] \leq CT \cdot h \cdot \varepsilon^{-2} \tag{24}$$

$$E_{z(0)} [\|M_y^{h,\varepsilon}(t)\|_{H^\varepsilon}^2] \leq CT \cdot h \cdot \varepsilon^{-2} \tag{25}$$

where the conditional expectation is taken with respect to the initial condition $z(0)$ of the process $z(\cdot)$ and where C is a constant depending only on Ω and of the coefficients of the transition rates.

Proof. We will prove only the first estimate, the computations for the second one being completely similarly (even simpler, since for this component the diffusion is absent). For any bounded C^1 -function g and for $z(t) = (x(t), y(t)) \in \mathbb{E}$ we have the standard identity (see e.g. [6]):

$$E_{z(0)} [g(M_x^{h,\varepsilon}(t))] = \int_0^t E_{z(0)} \left[\sum_{z \rightarrow \tilde{z}} \left(g(\tilde{x} - x + M_x(s)) - g(M_x(s)) - (\tilde{x} - x)g'(M_x^{h,\varepsilon}(s)) \right) r_{z \rightarrow \tilde{z}} \right] ds$$

where the notation $z \rightarrow \tilde{z} = (\tilde{x}, \tilde{y})$ has the same meaning as in (7). By taking $g = (p_{G,1}^x)^2$, we have then:

$$\begin{aligned} E_{z(0)} [\|M_x^{h,\varepsilon}(t)\|_{H^\varepsilon}^2] &= \varepsilon^d \sum_{G \in \mathbb{G}_\varepsilon} E_{z(0)} [M_{x,G}^{h,\varepsilon}(t)^2] = \\ &= \varepsilon^d \sum_{G \in \mathbb{G}_\varepsilon} \int_0^t \left\{ h^2 h^{-1} d(y_G) x_G + h^2 h^{-1} a(y_G)(1 - x_G) \right. \\ &\quad \left. + \sum_{J' \in \mathbf{N}(J)} [(-hE_J^* + hE_{J'}^*)_G]^2 \varepsilon^{-2} h^{-1} \chi_{[0,1]}(x_{J'}) x_J(s) \right\} ds \\ &= \varepsilon^d h \sum_{G \in \mathbb{G}_\varepsilon} \int_0^t \left\{ d(y_G) x_G + a(y_G)(1 - x_G) \right. \\ &\quad \left. + \varepsilon^{-2} x_G(s) \sum_{G' \in \mathbf{N}(G)} \chi_{[0,1]}(x_{G'}) + \varepsilon^{-2} \chi_{[0,1]}(x_G) \sum_{G' \in \mathbf{N}(G)} x_{G'}(s) \right\} ds \tag{26} \end{aligned}$$

Since $0 \leq x_G \leq 1$ for all G (ensured by definition), relation (26) implies immediately that the statement of the lemma is valid if we choose a sufficiently small ε . \square

Proof of Theorem 1 Throughout this proof, by *const* we will denote a generic constant, which is independent on h, ε, t and on the considered stochastic or deterministic processes. It may depend on the domain Ω and on the polynomial reaction functions. We will show first that the statement holds locally in time, more precisely on the time interval $[0, T_1]$ given by Lemma 2 for

$$r = 1 - \|x(0)\|_{L^\infty} \quad (27)$$

Define the stopping time

$$\tau^{h,\varepsilon} = \inf\{t \geq 0 : \|x^{h,\varepsilon}(t)\|_{L^\infty} = 1\} \wedge T_1 \quad (28)$$

That is, we stop the process as soon as we reach saturation in one of the cells or at the end of the considered time interval. Before this time moment, the exclusion property of the process does not show up, and we have just simple, discrete diffusion of the particles. For simplicity, we will denote from now on this stopping time as τ . From identity (12) we have thus:

$$\begin{aligned} x^{h,\varepsilon}(t \wedge \tau) - M_x^{h,\varepsilon}(t \wedge \tau) &= x^{h,\varepsilon}(0) + \int_0^{t \wedge \tau} \left\{ \Delta_\varepsilon(x^{h,\varepsilon}(s) - M_x^{h,\varepsilon}(s)) \right. \\ &\quad \left. + \Delta_\varepsilon M_x^{h,\varepsilon}(s) + a(y^{h,\varepsilon}(s)(1 - x^{h,\varepsilon}(s)) + d(y^{h,\varepsilon}(s))x^{h,\varepsilon}(s)) \right\} ds \end{aligned} \quad (29)$$

