Calculation of Minimum Pressure for Liquid Metal Infiltration of a Fiber Array

SAID NOURBAKHSH, FEI-LIN LIANG, and HAROLD MARGOLIN

The effect of capillary forces on the pressure differential required for infiltration of square and hexagonal arrays of parallel fibers has been evaluated by deriving equations which include the contact angle, fiber diameter, and volume fraction. Three models have been considered as follows: (a) unidirectional liquid flow normal to the fibers, (b) bidirectional flow normal to the fibers, and (c) three-dimensional flow. The three-dimensional model predicts the lowest pressure for infiltration. A comparison is made between the required pressure differential for infiltration based on the work of immersion[1] and capillary forces. The required pressure differentials based on capillary forces for contact angles greater than 90 deg are always greater than pressure differentials calculated from the work of immersion.

I. INTRODUCTION

CONSIDERABLE interest has been developed during the past two decades in producing metal matrix composites. Among the various techniques for fabricating metal matrix composites, liquid metal infiltration offers a promise of considerable economy. Molten metals usually do not wet ceramic fibers, and, consequently, an external pressure must be applied to cause infiltration. We focus our attention on the minimum pressures required for infiltration.

Recently, Mortensen and Cornie[1] calculated the pressure differential for infiltration by using the work of immersion. The expression for the minimum pressure differential required to produce infiltration of a fiber preform was given by

$$\Delta P = \frac{4f(\gamma_{fl} - \gamma_{fa})}{(1 - f)d} \qquad [1]$$

where f is the fiber volume fraction, d is the fiber diameter, and γ_{fl} and γ_{fa} are energies of fiber/liquid and fiber/atmosphere interfaces, respectively.

In deriving this equation, the authors considered only the initial and final energy states. In the following, it will be shown that for the system to reach the final state, *i.e.*, complete infiltration, the system must overcome an energy barrier. Consequently, the work required to overcome the energy barrier determines the minimum pressure, which is higher than that calculated from the work of immersion alone (Eq. [1]).

The energy barrier arises from the need to overcome capillary forces as the liquid front moves around the fibers in the preform. A calculation considering capillary forces has already been made for a perfectly nonwetting system, *i.e.*, contact angle 180 deg.[2] In the current analysis, the effect of the contact angle, θ, on the required pressure is considered. Calculations are carried out for both square and hexagonal arrays of parallel fibers. In these calculations, viscous drag and gravitational forces are ignored, and for this reason, the calculated pressures are the minimum required pressures.

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II. THE MODELS

Three different models are considered, *i.e.*, unidirectional, bidirectional, and three-dimensional flow. In the first two flow models, the flow of the liquid metal is normal to the fibers, while in the three-dimensional case, it is parallel and normal to the fibers.

A. *The Unidirectional Flow Model*

Figure 1(a) reveals the fiber and liquid front configuration for the unidirectional flow case. As the liquid front moves through the array, the contact angle θ is maintained. Figure 1(b) illustrates the various quantities involved in the calculations. The variable R is the curvature of the liquid metal front, α is the angle between the line connecting the centers of two adjacent fibers and a radial line drawn to the point of contact between the liquid and the fiber, and d is the fiber diameter.

It can be shown that the radius of liquid metal curvature at any instant during infiltration of a square or a hexagonal array of fibers is given by

$$R = \frac{d}{2} \frac{\cos \alpha - \lambda}{\cos (\theta + \alpha)} \qquad [2]$$

where $\lambda = \sqrt{\pi/4f}$ for the square array and where $\lambda = \sqrt{\pi/2\sqrt{3}f}$ for the hexagonal array.

The minimum value of R for a given fiber volume fraction f and contact angle θ is obtained by differentiating R with respect to α. When the minimum value of R is substituted into the Gibbs-Thomson equation,

$$\Delta P = \frac{2\gamma_{la}}{R} \qquad [3]$$

the required pressure differential for infiltration is obtained. The surface tension of liquid metal in atmosphere is represented by γ_{la}.

