Lattice Boltzmann simulation of the surface growth effects for the infiltration of molten Si in carbon preforms

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The infiltration of molten silicon into carbon preforms is a widespread technique employed in the industry in order to enhance the thermal and mechanical properties of the final ceramic products. A proper understanding of this phenomenon is quite challenging since it stems from the reciprocal action and reaction between fluid flow, the transition to wetting, mass transport, precipitation, surface growth as well as heat transfer. As a result, the exhaustive modeling of such problem is an involved task. Lattice Boltzmann simulations in 2D for capillary infiltration are carried out in the isothermal regime taking into account surface reaction and subsequent surface growth. Precisely, for a single capillary in the linear Washburn regime, special attention is paid to the retardation for the infiltration process induced by the thickening of the surface behind the contact line of the invading front. Interestingly, it turns out that the process of surface growth leading to pore closure marginally depends on the infiltration velocity. We conclude that porous matrices with straight and wide pathways represent the optimal case for impregnation. Our analysis includes also a comparison between the radii characterizing the infiltration process (i.e., minimum, hydraulic, average and effective radii).

Keywords: Capillarity; Lattice Boltzmann method; Surface growth; Liquid silicon infiltration

I. INTRODUCTION

The Lattice Boltzmann (LB) method is gaining increasing consideration for hydrodynamic simulations. A variety of systems can be studied in the incompressible limit [1–4]. The advantage of the LB method over other numerical schemes resides in its ability to deal with non-equilibrium dynamics, interface phenomena (wetting) and complex geometries (porous media). Our aim is to simulate the infiltration of molten silicon (Si) into carbon (C) preforms, taking into account the reaction of silicon carbide (SiC) formation [5–16]. The benefits of this process mainly have to do with the thermal properties.

In the simulations, the chemical reaction responsible for surface growth is based on mass precipitation [17–22]. The effects caused by the thickening of the surface are analyzed for a single capillary exhibiting the linear Washburn law [23–25]. This time dependence for the penetration of the invading front is typical of reactive Si infiltration [12, 13]. It is important to stress here that the LB models reside on assumptions that are not fulfilled in experiments. For example, in the simulations the liquid and vapor phases have almost the same density [15]. But this inconsistency is not dramatic since the density difference at the interface determines the surface tension (Laplace law) and in turn the contact angle. So, once the experimental contact angle of 30° [11, 12] is reproduced the results can be regarded as reliable in that respect. The discrepancies with experimental data are mainly due to the fact that in the simulations the Reynolds number overestimates the experimental value, particularly low (micron scale and weak velocities) [12, 14]. This means that matching with experimental results could be restored in the limit of capillaries of infinite length. However, the shortcomings arising from this inaccuracy should be limited since the reactivity is expected to be affected by the infiltration velocity not significantly [19, 20, 22]. In any case, our investigation is carried out under severe simplifying conditions. Last but not least, thermal effects are disregarded [26].

With our simulations we can ascertain that surface growth retards the process of capillary infiltration. We are also in a position to discuss the process of pore closure. The main result of our work is that the phenomenon of pore closure is to a large extent independent of the infiltration velocities. Guidelines for optimal melt infiltration are formulated accordingly.

II. LB MODELS

The LB method is based on the discretization of the velocity space [1–4], besides the discretization of the simulation domain, common to other numerical schemes. The LB method simulates the hydrodynamic behavior at a mesoscopic level since combining rules reminiscent of microscopic mechanisms and statistical treatment. The evolution of the systems is determined by iterating the BGK equation [27]. The bounce-back rule is applied for the collisions between the fluid and solid phases. It has been proven that the physics of capillary infiltration is better described by multicomponent systems [24, 25, 28, 29]. Interface phenomena (surface tension) are introduced by taking into account fluid-fluid interactions (cohesive forces). Wetting phenomena can be accounted for by incorporating solid-fluid interactions (adhesive forces). In our simulations, both types of interactions are implemented using the models proposed in Ref. [30].

In the LB framework, mass transport is described under the assumption of weak coupling with fluid flow. Also in this case, the dynamics satisfies the BGK equation [18, 22], in full analogy with fluid motion. The reaction at the surface is based on mass precipitation [18, 22] in the course of time mass deposits on the surface and when the cumulated mass exceeds a threshold value, the solid phase expands. In our simulations, the relative effects between reactivity and the characteristics of mass transport is varied only by considering different values of the reaction-rate constant $k_r$ as done in Ref. [31]. A thorough account on this aspect is proposed in Ref. [20].
In the next section, we shall present the results for capillaries of different length, height and reaction conditions. The simulations generate data in model units. The units of all quantities will be expressed in terms of the units for length $l_{u}$, for mass $m$ and for time $t_{s}$. Direct comparison with quantities given in ordinary units is possible after suitable transformation for mass $m$ and for time $t_{s}$. Direct comparison with quantities will be expressed in terms of the units for length $l_{u}$, the hydraulic radius is determined by means of the formula $r_{h} = P_{w}/A$ where $P_{w}$ is the average pressure drop across the wall.

