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A cellular automata approach to chemical reactions; 1 Reaction controlled systems

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HIGHLIGHTS

- Cellular automata approach is suitable to simulate the chemical controlled reactions.
- Therefore a modification of dissolution routine of CEMHYD3D has been introduced.
- A general and original linear relation between time and cycles was derived.

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ABSTRACT

A direct link between the chemical reaction controlled (shrinking core) model and cellular automata, to study the dissolution of particles, is derived in this paper. Previous research on first and second order reactions is based on the concentration of the reactant. The present paper describes the reaction kinetics based on particles and takes into account the shape and specific surface of these particles.

As a vehicle for the present study of cellular automata, a simplified version of the CEMHYD3D model is used. During the research it was found that during the dissolution of particles, additional reactive surface was created due to the dissolution of voxels in the middle of the top-surfaces. Therefore a modification of the dissolution routine within CEMHYD3D was introduced. This modification introduced the preference of the system to dissolve voxels on the outside of the particles rather than the middle of the top-surfaces of the particles. In this way the increase of reactive surface is prohibited and a spherical shape maintained.

Using this modification, it is proven that the dissolution of digitized particle can be describe based on the chemical reaction controlled system. Based on 165 simulations a general linear relation between cycles and time was derived. The derived model can describe the reaction sufficient up to 99.9%. Therefore it can be concluded that the single 'cellular automata' particle unambiguously related to the chemical controlled reactions.

1. Introduction

Chemical reactions are studied since centuries. Reactions can be described based either on concentration or on particle geometry. Levenspiel [1,2] and Szekely et al. [3], besides others, describe the progression of chemical reactions based on the particle geometry. From the described reactions, the chemical reaction controlled shrinking core model is the most simple reaction [2]. This reaction is relevant for particles and used for instance to describe the burning of carbon in air. The reaction kinetics are usually analyzed using numerical and analytical models. Besides these models also other methods have become available such as cellular automata (CA).

Cellular automata are already commonly used for the modeling of chemical systems [4,5]. Cellular automata systems are among others applied for studying biology, cement reaction, population growth, computability theory, mathematics, physics, complexity science, theoretical biology and microstructure modeling. Kier et al. [6] uses cellular automata to describe the percolation within chemical systems. Furthermore Kier and co-authors described a broad range of chemical systems and processes in which the cellular automata approach can be applied, ranging from aqueous systems (water), dissociation of organic acid in solutions and bond interactions [7–14]. Berryman and Franceschetti [15] described the simulation of diffusion controlled reaction kinetics using cellular automata. Kier and Cheng [13] gave a cellular automata model for dissolution. Goetz and Seetharaman [16] used cellular automata to describe the static recrystallization kinetics with homogeneous and heterogeneous nucleation. Liu et al. [17] and Raghavan and Sahay [18,19] simulated grain growth using cellular automata. Dab et al. [20] used a cellular-automaton for reactive systems containing particles. Zahedi Sohi and Khoshandam [21] used 2D

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cellular automata to describe non-catalytic gas–solid reactions of pellets consisting out of agglomerated grains with diffusion through a porous product layer and reaction on the surface of grains. In their work the cells were the grains and the conversion per cell was considered. This paper considers spheres in 3D which consists of several cells, being either product, reactant or void. Kier and Seybold and their co-authors [22–27] did research to the application of cellular automaton systems to describe chemical reactions. Their work describes first and second order reactions as well thermodynamically controlled reactions [26] and is closely related to our work. Their research on first and second order reactions is based on the concentration of the reactant, while in this research the shape is taking in account as well as the specific surface of the dissolving particle. Hence, this article considers the reaction as surface-based rather than volume-based.

A rigorous link between the (mathematical) chemical reaction models, like chemical reaction (shrinking core) model and the cellular automata approach to study the dissolution of particle has, to the authors' knowledge, not been achieved yet.