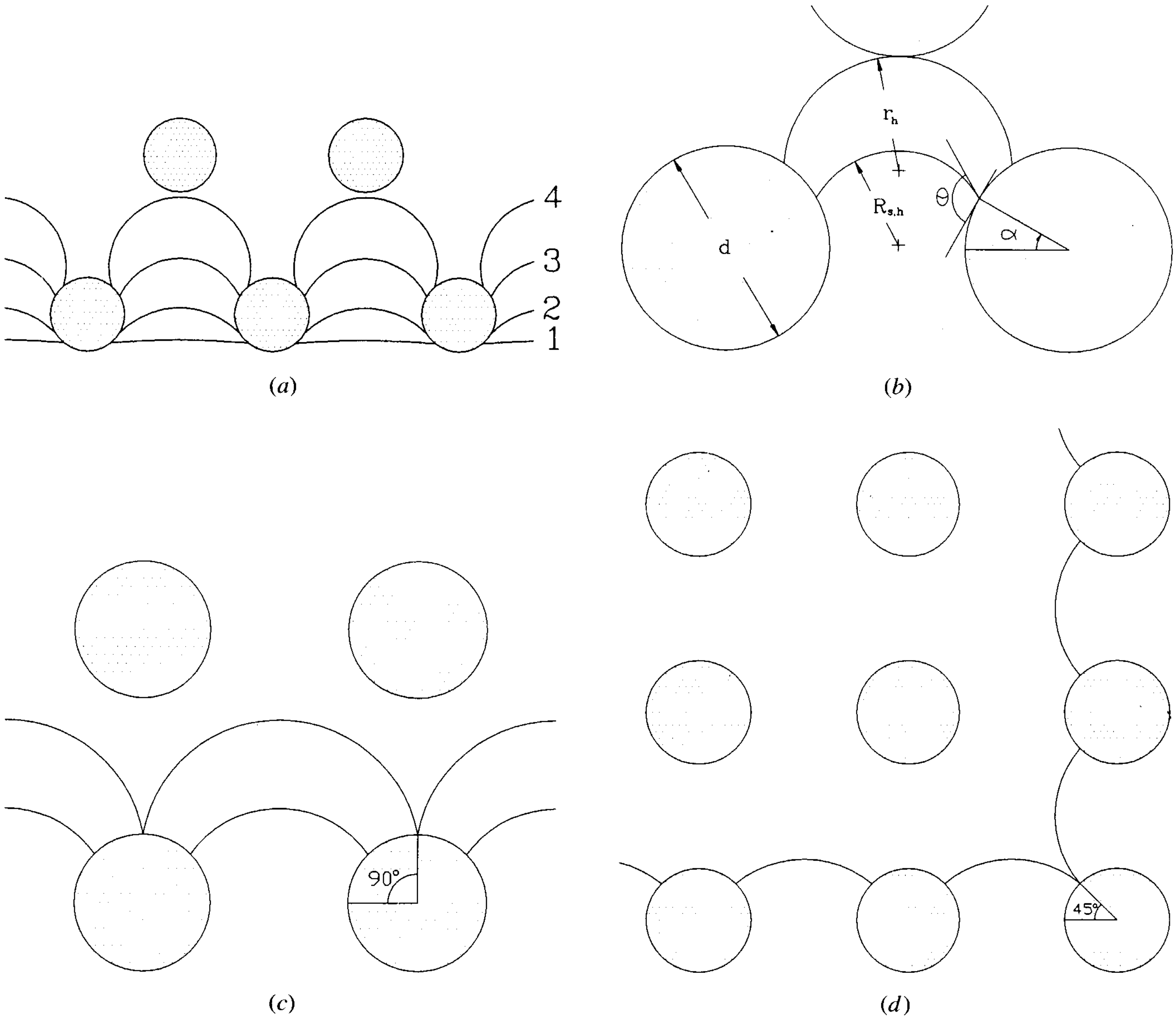

Fig. 1—(*a*) Positions of liquid front at various stages leading to infiltration. (*b*) Definition of quantities used in deriving equations of pressure differentials required for infiltration: d is the fiber diameter; θ is the contact angle; α is the angle between the line joining the centers of two adjacent fibers and a radial line drawn to the point of contact between liquid metal and fiber; $R_{s,h}$ is the radius of liquid metal curvature for square or hexagonal array; and r_h is the radius of liquid metal when the front contacts a fiber. (*c*) Illustration of liquid metal contact between two adjacent liquid fronts during unidirectional flow. (*d*) Illustration of liquid metal contact between two adjacent fronts during bidirectional flow.

When the minimum value of R is inserted in Eq. [3], the expression for ΔP becomes

$$\Delta P = \frac{4\gamma_{la}}{d} \left(\frac{\sqrt{1 - \left(\frac{\sin\theta}{\lambda}\right)^2}}{\lambda + \cos\left[\theta + \sin^{-1}\left(\frac{\sin\theta}{\lambda}\right)\right]} \right) \quad [4]$$

The values of ΔP for a square or a hexagonal array are obtained by substituting the appropriate values of λ for these two arrays. During their passage through the gaps between fibers, it is possible for two adjacent liquid fronts to meet at $\alpha = 90$ deg (Figure 1(c)) before the minimum radius of liquid curvature is reached. The pressure at this point must be taken to be the infiltration pressure. Meeting of the two liquid fronts does not occur before the minimum liquid radius is reached when $\theta \geq \tan^{-1}\lambda$; Eq. [4] is used for these values of θ.

For the case when two adjacent liquid fronts meet before the minimum radius of curvature of the liquid is reached, the infiltration pressures are given by

