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IMPREGNATION OF A HEATED FILLER WITH A VISCOUS LIQUID

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The kinetics of the penetration of a viscous liquid (connecting) inside a preliminarily heated porous body (the filler) moving inside it is considered.

When manufacturing many composite materials employed in technology the process of impregnating a certain porous body, which plays the further role of a filler of the composite material, with a viscous liquid is widely employed. The viscous liquid later changes into a solid due to crystallization or vitrification on cooling, polymerization, etc. and plays the part of a solid binding matrix. The viscosity of the liquid, even at high temperatures, is often too high, and the filler is too dense so that the hydraulic resistance experienced by the liquid when filtering through the filler is also high, and when impregnating it is necessary to apply extremely high pressure gradients which cannot easily be employed under practical conditions. Prolonged heating of the liquid to comparatively high temperatures to reduce its viscosity is undesirable in view of possible thermal expansion of the liquid or acceleration of other physicochemical processes occurring in it, and reactions which would reduce the quality of the composite material obtained. Such a situation usually arises when making many thermoplastics, glass-plastic materials, and a number of other composite materials.

One of the methods of eliminating these difficulties is by preliminary heating to high temperatures of the filler itself for relatively moderate preliminary heating of the liquid. This enables one to confine the duration of the intense heating of the liquid within permissible limits, which considerably facilitates its penetration into the filler. Hence, it is necessary to consider heat conduction in the filler—liquid system and filtering of the liquid simultaneously, taking into account the nonlinear dependence of its viscosity on the temperature.

The specific system considered is shown in Fig. 1. It represents a realistic model of certain systems used in practice for producing composite materials. In the region of negative x the filler is heated in a gaseous medium under a pressure p' (often reduced compared with atmospheric pressure) to a temperature T', which may sometimes reach thousands or more degrees Centrigrade. Hence, in region I of the filler, which may be a bunch of parallel fibers, a system of interlaced fibers, cloth material, etc., its pores are filled with gas under the pressure p'. The filler is drawn with constant velocity u into the chamber II, filled with liquid at a temperature $T'' < T'$ and a pressure $p'' > p'$. Under the action of the pressure drop which occurs, the liquid, heated by heat transfer with the filler, penetrates deeper into it, displacing gas, and at distances $x \geq L_x$ from the entry to the chamber II completely fills its pores. Thus, in addition to region I inside the filler there is a region III with pores filled with liquid; the boundary of the region is described by a certain function $Y(x)$. The length of the working part of the apparatus L_x (where $Y(L_x) = L_y$), its dependence on the various parameters of the process and the physical characteristics of the filler and liquid, and also possible methods of reducing this length while simultaneously increasing the rate of spread u, which helps to intensify the process, are of particular practical interest.

In this paper we will only investigate the plane problem (a "strip" of filler is drawn, the transverse dimensions of which in the direction perpendicular to the plane of the figure is far greater than L_y), and we will assume that the penetrability of the filler, the density and specific heat of the material of the filler and liquid and also the effective thermal conductivities are independent of the pressure and temperature.

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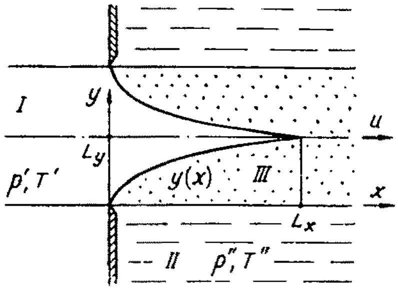

Fig. 1. Sketch illustrating the problem.

If the filler possesses a fine-pore structure, characterized by a high specific surface, we can assume that interphase heat and momentum exchange in region III occurs much more rapidly than the impregnation itself, so that when investigating the latter we can assume that the temperatures of the filler and the liquid at any physical point of this region are the same, while the component of the velocity of the liquid in the direction is simply the same as the drawing velocity u.

It follows from the equation of conservation of mass of the liquid $\partial v_y/\partial y = 0$ that the rate of filtering $v_y = v(x)$. On the basis of Darcy's law for the filtering of a liquid of variable viscosity, we have [1]

$$v(x) = -\frac{\alpha}{\mu(T)} \frac{\partial p(x, y)}{\partial y}. \tag{1}$$

The viscosity $\mu(T)$ depends on x and y implicitly in terms of the temperature dependence $T = T(x, y)$.

