capillary model of a WOUND thick-walled CYLINDER

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Keywords: solidification front, capillary, fiber-reinforced polymer, thick-walled cylinder, winding, viscous fluid, break of fluid, shrinkage

The mechanical phenomena in the case where the solidification front in a wound thick-walled cylinder made of a reinforced polymer moves from its outer to inner surface is investigated. The problem on motion of a viscous fluid opposite to the motion of the solidification front is solved. Experimental results obtained in modeling the motion of fluids of various viscosity are presented, and the possibilities of their break are discussed.

Ideally, in winding thick-walled products, for example, cylinders, each fiber is covered by an initially liquid binder. As a result, a capillary system is formed through which the binder, when heated and cured, is able to migrate (to percolate) under the action of the pressure drop created by tension in winding and by expansion of the binder on heating [1].

The problem of technological impermeability of wound thick-walled cylindrical products made of reinforced plastics lies in the fact that, during the manufacturing process (curing and cooling) of such products, ring cracks frequently arise in them. The basic reasons for this phenomenon at the macrolevel are the form of the bodies and the anisotropy of material properties (in curing and cooling, tensile radial stresses arise in the products); at the microlevel, the reason is that, owing to the adhesive interaction with a rigid fiber, upon curing and cooling, the binder in the interfiber space practically always is hydrostatically stretched [2]. In some studies (see, for example, [3-8]), it is mentioned that a nonuniform curing, in particular, the frontal one, can be one of the reasons for this phenomenon. In these investigations, theories (both discrete and continuous) of the mechanical phenomena of frontal curing are discussed. In the case of frontal curing, significant tensile radial stresses can appear. However, the mechanism of filtration of a liquid viscous binder opposite to the motion of the solidification front still remains undiscussed. The front, where the liquid binder is transformed into a solid, more dense state, needs some inflow (filtration) of the liquid binder from the area ahead of the front. This is the case where tensile stresses can arise in the viscous fluid, which can lead to the formation of ring breaks in it.

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Translated from Mekhanika Kompozitnykh Materialov, Vol. 50, No. 1, pp. 91-100 , January-February, 2014. Original article submitted July 10, 2013.

Fig. 1. Capillary model of a wound composite. Explanations in the text.

It is of interest to examine a model of the composite in the form of an idealized capillary system. For a cylinder, such a model can be replaced with an elementary one (Fig. 1) representing a disk-shaped slot 3 of height 2*h*, formed by end faces of two hollow cylinders 1 and filled with a viscous fluid (an oligomer with a hardener). The front of solidification reaction moves in the slot from its outer surface in the direction of decreasing radius (zone 2), i.e., toward the inner radius *a*.

Let us assume that, on the side of inner radius, i.e., from the cavity *a*, an inflow of fluid proceeds at a constant pressure p_0 . Here, we will restrict our consideration to the flow of a normal viscous incompressible fluid. However, we should bear in mind that a liquid polymer refers to the class of anomalous (non-Newtonian) fluids, which obey more complex equations [2, 9]. For an incompressible fluid, we can write the basic equations [3, 4] in a cylindrical system of coordinates

$$
-\frac{\partial p}{\partial r} + \eta \nabla^2 v_r = \rho \frac{\partial v_r}{\partial t},
$$

$$
-\frac{1}{r} \frac{\partial p}{\partial \varphi} + \eta \nabla^2 v_\varphi = \rho \frac{\partial v_\varphi}{\partial t},
$$

$$
-\frac{\partial p}{\partial z} + \eta \nabla^2 v_z = \rho \frac{\partial v_z}{\partial t},
$$
 (1)

where *p* is pressure, η is density, *v* is the flow rate (discharge) of fluid, and

$$
\nabla^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \cdot \frac{\partial^2}{\partial \varphi^2} + \frac{\partial^2}{\partial z^2}.
$$

The continuity equation has the form

$$
\operatorname{div} \upsilon = \frac{1}{r} \cdot \frac{\partial}{\partial r} \left(r \upsilon_r \right) + \frac{1}{r} \cdot \frac{\partial \upsilon_\varphi}{\partial \varphi} + \frac{\partial \upsilon_z}{\partial z} = 0. \tag{2}
$$

Due to axial symmetry, the functions p and v do not depend on φ , and, owing to narrowness of the gap, $h \ll R$ and $v_z = 0$. Then, from Eq. (2), we have

$$
v_r = \frac{f(z,t)}{r},\tag{3}
$$

and from Eqs. (1) —

$$
-\frac{\partial p}{\partial r} - \eta \frac{1}{r} \cdot \frac{\partial}{\partial r} \left[\frac{f(z,t)}{r} \right] + \eta \frac{\partial^2 f(z,t)}{r \partial z^2} = \rho \frac{\partial f(z,t)}{r \partial t}.
$$
 (4)

Let us consider the case of a stationary flow

$$
\frac{\partial v_r}{\partial t} = 0.
$$

As follows from Eq. (3), in this case, the second term in (4) is proportional to the ratio v_r/r^2 . Since we consider here the case of small flow rates (a laminar flow) and rather great values of *r*, this term can be neglected. Then, we have

$$
-\frac{\partial p}{\partial r} + \eta \frac{\partial^2 v_r}{\partial z^2} = 0.
$$
 (5)