$$\Delta P = \frac{4\gamma_{la}}{d}\frac{\sin\theta}{\lambda} \quad [5]$$

For this situation, $\theta \leq \tan^{-1}\lambda$.

During its passage through the gaps between the fibers, the liquid metal front may meet another fiber before reaching the minimum radius of curvature or meeting an adjacent liquid front. For this case, the radius of curvature of the liquid metal at the point of contact is taken to be the minimum radius, *i.e.*, $R = r$, where r is given for the square array by

$$r_s = \frac{d}{4}\left(\frac{(1-\cos\theta) - \sqrt{\frac{2\pi}{f} + \frac{4f}{\pi}(1+\cos\theta)^2 - 5 - 6\cos\theta - \cos^2\theta}}{\frac{f}{\pi}(1+\cos\theta)^2 - 1}\right) \quad [6]$$

and for the hexagonal array by

$$r_h = \frac{d}{4}\left(\frac{\frac{2\pi}{\sqrt{3}f} - \sqrt{\frac{2\pi\sqrt{3}}{f}}}{\sqrt{\frac{\pi\sqrt{3}}{2f}} - 1 - \cos\theta}\right) \quad [7]$$

Plots of the required pressure for the unidirectional infiltration of a square and a hexagonal array of fibers *vs* contact angle for fiber volume fractions of 0.1, 0.3, 0.5, and 0.7 are given in Figures 2 and 3, respectively. In these plots, the liquid metal surface tension and the fiber diameter are assumed to be 2 J/m^2 and 20 μm, respectively. All the constraints which affect ΔP are taken into account in Figures 2 and 3.

Figures 2 and 3 both show that at constant volume fraction, the pressure required for infiltration increases as the contact angle increases. Above a volume fraction of 0.5, there is a tendency for infiltration pressure to increase drastically, as the contact angle increases above 90 deg. For all volume fractions, the square array requires a higher pressure for infiltration than the hexagonal array.