Integrating (1) and taking into account the fact that in regions I and II the pressure is p' and p'', respectively, we obtain

$$p_0 = p'' - p' + p_c = \frac{v(x)}{\alpha} \int_0^{Y(x)} \mu(T) \Big|_{T=T(x,\, y)} dy, \tag{2}$$

where p_c is the capillary pressure, consideration of which may be important when the pores of the filler are small [1].

From geometrical considerations we have for the function Y(x)

$$\frac{dY(x)}{dx} = \frac{v(x)}{\varepsilon u}. \tag{3}$$

The equation of convective heat conduction in region III can be written, for the same temperatures of the phases, in the form (see [2])

$$[\varepsilon\rho_0 c_0 + (1-\varepsilon)\rho_1 c_1]\, u \frac{\partial T}{\partial x} + \rho_0 c_0 v(x) \frac{\partial T}{\partial y} = \lambda_x \frac{\partial^2 T}{\partial x^2} + \lambda_y \frac{\partial^2 T}{\partial y^2}. \tag{4}$$

Generally speaking, the material is not necessarily isotropic, so that in general $\lambda_x \neq \lambda_y$. Practical methods of calculating these coefficients for heterogeneous materials of different structure can be found in [3]; the theory of the heat conduction of fiber materials, which are usually employed as fillers, is given in [4].

Equation (4) should, in the most general case, be solved simultaneously with similar equations written for regions I and II, under conditions of continuity of temperature and the normal component of the heat flux at the boundaries of the regions. Here, bearing in mind that we merely wish to obtain the simplest estimates, we will simplify the problem and we will only consider Eq. (4) for the boundary conditions

$$T = T',\ y = Y(x);\ T = T'',\ y = 0. \tag{5}$$

Hence, we have obtained the problem of heat conduction (4) and (5) in a region with unknown boundary Y(x), defined by Eq. (3), for a strong nonlinear dependence of the coefficient v(x) in (4) on the temperature, in accordance with Eq. (2).

It is not possible to solve such a complex problem directly in a complete formulation. We will therefore simplify the problem by assuming that in situations of particular practical interest we have $L_x \gg L_y$. In this case it is quite natural to use the approximation of the thermal boundary layer [5] to solve the problem. If we introduce in the usual way the dimensionless coordinates $x/L_x \sim y/L_y$, take into account the fact that $v(x)/u \sim L_y/L_x$, and estimate the terms of Eq. (4), typical for boundary layer theory [5], we arrive at the conclusion that both convective terms in (4) have the same order of magnitude, but the longitudinal heat conduction (in the x direction) can be neglected compared with the transverse heat conduction. Introducing the dimensionless parameters

$$a = \frac{\lambda_y}{\varepsilon\rho_0 c_0 + (1-\varepsilon)\rho_1 c_1}, \quad \varkappa = \frac{\rho_0 c_0}{\varepsilon\rho_0 c_0 + (1-\varepsilon)\rho_1 c_1}, \tag{6}$$

we can rewrite Eq. (4) in the form

$$u\frac{\partial T}{\partial x} + \varkappa v(x)\frac{\partial T}{\partial y} = a\frac{\partial^2 T}{\partial y^2}. \tag{7}$$

From the structure of the problem we can now suggest that it should be amenable to self-similar solutions. In the usual way, typical for the theory of thermal or hydrodynamic boundary layers, we will introduce the self-similar variable h and the parameter H

$$h = \left(\frac{u}{2a}\right)^{1/2}\frac{y}{\sqrt{x}}, \quad H = \left(\frac{u}{2a}\right)^{1/2}\frac{Y(x)}{\sqrt{x}}. \tag{8}$$

From (5) and (7), after reduction, we then obtain the following problem:

$$(\varkappa H - h)\frac{dT}{dh} = \frac{d^2T}{dh^2}, \quad T = T'(h = H), \quad T = T''(h = 0). \tag{9}$$

In deriving (9) we used the following representation for the rate of filtering of the liquid, which follows from (3) and (8) assuming H is constant:

$$v(x) = \varepsilon\left(\frac{ua}{2}\right)^{1/2}\frac{H}{\sqrt{x}}. \tag{10}$$

