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Critical wetting angle for spontaneous liquid infiltration into orderly packed fibres or spheres

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The static mechanics requirement for liquids to infiltrate spontaneously into orderly packed monosized spheres or fibres is studied. It is found that a wetting angle much lower than 90° is required to achieve spontaneous infiltration, and this wetting angle also varies considerably with respect to the infiltration direction, the arrangement of the fibres or spheres, and the packing density.

1. Introduction

Infiltration of liquids into porous media is a general physical phenomenon occurring in both natural and man-made environments. A study of this phenomenon has been conducted extensively in such diversified fields as soil mechanics, ground-water hydrology, petroleum engineering, filtration, and materials processing [1]. Infiltration of molten metals has also been recognized as a promising technique for producing low-cost ceramic–metal composites. However, molten metals (such as aluminium alloy) normally do not wet ceramics (such as SiC) well, and a high external pressure is usually applied to force the molten metals to infiltrate into porous ceramic preforms. So far, most investigations concerning infiltration of molten metals into porous ceramics have been devoted to the forced infiltration (see, for example, the work by Mortensen *et al.* at MIT [2]), but it has become increasingly clear that the cost of composite fabrication can be greatly reduced if the surface tension force can be utilized to achieve a spontaneous infiltration [3–7].

To obtain spontaneous infiltration, a low wetting angle between the molten metals and the ceramics is required. In the last few decades considerable effort has been made to study the wetting angle and its relationship with other factors, in order to find ways to reduce it. Despite this effort, it is still not clear how low a wetting angle is required to obtain spontaneous infiltration. Traditionally, 90° has been taken to be the critical angle for spontaneous infiltration. But experimental evidence has shown that spontaneous infiltration has not been achieved in many cases even when the wetting angle is lower than 90°. Edwards and Olsen [8] suggested this is due to the additional resistance caused by interfacial reaction kinetics and fluid flow end effect; Mortensen *et al.* blamed this on the complicated mechanical and dynamic phenomena involved in wetting during infiltration [2]. In our recent investigation, we have found that spontaneous infiltration of several porous media requires a wetting angle much lower than 90°, regardless of the dynamics or kinetics effects. This requirement is due simply to the static mechanics at the infiltration front. It is the purpose of this paper to communicate this finding and to draw attention to the static mechanics involved in spontaneous infiltration.

2. Mechanical equilibrium of a liquid surface in a capillary channel

Although it has been understood that a low wetting angle between the fluids and the porous media is a necessary condition for spontaneous infiltration, it has not been well recognized that the spontaneous movement of the infiltration front is decided by the surface tension force acting on the liquid surface when gravity force and external pressure are neglected (the gravity force is usually negligible in the case of molten metals infiltration into porous ceramics). If the liquid surface curved concavely, the surface tension force pulls the liquid forwards and spontaneous infiltration occurs; if the liquid surface curved convexly, the surface tension force pulls the liquid backwards and thus the liquid moves backwards. An equilibrium state is reached when the net surface tension force acting on the liquid surface is zero. Shown in Fig. 1 are two examples of liquid surfaces at equilibrium. In Fig. 1a the liquid climbs up until its surface becomes flat and the net surface tension force acting on the liquid

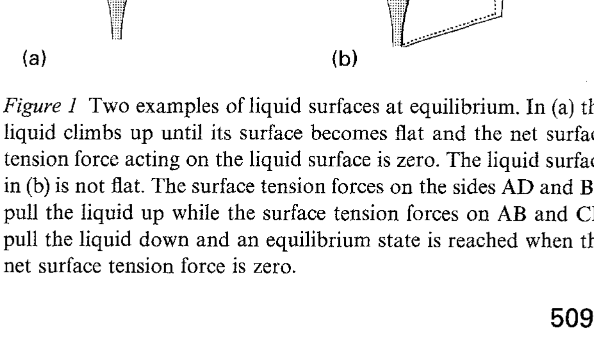

Figure 1 Two examples of liquid surfaces at equilibrium. In (a) the liquid climbs up until its surface becomes flat and the net surface tension force acting on the liquid surface is zero. The liquid surface in (b) is not flat. The surface tension forces on the sides AD and BC pull the liquid up while the surface tension forces on AB and CD pull the liquid down and an equilibrium state is reached when the net surface tension force is zero.