It is of interest to note that even when the contact angle is below 90 deg, the condition for wetting to occur, a positive pressure is still required for infiltration. It is further of interest to note that for the hexagonal array at low contact angles, crossovers occur in the pressure–contact angle curves in Figure 3. These crossovers occur because the liquid metal front touches another fiber before reaching its minimum radius of curvature. In the case of a volume fraction of 0.7, at a contact angle of 10 deg or less, infiltration is spontaneous, requiring no external pressure.

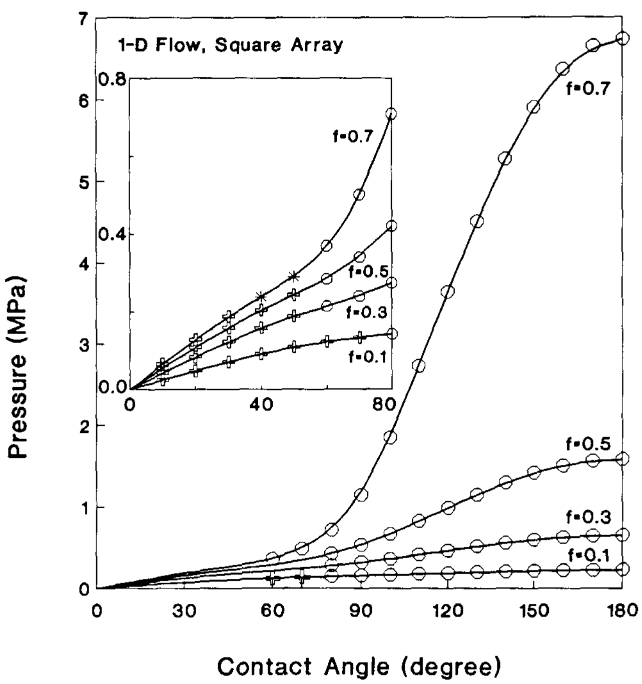

Fig. 2—Pressure differential required for liquid metal infiltration as a function of contact angle at various fiber volume fractions for unidirectional flow and square array calculated from Eqs. [4] through [6]. The cross symbols define the pressure differential governed by the contact between two adjacent liquid metal fronts. The asterisks denote the pressure differential determined by contact of the liquid metal front with a fiber. The hexagons indicate the pressure differential determined by the minimum radius of the liquid metal curvature.

B. *The Bidirectional Flow Model*

In the preceding discussion, we have considered only unidirectional flow normal to the fibers. It is possible

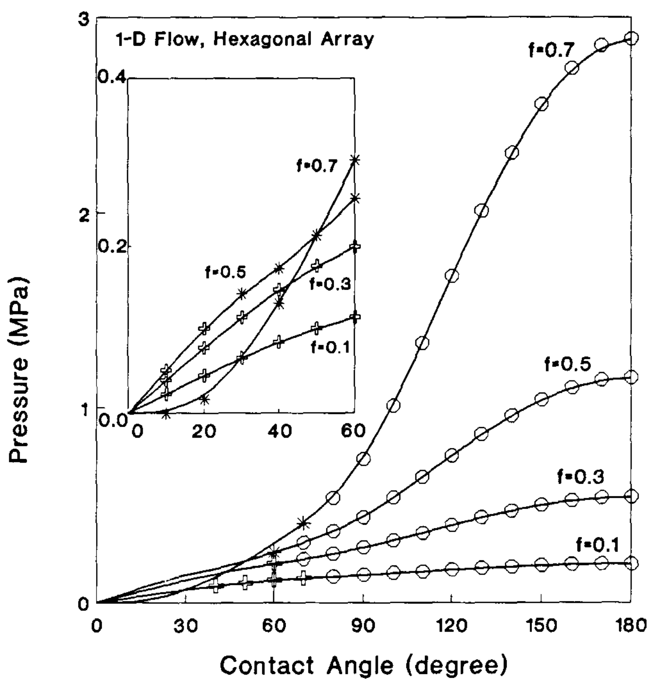

Fig. 3—Pressure differential required for infiltration of a hexagonal fiber array as a function of contact angle at various fiber volume fractions for unidirectional flow. For the definition of symbols, see Fig. 2.