2. Chemical reaction kinetics

Within the chemical reaction kinetics literature, two main reaction model families are distinguished the progressive-conversion model (PCM) and the shrinking unreacted core model (SCM). Within the progressive-conversion model (sometimes referred to as the zone reaction model [28]), the solid reactant is converted continuously and progressively throughout the particle, while within the shrinking core model, the reactions take place on the outer skin of the particle and the zone of reaction is moving into the solid leaving behind completely converted material and inert solids. The main assumption within the shrinking core model is that the zone of reaction is narrowly confined to the interface between the unreacted solid and the product, when the chemical reaction is very rapid and the diffusion of the fluids sufficiently low, for instance due to a low void fraction of the unreacted material [29]. If on the other hand the solid contains enough voids so that the fluid reactants can diffuse freely into the interior of the solid, the reactions between fluid and solid may be regarded as occurring homogeneously throughout the entire solid, this is described using the progressive conversion model [29].

Levenspiel [1] points out that in practice in most cases the shrinking core model approximates real particles more closely than the progressive conversion model does, as shown in a wide variety of situations. But in fact the PCM and SCM describe the two limiting cases of the reality and a combination of both models should be applied [29]. Ishida et al. developed a more general model for solid–gas reactions, which takes into account these two phases of reaction [28,30]. They presented the shrinking unreacted core model as a special case of the progressive conversion model.

Yagi and Kunii [31–33] distinguish five steps occur during reaction succession;

- Diffusion of gaseous reactant A through the film surrounding the particle to the surface of the solid.
- Penetration and diffusion of A through the blanket of ash to the surface of the unreacted core.
- Reaction of gaseous A with solid at this reaction surface.
- Diffusion of gaseous products through the ash back to the exterior surface of the solid.
- Diffusion of gaseous products through the gas film back into the main body of fluid.

Based on these reaction steps three resistance to reaction can be distinguished namely, film diffusion, ash diffusion and reaction controlled (surface area controlled). The highest resistance forms the rate-controlling step. An important parameter in the use of the shrinking core model is the assumption of unchanging or changing particle size, since this determines the available reaction surface. The ash diffusion model is only available for unchanging particle size, assuming the amount of product formed equals the amount of reactant used during reaction. The chemical reaction controlled 'model' in both cases uses the surface of the unreacted core and will therefore in both cases decrease. The main difference can be found in the film diffusion system. In the unchanging particle size case the surface area of the original particle is used, while in the changing particle size case the surface area of the unreacted core is used.

In the present paper, the surface-controlled system is studied. Both Levenspiel [1,2] and Szekely et al. [3] call this system chemical reaction controlled (CRC). The progress of the reaction within the reaction controlled model is unaffected by the presence of any ash/product layer, therefore the reaction rate is proportional to the available surface of the unreacted core.

$$\frac{1}{S_c}\frac{dV_c}{dt}=k \tag{1}$$

with k being the first-order rate constant for the surface reaction, S_c the surface area of unreacted core and V_c as the unreacted core volume. Using the standard equations for the volume and surface dependence on the radius of unreacted core (r_c), Eq. (1) can be rewritten to

$$\frac{dr_c}{dt}=k \tag{2}$$

Eq. (2) can be integrated and applying

$$r_c(t=0)=r_0 \tag{3}$$

results in

$$t=\frac{1}{k}(r_0-r_c) \tag{4}$$

The time τ required for complete conversion, i.e. when $r_c = 0$, reads

$$\tau=\frac{r_0}{k} \tag{5}$$

The decrease in radius or increase in fraction conversion of the particle in terms of τ is obtained by combining Eqs. (4) and (5)

$$\frac{t}{\tau}=1-\frac{r_c}{r_0} \tag{6}$$

This can be rewritten based on the reaction degree, which is described by the volume of unreacted core (V_c) and the initial volume of the particle (V_0).

$$\alpha=1-\frac{V_c}{V_0}=1-\left(\frac{r_c}{r_0}\right)^3 \tag{7}$$

Eqs. (6) and (7) can be combined to a general equation for the reaction controlled system, which reads

$$\frac{t}{\tau}=1-(1-\alpha)^{1/3} \tag{8}$$

In the next sections, cellular automata parameters such a particle geometry, dissolution probability and reaction time are related to this chemical reaction controlled model.