By variation of constants we have:

$$\begin{aligned} x^{h,\varepsilon}(t \wedge \tau) &= S_\varepsilon(t \wedge \tau)x^{h,\varepsilon}(0) + \int_0^{t \wedge \tau} S_\varepsilon(t \wedge \tau - s) \left\{ a(y^{h,\varepsilon}(s)(1 - x^{h,\varepsilon}(s)) \right. \\ &\quad \left. + d(y^{h,\varepsilon}(s))x^{h,\varepsilon}(s) + \Delta_\varepsilon M_x^{h,\varepsilon}(s) \right\} ds + M_x^{h,\varepsilon}(t \wedge \tau) \end{aligned} \quad (30)$$

Let $z = (x, y)$ be the unique fixed point of the operator A defined in (23), that is:

$$\begin{aligned} x(t \wedge \tau) &= S(t \wedge \tau)x(0) + \int_0^{t \wedge \tau} S(t \wedge \tau - s) \left[-d(y(s))x(s) \right. \\ &\quad \left. + a(y(s))(1 - x(s)) \right] ds \end{aligned} \quad (31)$$

$$y(t \wedge \tau) = y(0) + \int_0^{t \wedge \tau} \left[-q^1(x(s), y(s))y(s) + q^2(x(s), y(s))(1 - y(s)) \right] ds \quad (32)$$

Subtracting the equations (30) and (31), respectively (13) and (32) and taking L^2 -norms, we obtain:

$$\|x^{h,\varepsilon}(t \wedge \tau) - x(t \wedge \tau)\|_{L^2} \leq A_1^x + A_2^x + A_3^x \quad (33)$$

$$\|y^{h,\varepsilon}(t \wedge \tau) - y(t \wedge \tau)\|_{L^2} \leq A_0^y + A_1^y + A_2^y + A_3^y \quad (34)$$

where

$$A_1^x = \|S_\varepsilon(t \wedge \tau)x^{h,\varepsilon}(0) - S(t \wedge \tau)x(0)\|_{L^2} \quad (35)$$

$$A_2^x = \int_0^{t \wedge \tau} \left\| S_\varepsilon(t \wedge \tau - s) [a(y^{h,\varepsilon}(s))(1 - x^{h,\varepsilon}(s)) + d(y^{h,\varepsilon}(s))x^{h,\varepsilon}(s)] \right. \\ \left. - S(t \wedge \tau - s) [-d(y(s))x(s) + a(y(s))(1 - x(s))] \right\|_{L^2} ds \quad (36)$$

$$A_3^x = \sup_{t \leq T_1} \left\| \int_0^{t \wedge \tau} S_\varepsilon(t \wedge \tau - s) \Delta_\varepsilon M_x^{h,\varepsilon}(s) ds + M_x^{h,\varepsilon}(t \wedge \tau) \right\|_{L^2} \quad (37)$$

and

$$A_y^0 = \|y^{h,\varepsilon}(0) - y(0)\|_{L^2} \quad (38)$$

$$A_y^1 = \int_0^{t \wedge \tau} \left\| -q^1(x^{h,\varepsilon}(s), y^{h,\varepsilon}(s))y^{h,\varepsilon}(s) + q^1(x(s), y(s))y(s) \right\|_{L^2} ds \quad (39)$$

$$A_y^2 = \int_0^{t \wedge \tau} \left\| q^2(x^{h,\varepsilon}(s), y^{h,\varepsilon}(s))(1 - y^{h,\varepsilon}(s)) - q^2(x(s), y(s))(1 - y(s)) \right\|_{L^2} ds \quad (40)$$

$$A_y^3 = \sup_{t \leq T_1} \|M_y^{h,\varepsilon}(t \wedge \tau)\|_{L^2} \quad (41)$$

Let us estimate these terms, by using the properties of the operator semi-groups stated in Lemma 1.