surface is zero. The liquid surface in Fig. 1b is not flat; the surface tension forces on the sides AD and BC pull the liquid up while the surface tension forces on AB and CD pull the liquid down and an equilibrium state is reached when the net surface tension force is zero. Therefore, the necessary condition for spontaneous infiltration is that the liquid surface at the infiltration front curves concavely. Whether or not this can be satisfied is determined by both the wetting angle and the geometry shape of the porous channels. Using this simple geometry argument, we can obtain the necessary critical wetting angles required for spontaneous infiltration in a few simplified systems.

3. Spontaneous infiltration into porous ceramic preforms

Both ceramic particulate and fibre have been used as reinforcements for metal matrix composites. The actual arrangement and the shape of ceramic particles or fibres in a preform are far too complicated to be modelled exactly in the present investigation. We therefore will choose a few simplified models in which the particles are taken to be monosized spheres, the fibres are assumed to have a circular cross-section and an identical diameter, and both the fibres and the spheres are orderly packed. When a preform is submerged into a molten alloy, infiltration should occur in all directions. However, considering that the thickness of most components is usually much smaller than their width and length, it is thus reasonable to neglect the end effect and assume that the infiltration is practically one-dimensional along the thickness direction.

3.1. Unidirectional fibre preforms

Owing to the complexity involved in the analysis of the liquid surface geometry, only unidirectionally aligned fibre preforms will be considered. The critical wetting angle for spontaneous infiltration along the fibre axis direction is 90 ° regardless of the volume fraction and the fibre arrangement. In reality, fibres usually are aligned parallel to the surfaces of a component and thus infiltration in most cases proceeds perpendicularly to the fibre axis. The critical wetting angle required for spontaneous infiltration in such cases is dependent on the infiltration direction, the fibre arrangement, and the fibre volume fraction. Shown in Fig. 2 is a preform of fibres closely packed in a square arrangement. The critical wetting angles required for infiltration in the directions A and B are 0 ° and 45 °, respectively. In direction A, the liquid infiltrates into an interstice from one corner only and a 0 ° wetting angle is required to ensure that the liquid surface completely wets the fibres in layer 1 and reaches the fibres in layer 2 before it becomes flat (equilibrium); whereas in direction B, the liquid infiltrates an interstice from two corners and thus a wetting angle of 45 ° can ensure that the liquid surface completely wets the fibres in layer 1 and reaches the fibres in layer 3 before it becomes flat. Thus, the most favourable fibre arrangement for spontaneous infiltration is that the closely packed planes incline 45 ° from

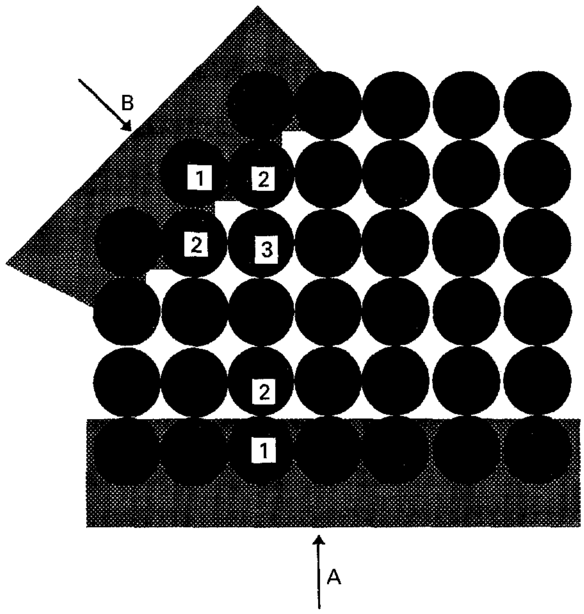

Figure 2 A fibre preform in a square arrangement. Spontaneous infiltration in the direction perpendicular to the closely packed planes requires a wetting angle of 0 °. If the wetting angle is larger than 0 °, the infiltration front becomes flat and stops before it reaches the second layer as shown in (A). However, the critical wetting angle required is only 45 ° for spontaneous infiltration in the direction inclining 45 ° from the direction (A). The infiltration front as shown in (B) is not in equilibrium state and will continue to infiltrate.