3. Cellular automata

Cellular automata (CA) were first proposed by Ulam [34,35] and Von Neumann [36], based on ideas of Zuse [25]. The first full comprehensive description of cellular automata was made by Wolfram [37]. Cellular automata are *'simple mathematical idealizations of natural systems. They consist of a lattice of discrete identical sites, each site taking on a finite set of integer values. The values of the sites evolve in discrete time steps according to deterministic rules that specify the value of each site in terms of the values of neighboring sites. Cellular automate may thus be considered as discrete idealization of the partial differential equations often used to describe natural systems'* [37,38]. The system consisting of triangles, squares, hexagons or other shapes cell in a one (1D), two (2D) or three-dimensional (3D) grid/array. The 2D square cells are used mostly in the last 50 years [25]. The first use of grids for study micro-structure of metals was done by Rosenhain [39]. Often the cellular automaton systems apply periodic boundaries, which mean that a cell moving off the system at the edge, reappears from the opposite edge to the grid, in order to avoid wall side effects.

According to Kier et al. [5,25], cellular automata are not restricted to only the use of deterministic rules, since probabilistic rules are used extensively for studying real physical and chemical systems. Kier et al. [5] point out that CA-models have the following four components;

- A grid composed of cells.
- A set of ingredients.
- A set of local rules governing the behaviors of the ingredients.
- Specified initial conditions.

In this paper a well known cellular automata model is used to study particle reaction kinetics (presented in Section 2). The CEMHYD3D-model [40–43], originally developed by NIST [40], present the microstructure of hydrating cement by using a $100 \cdot 100 \cdot 100$ box with 10^6 voxels of size $1 \cdot 1 \cdot 1$. Each voxel represents a chemical substance either fresh binder or hydrated material. The initial microstructure thus consists of cement particles and water, where each particle consists of a number of voxels. The particles are created by placing voxels in an approximately spherical shape called "digitized particle" within the box. A 1-μm particle in CEMHYD3D is represented by 1 voxel at a system resolution of 1 μm, while at a system resolution of 0.33 μm it is represented by 19 voxels with size of 0.33 μm. An example for the digitized particle of size 3, 5, 7, 11 and 21 voxels is shown in Fig. 1a, which contain 19, 81, 179, 739 and 4945 voxels, respectively [41].

The first step of the simulation is the creation of an initial microstructure, which representing the fresh paste. Therefore the particle size distribution and the chemical composition of the binder is taking into account. Based on the chemical composition, the (cement clinker) phases are assigned randomly to the (solid) voxels. This results in a homogeneous phase distribution throughout the microstructure. The second step of the simulation is the performance of the chemical reactions of clinker phases and dihydrate by using the cellular automation and random walk algorithms. The disadvantage of using a box is the presence of wall-effects. In order to eliminate these effects, periodic boundaries are applied within CEMHYD3D [44].

Reaction of the initial microstructure is simulated by using discrete cycles. Each cycle consists of three processes: dissolution, diffusion and reaction, which are performed in serie (Fig. 2). The dissolution step is performed by checking each voxel in the box, for its ability to dissolve. This ability depends on the distance of the voxel to the void fraction and if the phase is soluble at that moment in time. During the induction-time of the cement, some cement clinker phases are assumed not to be soluble. If a voxel is

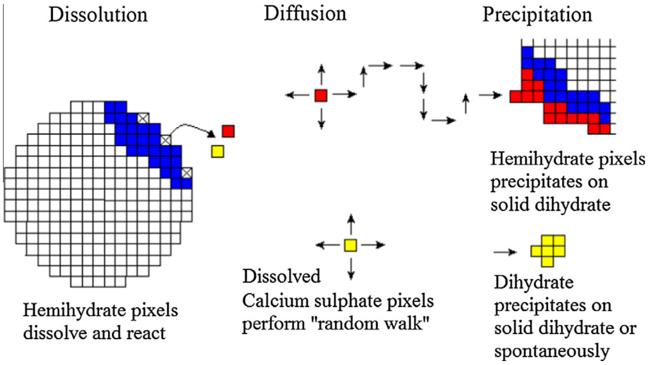

Fig. 2. Main reaction mechanism CEMHYD3D (modified from Bentz and Garboczi [45] and Van Eijk [41]).

