Journal of Engineering Physics and Thermophysics, Vol. 76, No. 3, 2003

INFLUENCE OF THE VISCOSITY OF FILLED EPOXY RESINS ON THEIR SPREADING ON A HORIZONTAL SURFACE

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UDC 678.674.4.01

The rheological properties of ÉD-20-epoxy-resin-based compositions filled (10–50 vol.%) with quartz sand, quartz flour, or their mixture have been investigated on Reotest-2 and RV-8 rotational viscosimeters. The relative viscosity of the filled compositions increased with their free volume according to the exponential equation with an exponent of 2.6. The compositions began to show an anomaly of viscosity in flowing for a content of the filler of more than 40 vol.%; the yield stress appeared for 50 vol.%. The rate of spreading of the compositions on a horizontal surface decreased with increase in the composition viscosity according to the exponential equation with an exponent of 1/8, which is similar to that calculated theoretically. The experiments conducted allowed calculation of the formula of epoxy-based, self-leveling pouring floors.

An important problem in creating polymer composites is production of economical filled materials based on thermosetting resins. Such materials are also used for creation of monolithic coatings of seamless pouring floors distinguished for their aesthetic beauty, sanitation, and ease of fabrication. Epoxy resins have gained the widest acceptance as polymer matrices for such floors, whereas less expensive mineral materials — quartz sand and flour — have found wide use as fillers. One uses different diluents to reduce the rather high viscosity of epoxy resins. To work out criteria for development of the formula of such compositions we have investigated the rheological properties of filled epoxy resins and their capacity for spreading on a horizontal surface. The kinetics of spreading of relatively thick layers of Newtonian fluids can be described using a simplified Navier–Stokes equation in the form [1]

$$r = \left(\frac{4m^3 g}{\pi^3 \rho^2 \eta}\right)^{1/8} t^{1/8} . \quad (1)$$

For practical application of this equation to the process of spreading of filled epoxy resins one must know their rheological properties.

We used the most widespread epoxy resin — ÉD-20 — as the basic component of the compositions. To reduce its high viscosity (as high as 50 Pa·sec) we employed a low-viscosity diluent ÉTAL-140 (0.018 Pa·sec), which represented the plasticizer solution of an epoxy-dated oligometer. The content of the diluent in a composition was changed from 18 to 52%. The viscosity of the resin–diluent compositions decreased with increase in the content of the diluent in accordance with the logarithmic additivity rule. The fillers of a composition were the fraction of quartz sand with a particle size of 0.1–0.3 mm and φ_{max} = 0.65, the fraction of quartz flour with a particle size of 0.005 mm and φ_{max} = 0.54, and their mixture with a 3:1 ratio of the components. The composition of the mixture was selected in accordance with the capacity of each fraction for packing according to a specially developed program [2]. The value of φ_{max} for such a mixture was 0.73.

The viscosity of the epoxy compositions containing a diluent and fillers was measured by Reotest-2 (in the regime of a constant rate of shear) and RV-8 (in the regime of a constant shearing stress) rotational viscosimeters of the "cylinder-and-cylinder" type. We investigated the spreading of the compositions, determining a change in the radius of a freely spreading spot with time.

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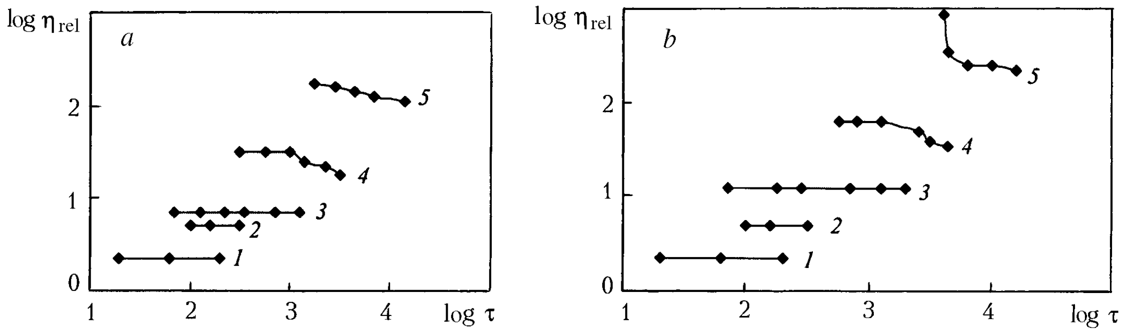
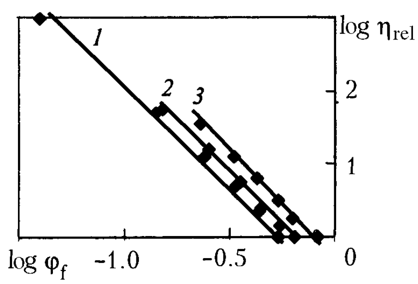


Fig. 1. Dependences of the relative viscosity of the compositions on the shearing stress in double logarithmic coordinates, which have been obtained on a Reotest-2 viscosimeter in the regime of constant rates of shear (a) and on an RV-8 viscosimeter in the regime of constant shearing stresses (b) for different concentrations of marshalite (vol.%): 1) 10, 2) 20, 3) 30, 4) 40, and 5) 50.



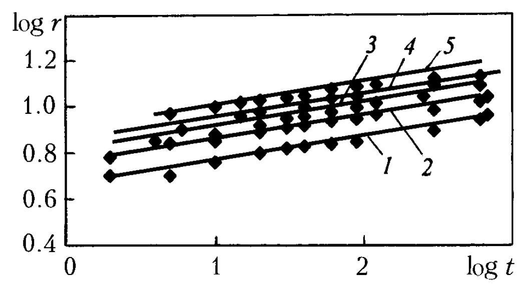


Fig. 2. Dependences of the relative viscosity of the compositions on the free volume of the disperse system in double logarithmic coordinates for different types of filler: 1) marshalite, 2) quartz sand, and 3) their mixture in a 3:1 ratio.