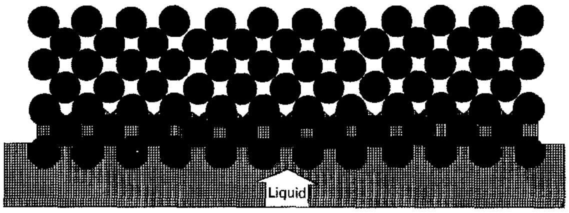

Figure 3 A preform with unidirectional fibres closely packed in a square arrangement. The closely packed fibre planes incline 45 ° from the thickness direction. Spontaneous infiltration occurs when the wetting angle is smaller than 45 °.

the thickness direction, as shown in Fig. 3. However, if the fibres are not closely packed, a still lower critical wetting angle is required (Fig. 4). Generally, for a given fibre arrangement, a lower fibre volume fraction requires a lower critical wetting angle. For a given wetting angle, θ, smaller than 45 °, the fibre volume fraction required for spontaneous infiltration must be

$$V \geq \frac{S_f}{S_{\Delta ABC}} = \frac{0.5\pi R^2}{(1+\cos\theta)^2 R^2} = \frac{\pi}{2(1+\cos\theta)^2} \qquad (1)$$

where $S_{\Delta ABC}$ and S_f are the areas of the triangle ΔABC and the area occupied by fibres within this triangle, and R is the radius of the fibres. When $\theta = 0°$, $V \geq 0.392$. Therefore, to be spontaneously infiltrated, a fibre preform must contain a fibre volume fraction higher than 39.2%. In other words, spontaneous infiltration cannot happen if the volume fraction is lower than 39.2% even when the wetting angle is 0 °.

For preforms with fibres packed closely in a triangular arrangement, the critical wetting angle for

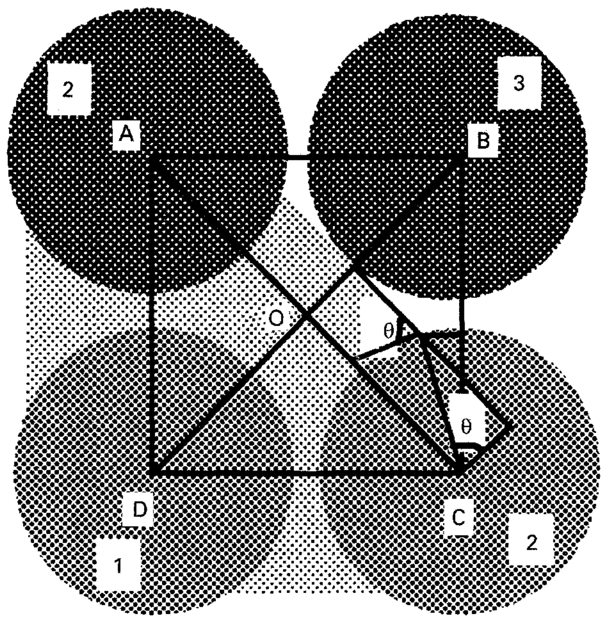

Figure 4 The critical wetting angle required for spontaneous infiltration is related to the volume fraction of the fibres. A looser packing requires a lower critical wetting angle.