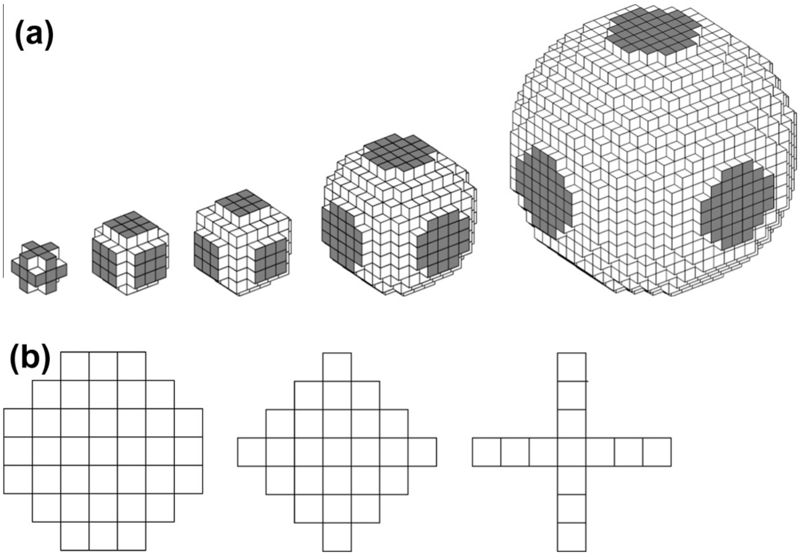

Fig. 1. (a) Digitized particle with a sizes of 3, 5, 7, 11 and 21 voxels with in gray indicated the top-surfaces and (b) a example of 2D-particle with the same perimeter and radius but with different surface area.

actually dissolving during a certain cycle, depends on the dissolution probability. The dissolution probability is defining by Bentz and Garboczi [45] as the likelihood of each phase to dissolve. Therefore a random number is generated which needs to be higher than the dissolution probability. The probability for the different (chemical) phases of the binder differs in order to model the different dissolution rates of these cement phases. The model corrects for the volume changes during reaction by adding one or more diffusing voxels. Due to the discrete behavior of cellular automata the volume stoichiometry of the reactions is preserved statistically.

The microstructure is updated after each reaction cycle. Reaction related properties, i.e. the reaction degree, are calculated after each cycle. The overall reaction degree of the binder is calculated on volume basis by diving remaining binder voxels by the total numbers of initial binder voxels. This can be converted into the degree on a mass basis by using the densities of phases. For this paper only one cement clinker phase is used and therefore both reaction degrees are equal.

4. Simulation results

Using simulations, the cycle in a certain α is reached has been determined for a dissolution probability (P_0) of 0.003–0.09 with steps of 0.003 and d = 7, 13, 21, 25 and 35. Analysis of the simulation data shows that the reaction rate depends on the available surface area of the reactant. Based on the description in the previous section, the chemical reaction controlled (CRC) model is most appropriate to describe a chemical system without precipitation of reaction products and the existence of a diffusion layer.

However when assessing the available surface area within CEMHYD3D during reaction, as shown in Fig. 3, one can notice an increase in available surface area rather than a decrease, as would be expected. This surface increase is caused by the dissolution of voxels from the middle of the flat top surface of the particles (indicated in gray in Fig. 1a), rather than the edge of the flat top surfaces. This results in a loss of the spherical shape and the creation of additional (reactive) surface area. Fig. 4a shows the cross-section of 35 particle during dissolution. From this figure, one can indeed conclude that the spherical shape is lost and that holes seems to be formed in the cross-section.