$$A_x^1 \leq \|S_\varepsilon(t \wedge \tau)x^{h,\varepsilon}(0) - S_\varepsilon(t \wedge \tau)\pi_\varepsilon x(0)\|_{L^2} \\ + \|S_\varepsilon(t \wedge \tau)\pi_\varepsilon x(0) - S(t \wedge \tau)x(0)\|_{L^2} \\ \leq \|x^{h,\varepsilon}(0) - \pi_\varepsilon x(0)\|_{L^2} + \|S_\varepsilon(t \wedge \tau)\pi_\varepsilon x(0) - S(t \wedge \tau)x(0)\|_{L^2} \\ \leq \|x^{h,\varepsilon}(0) - x(0)\|_{L^2} + \|x(0) - \pi_\varepsilon x(0)\|_{L^2} \\ + \|S_\varepsilon(t \wedge \tau)\pi_\varepsilon x(0) - S(t \wedge \tau)x(0)\|_{L^2} \\ \leq \|x^{h,\varepsilon}(0) - x(0)\|_{L^2} + c_1(\varepsilon) \quad (42)$$

where, according to (18) and Lemma 1 (iii), $c_1(\varepsilon) \rightarrow 0$, as $\varepsilon \rightarrow 0$. Let us make the following notations:

$$D_{h,\varepsilon}(s) = a(y^{h,\varepsilon}(s))(1 - x^{h,\varepsilon}(s)) + d(y^{h,\varepsilon}(s))x^{h,\varepsilon}(s) \quad (43)$$

$$D(s) = a(y(s))(1 - x(s)) - d(y(s))x(s) \quad (44)$$

We have then:

$$A_x^2 \leq \int_0^{t \wedge \tau} \|S_\varepsilon(t \wedge \tau - s) [D_{h,\varepsilon}(s) - \pi_\varepsilon D(s)]\|_{L^2} ds \\ + \int_0^{t \wedge \tau} \|(S_\varepsilon(t \wedge \tau - s)\pi_\varepsilon - S(t \wedge \tau - s))D(s)\|_{L^2} ds$$

$$\begin{aligned}
 &\leq \int_0^{t \wedge \tau} \|D_{h,\varepsilon}(s) - \pi_\varepsilon D(s)\|_{L^2} ds \\
 &\quad + \int_0^{t \wedge \tau} \|(S_\varepsilon(t \wedge \tau - s)\pi_\varepsilon - S(t \wedge \tau - s))D(s)\|_{L^2} ds \\
 &\leq \int_0^{t \wedge \tau} \|D_{h,\varepsilon}(s) - D(s)\|_{L^2} ds + \int_0^{T_1} \|D(s) - \pi_\varepsilon D(s)\|_{L^2} ds \\
 &\quad + \int_0^{T_1} \|(S_\varepsilon(t \wedge \tau - s)\pi_\varepsilon - S(t \wedge \tau - s))D(s)\|_{L^2} ds \\
 &\leq \int_0^{t \wedge \tau} \|D_{h,\varepsilon}(s) - D(s)\|_{L^2} ds + c_2(\varepsilon)
 \end{aligned} \tag{45}$$

where, according to (18), Lemma 1 (iii), and the dominated convergence theorem, $c_2(\varepsilon) \rightarrow 0$, as $\varepsilon \rightarrow 0$. By Lemma 1 (iv), we have:

$$\|D_{h,\varepsilon}(s) - D(s)\|_{L^2} \leq \text{const} (\|x^{h,\varepsilon}(s) - x(s)\|_{L^2} + \|y^{h,\varepsilon}(s) - y(s)\|_{L^2}) \tag{46}$$

which yields:

$$A_x^2 \leq \text{const} \int_0^{t \wedge \tau} (\|x^{h,\varepsilon}(s) - x(s)\|_{L^2} + \|y^{h,\varepsilon}(s) - y(s)\|_{L^2}) ds + c_2(\varepsilon) \tag{47}$$

We will discuss now the term A_3^x using the framework considered e.g. in [7]. Let \mathcal{H} be a separable Hilbert space and M an \mathcal{H} -valued martingale which is square integrable, right continuous and has left limits (cadlag). Consider on \mathcal{H} a C_0 -semigroup of operators $U(t)$ which satisfies $\|U(t)\|_{L(\mathcal{H})} \leq \exp(\mu t)$, and let A be its infinitesimal generator. By partial integration we have:

$$\begin{aligned}
 \int_0^t U(t-s)AM(s)ds &= - \int_0^t \frac{d}{ds} (U(t-s)) M(s)ds = U(t)M(0) - M(t) \\
 &\quad + \int_0^t U(t-s)dM(s)
 \end{aligned} \tag{48}$$