III. RESULTS AND DISCUSSION

Our starting point is the linear Washburn law for capillary infiltration:

$$z(t) = \frac{V_{\text{cap}}H \cos \theta}{6L}t_{a}[\exp(-t/t_{a}) + t/t_{a} - 1],$$

(1)

where $V_{\text{cap}} = \gamma/\mu$ and $t_{a} = \rho H^{2}/12\mu$. Furthermore, $z$ is the centerline position of the invading front, $\gamma$ the surface tension, $\mu$ the dynamic viscosity, $\rho$ the density, $H$ the capillary height, $L$ the length, $\theta$ the contact angle. The Lattice Boltzmann method can simulate accurately this phenomenon [31]. Equation (1) predicts that the depth of the front inside the capillary varies linearly with time. A typical capillary system is illustrated in Fig. 1. The behavior of the invading front in the presence of surface reaction is shown in Fig. 2. There appears that the thickening of the surface behind the contact line retards the infiltration process, leading also to pore closure. For narrower interstices, the retardation becomes important near the onset of pore closure, so that this phenomenon is quite abrupt. For larger widths the resistance increases gradually. It is also interesting to note that, for longer capillaries, the retardation effect is less marked but the infiltration process is slower (cf. Ref. [31]).

In order to shed more light on the process of pore closure, we consider the quantity $\Delta e$ defined as the maximal width of the solid phase. Values close to $H/2$ indicate that the phenomenon of pore closure occurred. Figure 3 shows the time dependence of $\Delta e$ for different lengths of the capillaries and for different reaction-rate constants. It is important to remark that, for a given reaction-rate constant, the curves are quite similar. Indicatively, Eq. (1) suggests that the infiltration velocity is reduced by a factor of 2 in passing from $L = 500$ $[l_{u}]$ to $L = 1'000$ $[l_{u}]$. Altogether, it turns out that the kinetics of surface growth is marginally influenced by the infiltration velocity. This means that narrow interstices are particularly detrimental for reactive infiltration because they are associated with smaller velocities and they occlude sooner and abruptly.

In Fig. 4 we compare the radii characterizing the infiltration process. $r_{\text{min}}$ is the minimum radius, $r_{h}$ the hydraulic radius, $r_{\text{ave}}$ the average radius and $r_{\text{eff}}$ the effective radius. The hydraulic radius is determined by means of the formula $r_{h} = A/P$. The hydraulic radius is also interesting to note that, for longer capillaries, the retardation effect is less marked but the infiltration process is slower (cf. Ref. [31]).

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Figure 3: Time dependence of the maximal width of the solid phase $\Delta r$.

Figure 4: Comparison between the minimum, hydraulic, average and effective radii, denoted respectively by $r_{\text{min}}$, $r_h$, $r_{\text{ave}}$ and $r_{\text{eff}}$. For all plots, $L = 500$ [lu] and $k_r = 1.6 \cdot 10^{-3}$ [lu/ts]. Similar curves are obtained for the other capillary lengths. For weaker reactivities, i.e. smaller $k_r$, the various ratios are closer to 1.
the comparison is not particularly speaking. In general, it can be seen that \( r_{\text{min}} \), \( r_{\text{a}} \) and \( r_{\text{ave}} \) vary linearly with time. Deviations from the effective radius \( r_{\text{eff}} \) are smaller for the hydraulic radius, with a few exceptions if \( H = 100 \) [lu]. The hydraulic and average radii differ especially in the second half of the simulations (see Fig. 4).

IV. CONCLUSIONS

In this work, we could single out simple criteria allowing to ease the capillary infiltration of molten Si into C preforms. Namely, there appears that ideally the porous matrix should be characterized by wide pathways as straight as possible. Importantly, the process of surface growth leading to pore closure is marginally influenced by the infiltration velocity. As a consequence, narrow interstices are especially detrimental: their smaller size is in no way beneficial in order to prevent the phenomenon of pore closure. Finally, our study suggests that the porosity characteristics of the carbon preforms are critical for the process of capillary infiltration. Future work will be devoted to this issue.

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