for bidirectional flow normal to the fibers to occur as well, as illustrated in Figure 1(d). For bidirectional flow, Eq. [4] defines the infiltration pressure. As in the case for unidirectional flow, it is possible for two adjacent liquid metal fronts to meet before the minimum radius of curvature is reached (Figure 1(d)). This would occur at $\alpha = 45$ deg for the square array and at $\alpha = 30$ deg for the hexagonal array. For this situation,

$$\Delta P = \frac{4\gamma_{la}}{d} \frac{\cos(\theta + \alpha)}{\cos\alpha - \lambda} \qquad [8]$$

For the square array, the two liquid metal fronts will not meet for

$$\theta \geq \tan^{-1}\left(\frac{\lambda}{\sqrt{2} - \lambda}\right)$$

and for the hexagonal array, when

$$\theta \geq \tan^{-1}\left(\frac{\lambda}{2 - \sqrt{3}\,\lambda}\right)$$

The pressure differentials required for bidirectional flow, depending on the volume fraction of fibers, are equal to or less than those for unidirectional flow. Plots of bidirectional flow calculations are not shown here.

C. *The Three-Dimensional Flow Model*

In the preceding discussion, we have considered liquid metal infiltration normal to the fibers, essentially a two-dimensional process. However, infiltration can occur parallel to the fibers also, and if this type of infiltration takes place, the required external pressures for infiltration are lower than for uni- or bidirectional flow, as will be shown. The principal difference between three- and two-dimensional flow is that infiltration can take place at a larger radius of liquid metal curvature than is the case in unidirectional or two-dimensional flow. Three-dimensional flow takes place when liquid metal flowing parallel to the fibers meets liquid metal attempting to flow normal to the fibers. This is illustrated in Figures 4(a) and (b) for two possible cases.

Case A: The liquid metal flows parallel to the fibers before the liquid metal flowing perpendicular to the fibers reaches the line joining the centers of two adjacent fibers.

Case B: The liquid metal moving perpendicular to the fibers extends beyond the line joining the centers of two adjacent fibers before the liquid metal flows parallel to the fibers.

As Figure 4 indicates for Case A, the liquid metal flowing parallel to the fibers can meet the liquid moving perpendicular to the fibers only by motion perpendicular to the fibers. The two liquid fronts meet along the line joining the centers of the two adjacent fibers. The external pressure required to push the liquid metal front to the line joining the centers of the two adjacent fibers and, therefore, to complete infiltration is given by

$$\Delta P_{A,s} = \frac{8\gamma_{la}\cos\theta}{d\left(1 - \frac{\pi}{4f}\right)} \qquad [9]$$