The term $\int_0^t U(t-s)dM(s)$ is called *stochastic convolution-type integral* and can be regarded as a Stieltjes integral of the evolution operators $U(t-s)$ with respect to $M(\cdot)$. Moreover, cf. [7] p.179 it satisfies the following Doob-like inequality:

$$P \left(\sup_{t \leq T} \left\| \int_0^t U(t-s)dM(s) \right\|_{\mathcal{H}} \geq \delta \right) \leq \frac{4 \exp(4\mu T)}{\delta^2} E [\|M(T)\|_{\mathcal{H}}^2] \tag{49}$$

for all $0 \leq t \leq T$ and $\delta > 0$. Returning to our problem where $\mu = 0$, we define

$$\begin{aligned}
 Y_{h,\varepsilon}(t) &:= \int_0^{t \wedge \tau} S_\varepsilon(t \wedge \tau - s)\Delta_\varepsilon M_x^{h,\varepsilon}(s)ds + M_x^{h,\varepsilon}(t \wedge \tau) \\
 &= \int_0^{t \wedge \tau} S_\varepsilon(t \wedge \tau - s)dM_x^{h,\varepsilon}(s)
 \end{aligned} \tag{50}$$

By (49) and Lemma 3 we have thus for any $\delta > 0$:

$$P(A_3^x \geq \delta) = P(\sup_{t \leq T_1} \|Y_{h,\varepsilon}(t)\|_{H^\varepsilon} \geq \delta) \leq 4\delta^{-2} \text{const} T_1 h \varepsilon^{-2} \quad (51)$$

By Lemma 1 (iv), we have:

$$A_y^1 + A_y^2 \leq \text{const} \int_0^{t \wedge \tau} (\|x^{h,\varepsilon}(s) - x(s)\|_{L^2} + \|y^{h,\varepsilon}(s) - y(s)\|_{L^2}) ds \quad (52)$$

For $\delta > 0$, we have by Doob's inequality for the submartingale $\|M_y^{h,\varepsilon}(t \wedge \tau)\|_{H^\varepsilon}^2$ and by Lemma 3:

$$P(A_y^3 \geq \delta) \leq \frac{1}{\delta^2} E[\|M_y^{h,\varepsilon}(T_1)\|_{H^\varepsilon}^2] \leq 4\delta^{-2} \text{const} T_1 h \varepsilon^{-2} \quad (53)$$

Summing up the relations (33) and (34) and using the estimates (42), (47), (52) we obtain:

$$\begin{aligned} & \|x^{h,\varepsilon}(t \wedge \tau) - x(t \wedge \tau)\|_{L^2} + \|y^{h,\varepsilon}(t \wedge \tau) - y(t \wedge \tau)\|_{L^2} \leq c_1(\varepsilon) + c_2(\varepsilon) \\ & + A_x^3 + A_y^3 + \|x^{h,\varepsilon}(0) - x(0)\|_{L^2} + \|y^{h,\varepsilon}(0) - y(0)\|_{L^2} \\ & + \text{const} \int_0^{t \wedge \tau} (\|x^{h,\varepsilon}(s) - x(s)\|_{L^2} + \|y^{h,\varepsilon}(s) - y(s)\|_{L^2}) ds \quad (54) \end{aligned}$$

Gronwall's inequality implies:

$$\begin{aligned} & \|x^{h,\varepsilon}(t \wedge \tau) - x(t \wedge \tau)\|_{L^2} + \|y^{h,\varepsilon}(t \wedge \tau) - y(t \wedge \tau)\|_{L^2} \leq (c_1(\varepsilon) + c_2(\varepsilon) \\ & + A_x^3 + A_y^3 + \|x^{h,\varepsilon}(0) - x(0)\|_{L^2} + \|y^{h,\varepsilon}(0) - y(0)\|_{L^2}) e^{\text{const} T_1} \quad (55) \end{aligned}$$

The convergence to 0 of $c_1(\varepsilon)$, $c_2(\varepsilon)$, relations (51), (53) and the hypotheses of the theorem imply that

$$\sup_{t \leq T_1} (\|x^{h,\varepsilon}(t \wedge \tau) - x(t \wedge \tau)\|_{L^2} + \|y^{h,\varepsilon}(t \wedge \tau) - y(t \wedge \tau)\|_{L^2}) \longrightarrow 0$$

in probability as $h, \varepsilon \rightarrow 0$. We will show now that the stopping time τ will be with high probability equal to T_1 , meaning that in the limit the saturation is not reached. Let us analyze the event $\{\tau < T_1\}$, that is, the situation when the saturation is reached before the end of the time interval. Considering now in relation (12) $t = \tau$, taking L^∞ -norms and using also Lemma 1 (ii), we have:

$$\begin{aligned} 1 & = \|x^{h,\varepsilon}(\tau)\|_{L^\infty} \leq \|x^{h,\varepsilon}(0)\|_{L^\infty} + \|Y_{h,\varepsilon}(\tau)\|_{L^\infty} + \int_0^\tau \|D_{h,\varepsilon}(s)\|_{L^\infty} ds \\ & \leq \|x(0)\|_{L^\infty} + \|x^{h,\varepsilon}(0) - x(0)\|_{L^\infty} + \varepsilon^{-d/2} \|Y_{h,\varepsilon}(\tau)\|_{L^2} + K\tau \quad (56) \end{aligned}$$

Where the constant K is defined in Lemma 2. Together with (27), this implies

$$KT_1 > K\tau \geq r - \|x^{h,\varepsilon}(0) - x(0)\|_{L^\infty} - \varepsilon^{-d/2} \|Y_{h,\varepsilon}(\tau)\|_{L^2} \quad (57)$$

Using the relation between T_1 and r from Lemma 2 we get:

$$K\|z(0)\|_{Z_{T_1}} T_1 \leq \|x^{h,\varepsilon}(0) - x(0)\|_{L^\infty} + \varepsilon^{-d/2} \|Y_{h,\varepsilon}(\tau)\|_{L^2} \quad (58)$$

Denote the event of this inequality by \mathcal{E} . We have shown that

$$\{\tau < T_1\} \implies \mathcal{E}$$

and thus

$$P\{\tau < T_1\} \leq P\{\mathcal{E}\}$$

But, by the hypothesis of the theorem together with (51),

$$P\{\mathcal{E}\} \rightarrow 0 \text{ as } h, \varepsilon \rightarrow 0$$

This shows that the convergence property holds on the entire time interval $[0, T_1]$. Since we considered a solution of (21) which, by the assumption on the reaction functions, stays uniformly bounded away from 1, we have $\|x(t)\|_{L^\infty} \leq 1 - \kappa$ for all $t \in [0, T_1]$, for some positive constant κ . This implies that $r \geq \kappa$ and that $1 - \|x(T_1)\|_{L^\infty} \geq \kappa > 0$. We can now repeat the construction from Lemma 2 starting from the initial value $(x(T_1), y(T_1))$ and obtain a prolongation of the solution on the interval $[T_1, T_2]$ with $T_2 = T_1 + K^{-1}(1 + \|z(T_1)\|_{Z_{T_2}})^{-1}(1 - \|x(T_1)\|_{L^\infty})$. We have then $T_2 - T_1 \geq (K/3)^{-1}\kappa$. Applying this procedure repeatedly, we obtain a solution on any arbitrary finite interval $[0, T]$, since the local existence intervals do not shrink. In every step we repeat the argumentation for the stochastic convergence which we made for the interval $[0, T_1]$ and cover thus the whole interval $[0, T]$, finishing the proof of the theorem. \square

References

- [1] Arnold, L., Theodosopulu, M. (1980) Deterministic limit of the stochastic model of chemical reactions with diffusion. *Adv.Appl.Prob.* **12**, 367-379
- [2] Blount, D. (1994) Density dependent limits for a nonlinear reaction-diffusion model. *Ann.Probab.* **22** No.4, 2040-2070
- [3] Blount, D. (1996) Diffusion limits for a nonlinear density-dependent space-time population model. *Ann.Probab.* **24** No.2, 639-659
- [4] Ethier, S., Kurtz, T.G. (1986) *Markov Processes: Characterization and Convergence.* J.Wiley & Sons, New York
- [5] Guias, F. (2001) Convergence properties of a stochastic model for coagulation-fragmentation processes with diffusion. *Stochastic.Anal.Appl.* **19** No.2, 245-278
- [6] Guias, F. (1997) A Monte Carlo approach to the Smoluchowski equations. *Monte Carlo Meth.Appl.* **3** No.4, 313-326
- [7] Kotelenetz, P. (1986) Law of large numbers and central limit theorem for linear chemical reactions with diffusion. *Ann.Probab.* **14** No.1, 173-193
- [8] Reichert, C., Starke, J., Eiswirth, M. (2001) *Stochastic Modelling of CO-Oxidation on Platinum Surfaces and Deterministic Limit.* (Preprint)
- [9] Smoller, J. (1983) *Shock Waves and Reaction-Diffusion Equations.* Springer