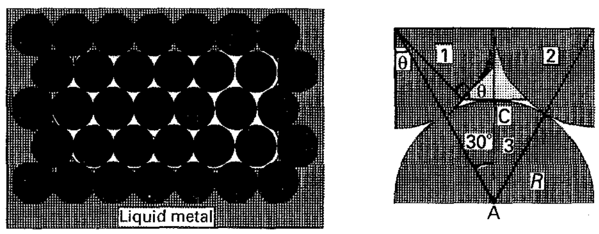

Figure 5 A preform with closely packed fibres in a triangle is submerged in a liquid. Spontaneous infiltration occurs when the wetting angle is smaller than 43 °.

spontaneous infiltration is the same for all directions. As shown in Fig. 5, where a preform is completely submerged in a liquid, to continue infiltration the liquid between fibres 1 and 2 must reach fibre 3 before its surface becomes flat (equilibrium), regardless of the infiltration direction. The critical wetting angle, θ, that satisfies this condition is shown in Fig. 5b and is 43 °. If the fibres are not closely packed, the required critical wetting angle decreases further. For a given wetting angle lower than 43 °, the minimum fibre volume fraction is

$$V \geq \frac{S_f}{S_{\Delta ABC}} = \frac{0.5\ \pi R^2}{(1+\cos\theta)^2\ R^2 \tan(30°)}$$

$$= \frac{\pi}{2\ \tan(30°)(1+\cos\theta)^2} \quad (2)$$

where the parameters are the same as those in Equation 1. Similarly, as the fibre volume fraction decreases, the critical wetting angle required for spontaneous infiltration also decreases. Taking $\theta = 0°$, we obtain $V \geq 0.68$. Thus, if the fibre volume fraction is lower than 68%, spontaneous infiltration cannot occur in a unidirectional fibre preform with triangular packing arrangement.

3.2. Orderly packed monosized spheres

Orderly packed monosized spheres can be arranged in a simple cubic, body centred cubic, or face centred cubic arrangement. The liquid surfaces at equilibrium in these preforms are usually curved in three dimensions and are difficult to analyse, except for some simple situations. In the present work, only a few simple cases in which the equilibrium liquid surfaces are flat will be treated.

For simple cubic arrangement, spontaneous infiltration in the directions of [0 0 1] requires that the wetting angle be zero (Fig. 6) and the packing be close with a packing density of $\pi/6$ (or 52.4%). Infiltration in the direction $[0\ \bar{1}\ \bar{1}]$ is much more complicated because three-dimensionally curved liquid surfaces are usually involved. Nevertheless, a simple situation shown in Fig. 6 can still be analysed, where the liquid has passed the spheres in layer 1 and is about to reach the spheres in layer 3. To achieve spontaneous infiltration the liquid surface must not become flat and the critical wetting angle required is the θ shown in Fig. 6, which is 65.5 °. This is not a sufficient, but a necessary, wetting angle for spontaneous infiltration, because, to reach the spheres in layer 3, the liquid must first pass the interstice between spheres in layers 1 and 2. The necessary wetting angle for that is not available in the present analysis. As the packing density decreases, the necessary wetting angle also decreases and their relationship can be written as

$$V \geq \frac{V_s}{V_{\text{unit}}} = \frac{4/3\ \pi R^3}{[\sqrt{2}(1+\cos\theta)]^3 R^3} = \frac{\sqrt{2}\pi}{3(1+\cos\theta)^3} \quad (3)$$