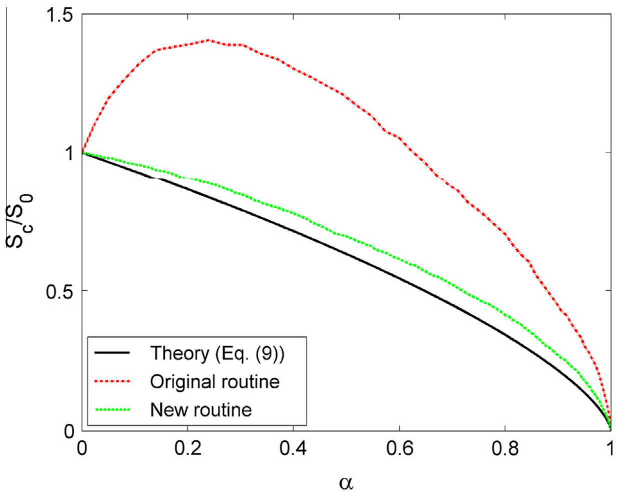

Fig. 3. The available reactive surface during reaction in CEMHYD3D using the original and new dissolution routine compared to the theoretical surface (Eq. (9)) according to chemical reaction controlled model of Levenspiel [1].

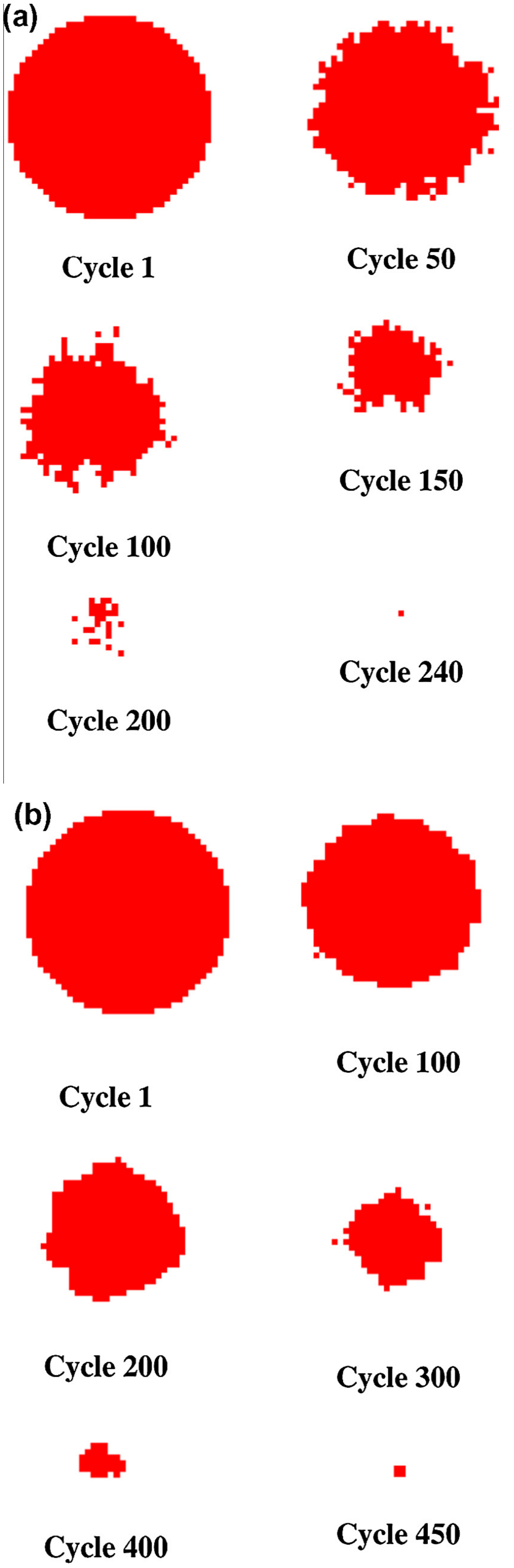

Fig. 4. Example of a dissolving d_p = 35 particle and the developing shape during its dissolution (in red/black the unreacted voxels) using (a) the original and (b) new dissolution routine. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Therefore a new routine has been added to the dissolution module of CEMHYD3D. Within this routine the determination whether a voxels dissolves or not (the probability part) is done in the regular way as done in CEMHYD3D. In case the voxels will dissolve, the position of the voxels is checked. When the number of surfaces of the particular voxel exposed to the pore solution is three or higher the voxel will dissolve immediately. Otherwise the edge of the particle is searched and a voxel at this edge will dissolve instead. Whenever a voxel has 3 free neighboring voxels, it will dissolve. Fig. 1b shows three particles in 2D with the same perimeter but with different surfaces. For 3D comparable images can be produced. Due to the way the dissolution module is programmed it is always searching for the outer pixels (it is so programmed that an increase of the external surface is prohibited). Fig. 3 shows the dissolution of particle of size 35 using both the original and the new routine. As one can notice the particle in the original routine dissolves faster than the new routine. This is caused by higher reactive surface area, which leads to an increase of reaction speed in the system. Furthermore, a particle dissolving using the new routine retains its spherical shape (Fig. 4b).