The solution of problem (9) for certain H can be written in the form

$$\frac{T-T''}{T'-T''} = \left[\int_0^H \exp\left(\varkappa Hh - \frac{h^2}{2}\right)dh\right]^{-1}\int_0^h \exp\left(\varkappa Hh - \frac{h^2}{2}\right)dh = \left[\operatorname{erf}\left(\frac{\varkappa H}{\sqrt{2}}\right) - \operatorname{erf}\left(\frac{(\varkappa-1)H}{\sqrt{2}}\right)\right]^{-1}\times$$
$$\times\left[\operatorname{erf}\left(\frac{\varkappa H}{\sqrt{2}}\right) - \operatorname{erf}\left(\frac{\varkappa H - h}{\sqrt{2}}\right)\right], \quad \operatorname{erf}(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt, \quad \operatorname{erf}(-x) = -\operatorname{erf}(x). \tag{11}$$

Here erf(x) is the probability integral; when writing (11) we used an expression for the integrals in (11) given in [6].

Hence v(x) and T(x, y) = T(h) are completely defined apart from the constant H. A transcendental equation for this quantity is obtained from (2) if we use as the argument of the function $\mu(T)$ the representation for T which follows from (11). Using (8) and (10) we can write this equation in the form

$$p_0 = \frac{\varepsilon a H}{\alpha}\int_0^H \mu(T)\,dh. \tag{12}$$

For an arbitrary functional dependence of $\mu(T)$, integral (12) can be obtained numerically, which enables one to find H in the form of a function of p_0 and other parameters and thereby complete the solution of the problem.

From the practical point of view an interesting situation is when $h \leqslant H \ll 1$, which, as can easily be seen from (8) assuming there that $x = L_x$, $Y(x) = L_y$, corresponds to satisfaction of the inequality

$$\left(\frac{uL_y}{2a}\right)^{1/2}\left(\frac{L_y}{L_x}\right)^{1/2} \ll 1. \tag{13}$$

In this case we obtain from (11)

$$\frac{T-T''}{T'-T''} \approx \frac{h + \varkappa Hh^2/2 - h^3/6}{H + (\varkappa - 1/3)H^3/2} \approx \frac{h}{H}. \tag{14}$$

The latter equation in (14) corresponds to the quasistatic model of heat conduction when convective heat transfer is negligibly small compared with conduction, and its use considerably simplifies the evaluation of the integral in (12).

In practice the quantity $\mu(T)$ can usually be approximated using more or less simple functions. We will consider examples of the calculations corresponding to different versions of this approximation. Suppose

$$\mu(T) \approx \mu_0 \left(1 - \frac{T - T''}{T_0}\right), \tag{15}$$

where T_0 is a certain quantity having the dimensions of temperature. This approximation is admissible when T' does not differ too much from T''. Evaluating the integral in (12) using (14) and (15) and expressing L_x in terms of L_y from the requirement that when $x = L_x$ and $y = L_y$, in accordance with (8), h is identical with H, we obtain

$$H \approx \left(\frac{\alpha p_0}{\varepsilon a \mu_0}\right)^{1/2} \left(1 - \frac{T' - T''}{2T_0}\right)^{-1/2}, \quad L_x \approx \frac{\varepsilon u \mu_0}{2\alpha p_0} \left(1 - \frac{T' - T''}{2T_0}\right) L_y^2. \tag{16}$$

Over a wider temperature range, instead of (15) one often uses the approximation

$$\mu(T) \approx \mu_0 \exp\left(-\frac{T - T''}{T_0}\right). \tag{17}$$

In this case, instead of (16) we obtain

$$H \approx \left(\frac{\alpha p_0}{\varepsilon a \mu_0} \frac{T' - T''}{T_0}\right)^{1/2} \left[1 - \exp\left(-\frac{T'' - T'}{T_0}\right)\right]^{-1/2}, \quad L_x \approx \frac{\varepsilon u \mu_0}{2\alpha p_0} \frac{T_0}{T' - T''} \left[1 - \exp\left(-\frac{T'' - T'}{T_0}\right)\right] L_y^2. \tag{18}$$