Since $v_r = f(z)$ and $p = p(r)$, the integration of (5) yields

$$
v_r = \frac{1}{\eta} \cdot \frac{\partial p}{\partial r} \cdot \frac{z^2}{2} + c_1 z + c_2.
$$
 (6)

The boundary conditions for v_r are

$$
z = \pm h, \ \upsilon_r = 0. \tag{7}
$$

From Eqs. (6) and (7), we derive the expression

$$
\upsilon_r = \frac{h^2}{2\eta} \cdot \frac{\partial p}{\partial r} \left(\frac{z^2}{h^2} - 1 \right). \tag{8}
$$

Taking into account Eqs. (8) and (3), we can write an equation for determining the quantity $p(z)$ in the case of a stationary flow:

$$
p(r) = f(z)\varphi(z)\ln r + c_3.
$$

But *p* depends only on the value of *r*. Therefore, $f(z)\varphi(z) = c_4$, and finally

$$
p(r) = c_4 \ln r + c_3. \tag{9}
$$

The unknown constants can be found by using the following boundary conditions for *p*:

at
$$
r = a
$$
: $p = p_0$,
at $r = b$: $p = p_1$. (10)

Then,

$$
p(r) = \frac{1}{\ln(b/a)} [(p_1 - p_0) \ln r + p_0 \ln b - p_1 \ln a],
$$
\n(11)

and, from Eqs. (11) and (8) , we have

$$
v_r = \frac{h^2 \pi (p_1 - p_0)}{2\eta \ln \frac{b}{a} r} \left(\frac{z^2}{h^2} - 1\right).
$$
 (12)

Now, we will apply the resulting solution to the case of frontal curing.

Let the rate V_s of increase in the volume of the solid layer (zone 2 in Fig. 1) be given. For its formation, the volume of fluid equal to

$$
V_h = V_s \frac{\rho_s}{\rho_h}.\tag{13}
$$

is spent. At $\rho_s > \rho_h$, this means that the surface of the front of polymerization (solidification) requires an inflow of fluid from outside with the volumetric speed

$$
\upsilon = V_h - V_s = V_s \frac{\rho_s - \rho_h}{\rho_h} = V_s \frac{\Delta \rho}{\rho_h}.
$$
\n(14)

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In addition, a motion of fluid can also be caused by deformation of the formed solid ring layer (for example, as a result of chemical shrinkage). Then, the equation for the volumetric flow rate has the form

$$
\int_{-h}^{h} 2\pi r v_r(r, z, t) dz = V_s(r, t) \frac{\Delta \rho}{\rho_h} + \int_{-h}^{h} \frac{\partial u}{\partial t} 2\pi r_2 dz,
$$
\n(15)

where $r_2(z,t)$ is the coordinate of the solidification front, and $u(r_2)$ is the displacement of the internal boundary of the ring of the solid layer forming owing to its shrinking deformation.

Let us consider the simplest case where $u \equiv 0$ and $V_s = \text{const}$, i.e., if, outside the crack, there is a rigid ring (maybe, a polymer layer hardened on the outside) with a good adhesion to the forming solid polymer. In this case, Eq. (15) can be written in the form

$$
2\pi r \int_{-h}^{h} \nu_r(r, z, t) dz = V_s \frac{\Delta \rho}{\rho_h}.
$$
 (16)

Inserting here expression (8), we derive the equation for $p(r)$

$$
-\frac{4\pi h^3}{3\eta}r\frac{\partial p}{\partial r} = V_s\frac{\Delta p}{\rho}.\tag{17}
$$

After integration of Eq. (17), we obtain an expression of $p(r)$ for the boundary condition $r = a$, $p = p_0$:

$$
p(r) = p_0 - \frac{3V_s \Delta \rho \eta}{4\pi h^3 \rho_h} \ln \frac{r}{a} \quad (a \le r \le r_2).
$$
 (18)

Here, pressure is considered positive.

It follows from Eq. (18) that the minimum pressure occurs on the front, i.e., at $r = r₂$. Then, from the inequality $p(r) \le p_{cr}$, where p_{cr} is the critical pressure at which the fluid can disrupt or boil up [2], we can determine the value of r_{2cr} beginning with which (i.e., for $r \ge r_{2cr}$), in conditions of the given problem, the rigid phase formed in such a disk-shaped slot will be porous (loose).

If we take into account the deformation of the forming solid ring layer, the solution of the problem will not be so trivial, because, first, with increasing thickness, the ring will contract and may even force out the fluid from the slot; then, after the built-up of a certain thickness, the ring will need some inflow of fluid, and only then can some critical conditions, when $p \leq p_{cr}$, arise. However, the variations in v_r and p also depend on the relation between the shrinkages ε_h and ε_s .

Experimental Investigation of Boiling (Breaking) of Fluids in a Capillary

As a result of solution of problems on the mechanical behavior of homogeneous and composite materials upon frontal curing, it was found that, ahead of a closed front, if it moves from the outer surface to the center, the pressure in the liquid phase can fall down to the emergence of considerable multidirectional tensile stresses. In other words, at such a cure, there is a risk of appearance of pores (breaks) in the fluid.