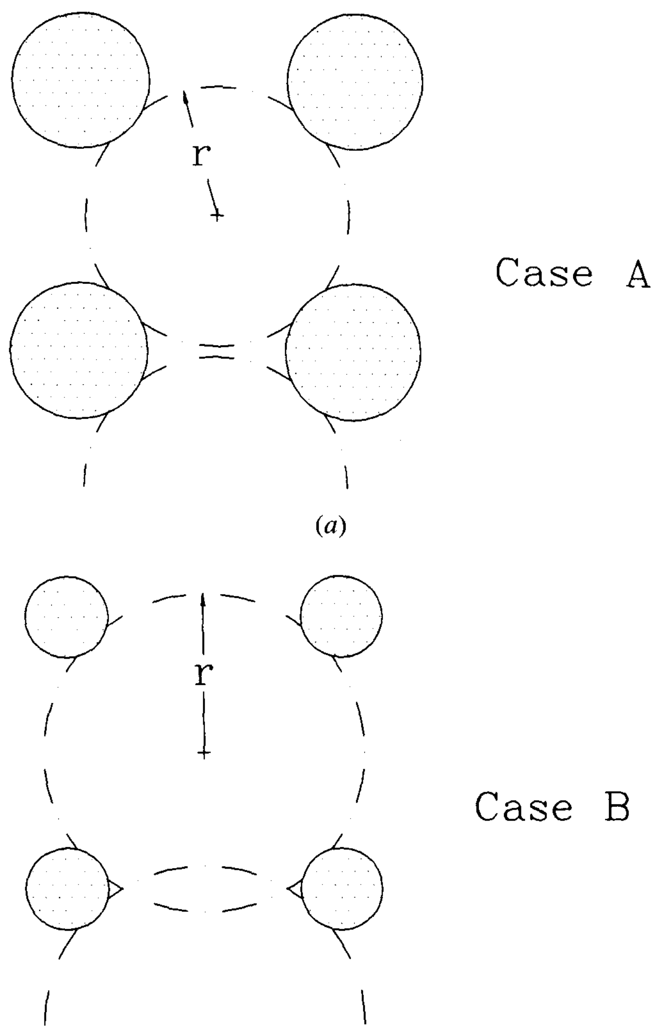

Fig. 4—Two possible liquid metal configurations during three-dimensional flow. (*a*) *Case A*: Liquid metal flow parallel to the fibers takes place before flow normal to the fibers reaches the line drawn between the centers of two adjacent fibers. (*b*) *Case B*: Liquid metal flow normal to the fibers extends beyond the line joining the centers of two adjacent fibers before liquid metal flow parallel to the fibers takes place. Sections are taken normal to the fiber axes.

for the square array and

$$\Delta P_{A,h} = \frac{8\gamma_{la}\cos\theta}{d\left(1 - \frac{\pi}{2\sqrt{3}f}\right)} \qquad [10]$$

for the hexagonal array.

As Figure 4 implies for Case B, infiltration is completed when the liquid metal flows parallel to the fibers. In this case, the required pressure to complete infiltration is given by

$$\Delta P_{B,s} = \frac{4\gamma_{la}\cos\theta}{d\left(1 - \sqrt{\frac{\pi}{2f}}\right)} \qquad [11]$$

for the square array and

$$\Delta P_{B,h} = \frac{4\gamma_{la} \cos\theta}{d\left(1 - \sqrt{\frac{2\pi}{3\sqrt{3}f}}\right)} \qquad [12]$$

for the hexagonal array.

Case A applies when ΔP_A is larger than ΔP_B. Since the dependencies of ΔP_A and ΔP_B on γ_{la}, θ, and d are the same, the relative magnitude of ΔP_A and ΔP_B is governed by the volume fraction, f. Case A for a square and hexagonal array occurs at volume fractions above 0.135 and 0.302, respectively.

The results for three-dimensional calculations of infiltration are presented in Figures 5 and 6 for the square and hexagonal arrays. The liquid metal surface tension, γ_{la}, and the fiber diameter are assumed to be 2 J/m^2 and 20 μm, respectively. Both figures show that for contact angles of 90 deg or less, infiltration is spontaneous. The required pressures for infiltration at contact angles above 90 deg increase with increasing contact angle and volume fraction in a manner similar to that seen in Figures 2 and 3 for unidirectional flow. Comparisons of Figures 5 and 6 with Figures 2 and 3 indicate that pressures required for three-dimensional flow are less than unidirectional and bidirectional (not shown) flow. For three-dimensional flow, the required pressures for infiltration are higher for square arrays than for hexagonal arrays at fiber volume fractions above 0.201.