Finally, the general formula describing the dependence of the viscosity of a Newtonian liquid on the temperature has the form [7]

$$\mu(T) \approx \mu_0 \exp\frac{T_0}{T}, \tag{19}$$

where μ_0 is a slowly varying function of T, which to a first approximation can be assumed constant, while T_0 means the characteristic "activation temperature." In this case we obtain

$$\int_0^H \mu(T)\, dh = \frac{\mu_0 H T_0}{T' - T''} F\left(\frac{T'}{T_0}, \frac{T''}{T_0}\right),$$

$$F = \frac{T'}{T_0} \exp\frac{T_0}{T'} - \frac{T''}{T_0} \exp\frac{T_0}{T''} + \mathrm{Ei}\left(\frac{T_0}{T''}\right) - \mathrm{Ei}\left(\frac{T_0}{T'}\right),$$

$$\mathrm{Ei}(x) = \int_{-\infty}^{x} \frac{e^t}{t}\, dt, \tag{20}$$

where Ei(x) is the integral exponential function. Instead of (16) or (18) we have

$$H \approx \left(\frac{\alpha p_0}{\varepsilon a \mu_0}\right)^{1/2} \left(\frac{T' - T''}{F T_0}\right)^{1/2}, \quad L_x \approx \frac{\varepsilon u \mu_0}{2\alpha p_0} \frac{F T_0}{T' - T''} L_y^2. \tag{21}$$

The dependence of H and L_x on the fundamental parameters of the process corresponding to the different approximations for $\mu(T)$ have the same form and are extremely simple, so that there is no point in illustrating them graphically unrelated to the manufacture of some actual material. According to (16), (18), and (21), the length L_x, in which the impregnation is completed, is independent of the thermal characteristics of the filler, filled with liquid. It can be shown that this is due to the establishment of the quasistatic temperature distribution (14) when inequality (13) is satisfied. In the more general case the temperature field (11) and L_x depend on the thermal parameters.

In conclusion, we will point out possible further developments of this theory. Firstly, in a number of important practical cases it is also necessary to take into account the existence of a thermal boundary layer in the region II filled with liquid, i.e., to solve the conjugate problem of thermal conduction instead of the problem considered above defined only for region III. In fact, the imposition of a second boundary condition in (5) corresponds to the assumption of "rapid" heat exchange in the liquid, which occurs, e.g., due to good mixing. Similar considerations also apply to the effect of heat conduction in region I. In addition, it is sometimes important to take into account in the equations the thermal effect occurring in homogeous and heterogeneous chemical reactions, which have been ignored above.

We have only considered the plane problem corresponding to a strip of filler being drawn into a liquid. The axisymmetrical problem, when we are concerned with the impregnation of a cylindrical rope of filler is also of considerable interest. In this case a quantity occurs which has the dimensions of length, viz., the diameter of the rope, so that the process ceases to be self-similar and the mathematical problem is much more complex. In a number of cases a whole system of strips or ropes is drawn through the chamber with the liquid and collective effects of the hydrodynamic and thermal interaction between them become important; these have also not been considered.

Finally, we have only investigated the impregnation of a porous filler with a Newtonian liquid. In practice processes are widely used when the impregnating liquid possesses appreciable non-Newtonian properties, such as occurs when impregnating with resins, polymer melts, etc. The non-Newtonian properties of the liquid have an appreciable effect on its filtering characteristics, and this requires an independent analysis.

NOTATION

a, thermal diffusivity defined in (6); c_0 and c_1, specific heats of the liquid and material of the filler; F, function in (20) and (21); h and H, dimensionless quantities introduced in (8); L_x and L_y, characteristic dimensions; p, pressure; p_0, effective pressure drop taking the capillary pressure into account; T, temperature; T_0, a quantity occurring in the approximating relations for the viscosity; u, rate of pull through; v, rate of filtering of the liquid; x and y, longitudinal and transverse coordinates; Y(x), thickness of the thermal boundary layer; α, permeability of the filler; ε, porosity of the filler; $\varkappa$, a parameter defined in (6); λ_x, λ_y, thermal conductivities; $\mu(T)$, viscosity of the liquid; μ_0, characteristic value of the viscosity; ρ_0, ρ_1, densities of the liquid and material of the filler; the prime and double prime refer to the parameters in regions I and II in the figure, respectively.

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