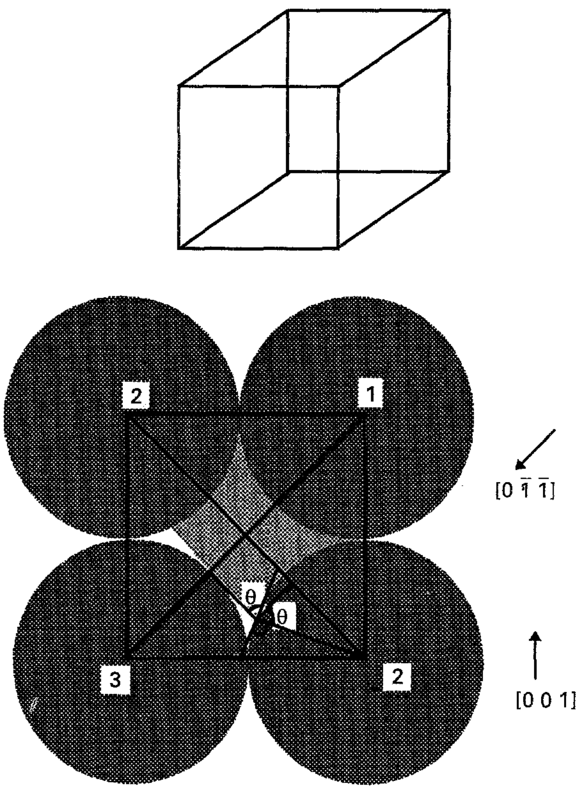

Figure 6 Spheres packed in a simple cubic arrangement. When the liquid infiltrates in the [0 0 1] direction, a wetting angle of 0 ° is required to ensure that the liquid can completely wet the spheres 2 and 3 before the liquid surface becomes flat, whereas infiltration in the $[0\bar{1}\bar{1}]$ direction requires a wetting angle, θ, as shown in the figure ($\theta = 65°$) to ensure a spontaneous infiltration.

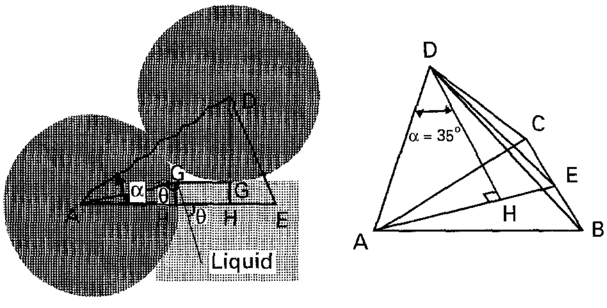

Figure 7 Infiltration of a tetrahedral interstice in f c c or h c p structures. To infiltrate spontaneously, the liquid that comes in the direction perpendicular to the closely packed planes (ΔABC) must reach the sphere D before it becomes flat (equilibrium).

where V_{unit} and V_s are the volume of the cubic unit and the volume occupied by spheres in this unit, respectively, and R is the sphere radius. When $\theta = 0°$, $V \geq 18.5\%$. Therefore, the minimum sphere packing density is 18.5% for spontaneous infiltration.

In f c c or h c p packing, spheres are closely packed in hexagonal-forming closely packed planes, and the planes are stacked in ABCABC or ABAB sequence. In the present study, we will analyse the infiltration in the direction perpendicular to the closely packed planes. This infiltration process can be simplified to be the infiltration of a tetrahedron interstice, as shown in Fig. 7. To ensure spontaneous infiltration, the liquid that is passing the spheres in the triangle ABC plane must reach the sphere at the opposite corner D before it becomes flat. The critical wetting angle satisfying this condition is 52.7°. Similarly, as the packing density decreases, the required wetting angle also decreases. For a given wetting angle lower than 52.7°, the minimum sphere volume fraction for spontaneous infiltration is

$$V = \frac{16\pi}{9\sqrt{3(1 + \cos\theta)^3}} \quad (4)$$

When $\theta = 0°$, $V = 0.4$. Thus, preforms with a sphere volume fraction lower than 40% cannot be spontaneously infiltrated even when the wetting angle is zero.

4. Conclusion

The critical wetting angles for spontaneous infiltration of liquids into several orderly packed porous media have been analysed using a simple geometry requirement. The present study involves only a few simple situations in which the liquid surfaces are flat when they reach equilibrium. Situations that involve curved equilibrium surfaces require a much more complicated mathematical analysis and the help of a three-dimensional computer graphical illustration. Results obtained from the present study offer valuable insight into the static mechanics requirement for spontaneous infiltration. It has been found that the critical wetting angle required for spontaneous infiltration is strongly associated with the packing arrangement of the fibres or monosized spheres. For given orderly arranged monosized spheres or fibres, the critical infiltration angle changes with respect to the infiltration direction, and is thus anisotropic. In all cases studied, a wetting angle much lower than 90° is required.

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