III. DISCUSSION

In order to compare the present three-dimensional results with the calculation of Mortensen and Cornie,[1] it

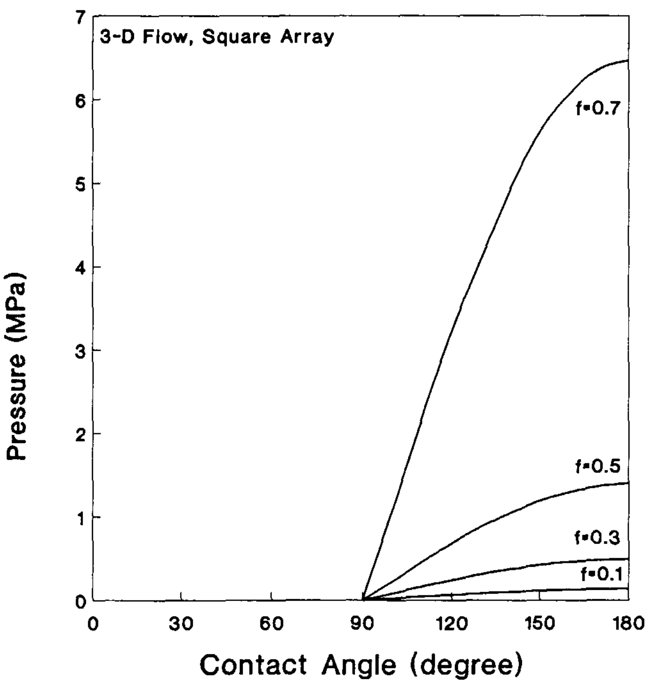

Fig. 5—Pressure differential required for infiltration as a function of contact angle at various fiber volume fractions for square array and three-dimensional flow.

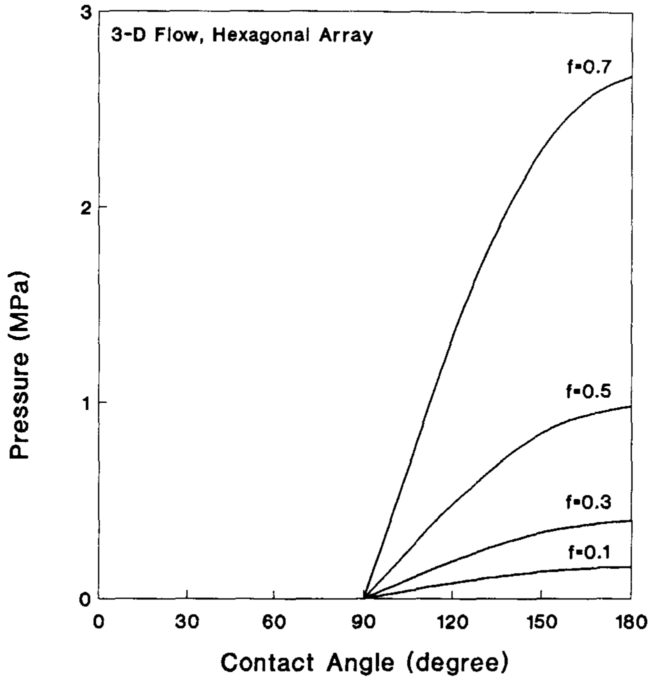

Fig. 6—Pressure differential required for infiltration as a function of contact angle at various fiber volume fractions for hexagonal array and three-dimensional flow.

is necessary to substitute $\gamma_{la} \cos\theta$ for $\gamma_{fl} - \gamma_{fa}$, in Eq. [1], according to Young's equation,

$$\gamma_{fl} - \gamma_{fa} = -\gamma_{la} \cos\theta \qquad [13]$$

The comparison is given in Figure 7. In this figure, the $\gamma_{la} \cos\theta$ term and fiber diameter are assumed to be 1 J/m^2 and 20 μm, respectively.