Fig. 3 also shows the result of applying the new routine at the dissolution of a 35-voxel particle. As can be noticed the surface-decrease during reaction is much closer to the theoretical surface decrease for a shrinking core. The theoretical dependence of the specific surface area on the reaction degree reads as

$$\frac{S_c}{S_0} = \left(\frac{r_c}{r_0}\right)^2 \tag{9}$$

with S_0 the initial surface of the digitized particle. Combining Eqs. (7) and (9), this results into

$$S_c = (1-\alpha)^{2/3} S_0 \tag{10}$$

Based on Fig. 3, one can notice that the surface area of the dissolving particles is slightly higher than one should expect based on these general models. According to this models a relation between available reactive surface area and reaction degree should exist.

Fig. 5 shows the results of the different particle radius for $p = 0.021$. The reaction degree of smaller particles ($d_p < 35$) has been recalculated so their starting point corresponds to moment that a 35 particle reach the same volume and the curves have been scaled. This enables a direct comparison of the curves. From the figure one can notice that the reaction rate indeed depends on the particle size.

5. Coupling cellular automata and reaction kinetics

Since the resemblance between the reaction curves obtained from the CEMHYD3D and the reaction controlled curves as, in this section a correlation between simulations and theoretical model is researched. Therefore a linear regression between cycle number