Fig. 3. Dependences of the radius of the spreading spot of the composition on the spreading time in double logarithmic coordinates for different concentrations of the diluent in the binder (vol.%): 1) 18, 2) 30, 3) 39, 4) 46, and 5) 52.

It was established that the composition viscosity increased with the degree of filling φ; the compositions showed Newtonian properties in flowing for a content of the fillers of 10–30 vol.%, an anomaly of viscosity appeared in them for $\varphi = 40\%$, and the yield stress appeared in the case of a 50% filling. The viscosities as functions of the shearing stress for different volume concentrations of the filler in the composition are given in Fig. 1.

The increase in the relative viscosity of the filled compositions was determined by their "free volume" in the following form [3]:

$$\eta_{rel} = K\,(\varphi_{max} - \varphi)^n, \tag{2}$$

where K is the number constant whose value depends on the type of filler and the shape of its particles; the exponent n is close to 2.5. The difference $\varphi_{max} - \varphi = \varphi_f$ is the "free volume of a filled composition" and it determines its rheological properties [3]. Indeed, in double logarithmic coordinates, the dependences of the relative viscosity of the compositions on their "free volume" were expressed as straight lines whose slope was about 2.6 (Fig. 2). For unfilled compositions we can calculate an additional point on these plots. If $\varphi = 0$, then $\varphi_f = \varphi_{max}$ and the relative viscosity of such a composition is equal to unity.

Experimental investigation of the process of spreading of unfilled compositions with different viscosities was carried out on model compositions not containing a curing agent. It showed that this process is well described by Eq. (1). Experimental curves were linearized in double logarithmic coordinates (Fig. 3). The first factor in (1), which is

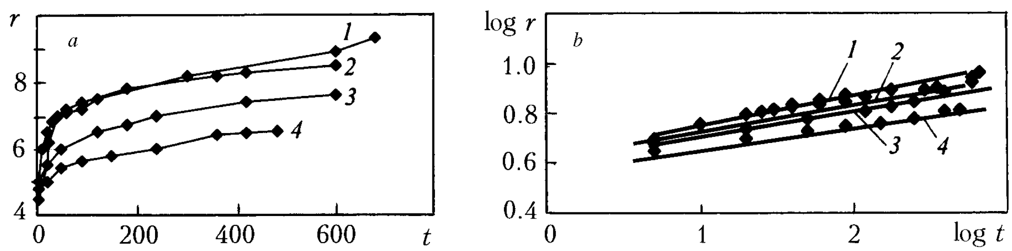


Fig. 4. Kinetics of spreading of compositions with a high-viscosity binder (18 vol.% of the diluent) which are filled with the mixture of quartz sand and marshalite in a 3:1 ratio in ordinary (a) and double logarithmic coordinates (b): 1) φ = 0, 2) 30, 3) 40, and 4) 50 vol.%.

TABLE 1. Time of Spreading (min) of Epoxy Compositions on an Area of 1 m^2

Concentration of the filler, vol.%	Concentration of the diluent, vol.%				
	52	46	39	30	18
30	0.16	0.3	0.4	1.0	2.7
40	0.6	0.7	1.2	2.0	5.2
50	3.8	4.9	6.2	9.5	17.0

TABLE 2. Residual Height of a Composition Layer (mm) after Different Periods of Spreading

Concentration of the filler, vol.%	Spreading time, min		Recommended thickness of the flow, mm
	30	10	
30	1.3	1.9	2.0
40	1.5	2.1	2.0
50	1.8	2.4	2.5
55	2.0	2.7	3.0
60	2.3	3.1	3.0
65	3.0	3.7	4.0

determined by the physical properties of the spreading composition, and the exponent of t corresponded to theoretical values. For filled resins based on binders with different viscosities the spreading curves (Fig. 4a) were also described by Eq. (1); they were linearized in double logarithmic coordinates (Fig. 4b), but the exponent of t decreased from 0.125 to 0.09.

Evaluation of the kinetics of spreading of the model compositions allowed calculation of the time parameters of spreading of actual compositions for pouring floors in the case of a significant change in the type of fillers and their concentration and in the viscosity of the resins. The calculated time of spreading of the compositions with different contents of a diluent on an area of 1 cm^2 is given in Table 1.

Also, the results presented above allow determination of the maximum height of a layer of the composition in the case of the free spreading of the latter during a given period. These values are given in Table 2 for different concentrations of the filler (the recommended thickness of the coating of the floor is presented in the last column).

The investigation carried out makes it possible to develop criteria for a substantiated selection of the formula of filled epoxy compositions for self-leveling pouring floors on the basis of solution of Eq. (1). The dependences obtained allow description of the capacity of filled compositions for spreading in a wide range of concentrations of the fillers and viscosities of the binding matrix.

NOTATION

g, free-fall acceleration; m, mass of the composition experiencing spreading; n, exponent in Eq. (2); r, radius of the spreading spot of the composition, cm; t, time of spreading of the composition, sec; η, effective (variable) viscosity of the composition, Pa·sec; η_{rel}, relative viscosity of the composition, i.e., ratio of the viscosity of a filled composition to the viscosity of its binder; φ, volume fraction of the filler in the composition, %; φ_{max}, coefficient of the maximum packing of the filler in any volume, %; φ_f, free volume of the dispersed phase in the composition, %; τ, shearing stress, Pa·sec; ρ, density of the spreading composition. Subscripts: max, maximum; f, free; rel, relative.

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