The hexagonal array is chosen for comparison, because it gives the smallest pressures for most volume fractions. The substitution of $\gamma_{fl} - \gamma_{fa}$ is justified as long as $\cos\theta$ in Eq. [13] lies in the range of -1 to $+1$. Since the dependencies of pressures in Eqs. [1], [10], and [12] on fiber diameter d, contact angle θ, and liquid metal surface tension γ_{la} are the same, the variable which determines the relative required pressures for complete infiltration is the volume fraction of fibers. As demonstrated in Figure 7, the required pressure for infiltration when capillary forces alone are considered is always higher than that predicted by the work of immersion for any given fiber volume fraction.

It has been argued that the required pressure for infiltration can be reduced by reducing γ_{la}.[2,3] It has also been counterargued that a reduction in γ_{la} would raise the contact angle θ in such a way that the $\gamma_{la} \cos\theta$ would remain unchanged, resulting in no change in the required pressure.[4] By using Young's equation (Eq. [13]), and Eqs. [9] through [12] which indicate that ΔP is directly proportional to $\gamma_{la} \cos\theta$, it can be shown that for systems in which $\gamma_{fl} - \gamma_{fa} > \gamma_{la}$, which is the most widely observed case, reducing γ_{la} would have no effect on infiltration pressure. In these systems, an effective way of reducing the infiltration pressure is to reduce the liquid metal/fiber interface energy and, therefore, $\cos\theta$. This can be achieved through the addition of suitable alloying elements to the melt to promote reaction with the fibers.

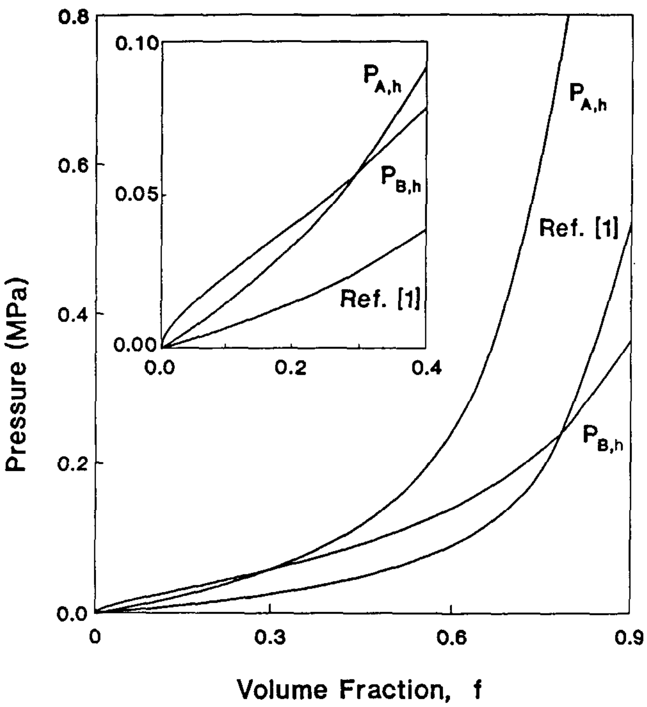

Fig. 7—Comparison of pressure differentials for infiltration determined by capillary forces and the work of immersion after Ref. 1. When capillary forces are considered for volume fractions up to 0.302, the curve labeled $P_{B,h}$ defines the required pressure differential; beyond the volume fraction of 0.302, the pressure differentials are defined by the curve $P_{A,h}$. For all volume fractions, the pressure differentials determined by capillary forces are higher than those given by the work of immersion.

For other systems in which $\gamma_{fl} - \gamma_{fa} < \gamma_{la}$ and which, therefore, would require $\cos\theta$ to be smaller than -1, a completely nonwetting system reducing γ_{la} would result in a decrease in infiltration pressure.

IV. SUMMARY

Equations for the effect of capillary forces on the pressure differentials required for the infiltration of square and hexagonal arrays of fibers have been derived. Unidirectional, bidirectional, and three-dimensional liquid flow models have been used. It is shown that the lowest pressure differentials occur for three-dimensional flow.

ACKNOWLEDGMENTS

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