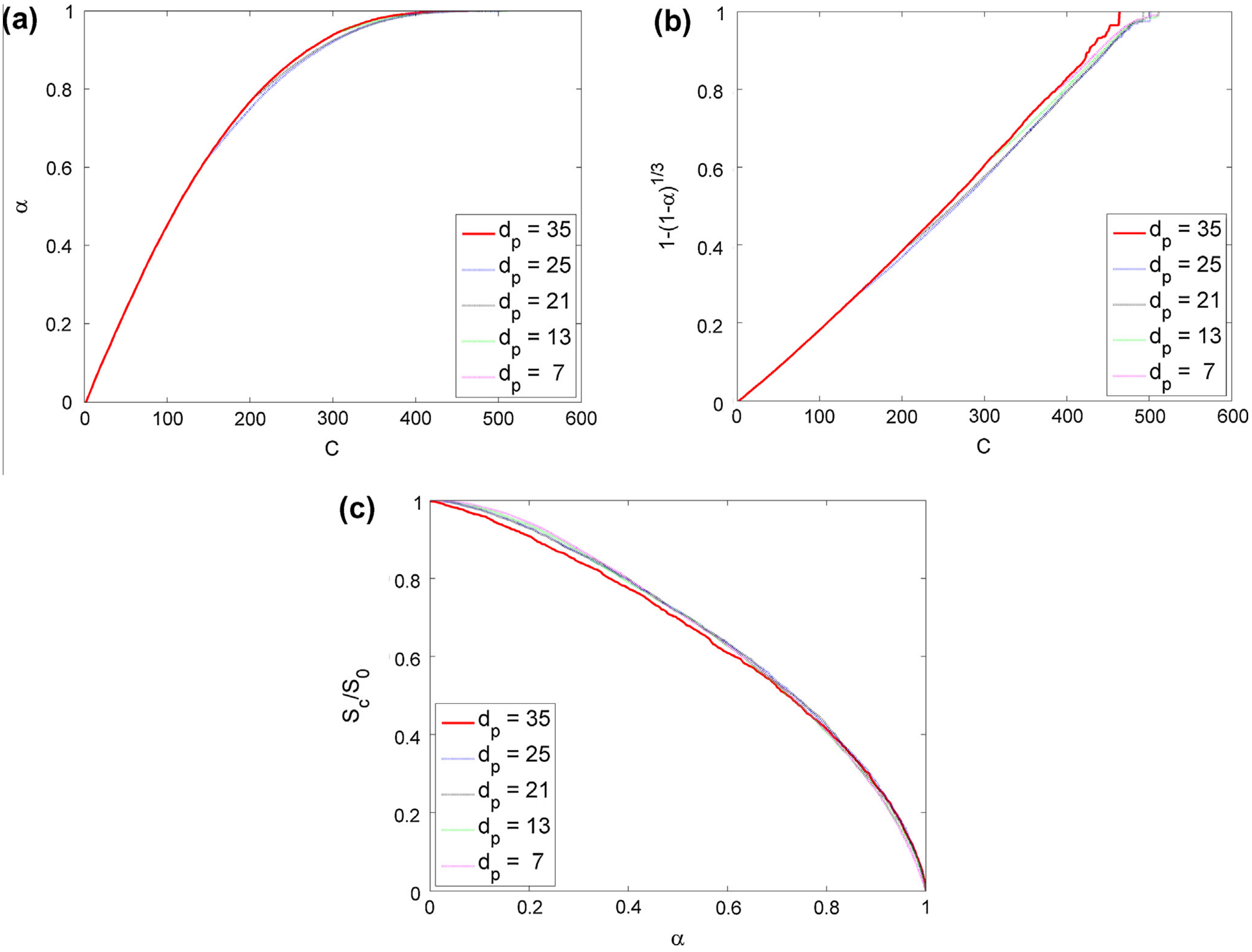

Fig. 5. Partial reaction of particles for $P_0 = 0.021$ (a) relation between cycles (C) and reaction degree (α), (b) the relation between cycles and chemical reaction model and (c) the relation between reaction degree and surface fraction of unreacted core (S_c/S_0).

(C-2) and $(1-(1-\alpha)^{1/3})$ is tested, since according to the chemical reaction controlled model a linear relation between time and $(1-(1-\alpha)^{1/3})$ exists and linear relation between cycle and time is assumed here. The relation with C-2 rather with C is used because the reaction within CEMHYD3D always start not earlier than cycle 2, as described earlier in this paper. Fig. 6 shows the simulation of three reaction curves scaled both on the x- and y-axis. As one can notice the scaled curves are similar and can be described with the following equation;

$$\left(1-(1-\alpha)^{\frac{1}{3}}\right)=\beta\frac{(C-2)\cdot P_0}{r_p} \tag{11}$$

with β the fitting parameter, r_p the radius of particle size in voxels and P_0 the dissolution probability. Note that the here α is based on the volume of reacted voxels in the cellular automata system. Using the fitting toolbox of Matlab® the β has been determined for all reaction curves with $d = 7$, 13, 21, 25, 35 and $P_0 = 0.003$–0.09 for $0.05 < \alpha < 0.9$. Table 1a shows the statistical data on found values of β. A mean (μ) of 1.622 was found with a standard deviation (σ) of 0.059, i.e. a coefficient of variation of 0.036, which is a satisfactory outcome. When determining the β of ten simulations with the same initial microstructure, $P_0 = 0.003$ and $d = 35$, we found a standard deviation (σ) of 0.015. This statistical variation for identical systems is inherent on cellular automata and constitutes already 25% of the variation found for β of the whole range of diameters and probabilities. The other statistical data on these simulations can be found in Table 1b.