It seems likely that, for composite materials, the conditions of pore formation have to be less stringent than for homogeneous fluids. This presumption is based on the following factors:

(1) the presence of a great amount of pore formation centers introduced into the binder upon its manufacture (for example, captured air particles);

(2) an additional drop in pressure in the liquid binder due to its interaction with filler surface (capillary effect);

(3) the filler is an obstacle to the inflow of the liquid phase into the zone of solidification front.

An experimental investigation of the possibility of break (boiling) of fluids in a capillary was carried out on a simple unit.

The schematic of the experiment is shown in Fig. 2. A horizontally located glass capillary 1, open at both ends and containing a column of fluid 2, was placed in a transparent glass flask connected with a vacuum pump through a vacuum gage. The indications of a manometer were registered, and the column of fluid was observed visually.

Fig. 2. Schematic of the test for investigating the breaks of fluids in a capillary: 1 — cylindrical capillary 0.1 mm in diameter, 2 — column of fluid, and 3 — vapor bubble.

Fig. 3. Determination of the boiling pressure of fluid in a capillary.

Fig. 4. Pressure at the beginning of boiling of fluid as a function of temperature: 1 and 2 — water, 3 — glycerin, and 4 — ED-20 epoxy oligomer; 1, 3, and 4 — in an open vessel and 2 — in a capillary.

Fluids of different viscosity and structure were investigated. They behaved differently. For comparison, tests on boiling of the fluids in an open vessel were performed.

Let us employ the Clausius–Clapeyron equation on the assumption that the volume of the steam phase is much greater than that of the liquid one at the same mass of substance and the state of the steam phase obeys the equation of an ideal gas. Then, we come to the known formula for variations in the boiling temperature with changes in pressure, namely

$$
p = p^* e^{-\frac{q}{nk} \left(\frac{1}{T} - \frac{1}{T^*}\right)},\tag{19}
$$

where *q* is the latent heat of boiling, *n* is the amount of molecules in a bubble, *k* is the Boltzmann constant, *T* is temperature, and T^* is the boiling temperature at a pressure p^* . However, it is known that, in a wetting liquid in a capillary, the pressure

$$
p = p_0 - \frac{2(\sigma - \gamma T)}{r_{\text{cr}}} \tag{20}
$$

is lower than the external pressure p_0 due to the surface tension σ (γ is the factor of temperature dependence of σ , and r_{cr} is the radius of capillary). Thus, we can find the boiling temperature T_0 by equalizing Eqs. (19) and (20). Graphically, it is illustrated in Fig. 3, where it is seen that the boiling temperature T_{cr} of a fluid in a capillary is lower than T_0 in an open volume under the same external pressure p_0 . In the case of reinforced systems, this could mean that the break of fluid upon frontal curing has to occur not on the front area, but at some distance ahead of it, in the region of the maximum drop in pressure.

However, experimental results showed that this conclusion is valid only for low-viscosity fluids in capillaries of relatively great diameter (0.05 mm). The boiling diagram of such a fluid in an open vessel (curve 1) and in a capillary (0.1 mm, curve 2) is shown in Fig. 4. In this case, the following picture was observed. First, in the middle region of the column, a bubble was formed (as in Fig. 2). The bubble grew very quickly practically at a constant external pressure. The column of fluid disrupted into two parts, which flew apart and froze up. It is seen that water in the capillary boils (disrupts) at lower pressures than in an open vessel. The fluids of higher viscosity do not boil in capillaries. At some sufficiently high temperatures, a column several centimeters in length disappears in a few seconds.

This phenomenon is explained by the dynamic properties of fluids in a capillary and by the process of their boiling [2]. For bubbles of the steam phase to originate, an appropriate volume is needed, and it must be formed quickly enough. Its size must be greater than some critical volume $(r > r_{cr})$, otherwise the bubble will collapse under the action of surface tension.

In a capillary, this process can be carried out only at the expense of moving apart the column of fluid in different directions from the bubble. But such a movement is hindered by viscosity of the fluid and its bond with surface of the capillary, i.e., there arises a considerable dynamic counteraction impeding the emergence of nuclei of the steam phase. This means that the disturbance of continuity in frontal cure (as also in the case of uniform curing up to gel formation) of composites with an epoxy binder (in the absence of low-molecular easily boiling fractions in it) may be expected in the presence of tensile stresses ("negative" pressure) in the fluid. In fact, this phenomenon is similar to boiling up of a liquid upon its stretching in a closed cylinder with the help of a movable piston.

Thus, as follows from the theoretical solution, a drop in pressure ahead of the solidification front down to negative values is possible. Everything depends on different test parameters and characteristics of the fluid. Experimental investigations of the behavior of three different fluids demonstrated their distinctions and particular features of their behavior in capillaries. The statement of the tests was not quite adequate to the conditions ahead of the solidification front in a capillary, although it does not exclude the possibility of break of the fluid in this area. Such a break can stop the motion of the solidification front.

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