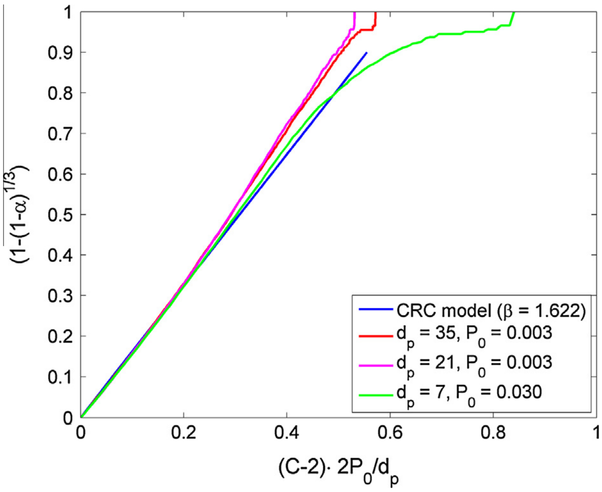

Fig. 6. The relation between $(C\text{-}2)\cdot 2P/d_p$ and $1-(1-\alpha)^{1/3}$ (with $\beta = 1.622$) for three simulations and the reaction controlled processes given by Levenspiel [1].

Table 1
(a) Statistical information on the derived values of β for $d = 7$, 13, 21, 25 and 35 and $P_0 = 0.003$–0.099 and (b) statistical information on derived values of β for 10 simulations with $d = 35$ and $P_0 = 0.003$ and $d = 35$ and $P_0 = 0.021$.

Parameter	Value
1a	
Average	1.622
Median	1.618
Standard deviation (σ)	0.059
Variance (σ^2)	0.003
Coefficient of variation	0.036

Parameter	$P_0 = 0.003$	$P_0 = 0.021$
1b		
Average	1.699	1.664
Median	1.704	1.664
Standard deviation (σ)	0.015	0.008
Variance (σ^2)	0.000	0.000
Coefficient of variation	0.009	0.005

Fig. 6 shows the results from the model compared to the (original) simulation curves for three different cases. From Fig. 6, one can notice that the assumption for linearity is valid for $1-(1-\alpha)^{1/3} < 0.9$, especially for slow reaction curves. This range of applicability corresponds to a reaction degree of 0.999. From the reaction curves (Fig. 6), one can notice that the model is sufficiently accurate to describe the reaction curve produced using CEMHYD3D.

In order to derive a mathematical correlation between the chemical reaction controlled model and the cellular automata simulations, Eqs. (5) and (8) can be combined rewritten as

$$\left(1-(1-\alpha)^{\frac{1}{3}}\right)=\frac{t\cdot k}{r_0} \tag{12}$$

Combining Eqs. (11) and (12), the following relation between time and cycles is derived,

$$t=b\frac{P_0}{k}\cdot\frac{r_0}{r_p}\cdot(C-2) \tag{13}$$

Hence, a universal and linear relation is established between cellular automata cycles (C) and digital particle size (d_p) and dissolution probability (P_0) on the one hand, and time for given initial sphere size of a reacting particle r_0 and reaction constant k. So, a direct relation is obtained between cellular automata parameters and the governing parameters of chemically controlled reaction systems. Furthermore, it can be seen that the P_0 and digitized particle size scale proportionally.

6. Conclusions

In this paper, the relation between the theoretical shrinking core reaction kinetic equations, as presented by Levenspiel [1,2], Szekely et al. [3] and others, and a cellular automata reaction approach has been investigated. As cellular automata tool CEMHYD3D was employed. In order to derive this relation a major improvement/modification was introduced. This modification introduced the preference of the system to dissolve voxels on the outside of the particles rather than the middle of the top-surfaces of the particles. In this way a spherical shape is maintained, which could not be avoided in the original version of the dissolution routine.

Using these modification, it is proven that a cellular automata approach is suitable to simulate the chemical reaction controlled model. Based on 165 simulations using a wide variation of dissolution probabilities (P_0) and particle diameters (d_p) a general and original linear relation between cycles and time was derived (Eq. (13)). The derived model can describe the reaction sufficiently accurate up to a reaction degree of 99.9%.

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