COMPARISON OF WETTABILITY AND CAPILLARY EFFECT EVALUATED BY DIFFERENT CHARACTERIZING METHODS

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ABSTRACT: The objective of this paper is to compare the capillary pressures and contact angles obtained by different characterizations including Wihelmy plate, droplet spreading, wicking, and infiltration experiments driven by air pressure or vacuum, respectively. Effects of temperature and fiber contents have been analyzed. The resultant contact angles and capillary pressures are quite different from the five measurements. Wihelmy plate method provides the quasi-static capillary pressure while in droplet spreading and wicking experiments capillary flow and spreading flow happen which depend closely on the fiber arrangement and resin viscosity. In addition, capillary pressures in axial impregnation show the same increasing tendency with the increase of fiber content. And the differences are found to be caused by the dynamic features of capillary pressure, especially for the infiltration flow driven by vacuum, which acts as a special drag force.

1 General Introduction

As a cost-effective processing technique, liquid composite molding (LCM) process has been increasingly used to manufacture composites with higher fiber volume fractions in recent years [1], in which capillary flow in the fiber tows plays an important role on adequate wetting of the fiber preform. The capillary pressure, one of the most important forces of capillary flow, can be estimated by the contact angle according to thermodynamic theories. Moreover, capillary flow should also depend on the fiber packing density, i.e. the distance between adjacent fibers, which can be directly measured by infiltration experiments with or without external applied pressures [2-5]. However, the results of different methods appear to be different even for the same fiber and liquid. Hence, it is necessary to understand the effects of factors on the contact angle (CA) and capillary pressure in details under the LCM processing conditions.

This paper aims to elucidate the wettability characterization and the resultant capillary behavior with different experimental conditions. Values of CA were measured by five ways including Wihelmy plate, droplet spreading, wicking, and infiltration experiments driven by compressed air and vacuum, respectively. Moreover, effects of temperature and fiber contents on the wettability and capillary pressure were analyzed.

2 Materials and experiments

SC8-12×20 glass fiber supplied by Nanjing Fiberglass Research and Design Institute, and carbon fiber produced by Toray were used. E-51 epoxy resin without hardener and silicone oil were used as impregnation liquids.

Wihelmy plate method [6] is widely used to measure the contact angle between single fiber and liquid. In the experiment, five fiber filaments were suspended in parallel onto a balance of tensionmeter DCAT21 Dataphysics Instrument, and the liquid was then raised to contact the filaments at a speed of 30μm/s to detect the wetting force. Advancing contact angle could be calculated by equation as follow:

$$\cos\theta = \frac{F}{\gamma P} \tag{1}$$

where F is the wetting force measured by the balance, P is filament perimeter, and γ and θ are surface tension of testing fluid and contact angle, respectively.

Droplet spreading experiment [7] was carried out with OCA21 Dataphysics Instrument. The liquid was put into a syringe, the tip of which was fixed a

few micrometers from the surface of fiber tow. The droplet volume was controlled at 1.5 μl. The apparent contact angle was recorded by video and analyzed according to Young-Laplace method.
In the wicking experiment, the liquid spontaneously impregnated aligned fiber bundles due to capillary effect. The fiber bundle was suspended onto an electric balance, which can detect the weight change of absorbed liquid. Impregnation experiments were carried out with the same specimens, and extra compressed air or vacuum pressures were applied to drive the liquid flow.

3 Results and discussion

3.1 Comparison between the quasi-static and the dynamic contact angles

Fig.1 shows typical images of the initial states of epoxy droplet on the carbon fiber tow at different temperatures, from which we can obtain the apparent CA and the wetting diameter as a function of time. Fig.2 shows that the apparent CA decreases with time until reaching the stable equilibrium state, meanwhile the wetting diameter increases with time. The higher the temperature, the faster the spreading speed revealed by the epoxy droplet. This is caused by two actions: wetting between the resin and the fiber surface, and unsaturated penetration of the resin into the space between fiber filaments simultaneously. Elevated temperature can considerably decrease the resin surface tension and the viscosity, both benefiting the wetting and wicking properties. From Fig.1 we can see that the initial apparent CA decreases with higher temperatures, which is also attributed to smaller surface tension and lower viscosity of the resin.

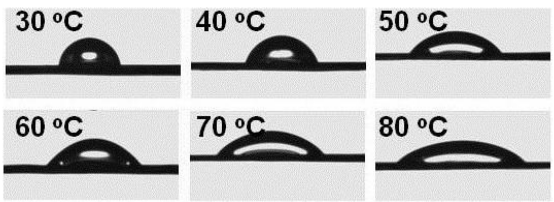
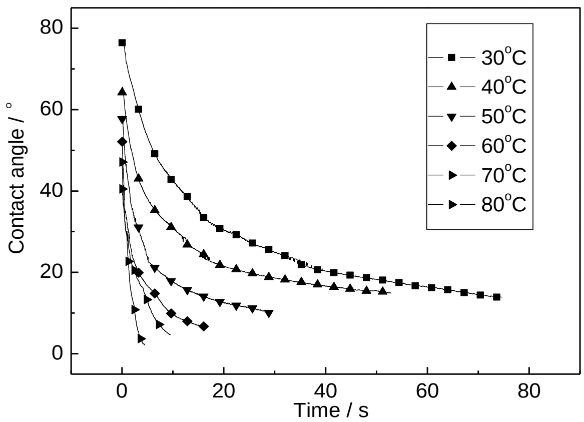
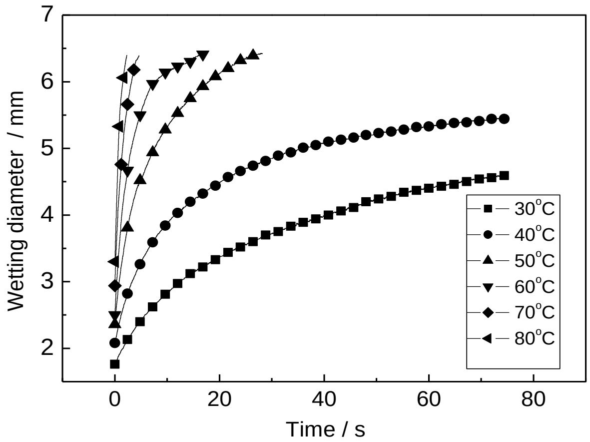


Fig.1. Initial images of epoxy droplet on the surface of carbon fiber tow at different temperatures.



(a) Contact angle



(b) Diameter of the epoxy droplet

Fig.2. Plots of contact angle and diameter of the epoxy droplet on the surface of carbon fiber tow as a function of spreading time.

Fig.3 shows the wicking height-time curves of epoxy in unidirectional carbon fiber bundles. The wicking height increases quickly at first stage, while slowly at later stage. Again, the infiltration of epoxy reveals considerable increment in velocity when the temperature increases from 40 to 80 ^{o}C. Based on Poiseuille law [8], we can estimate the wicking dynamics as follow:

$$t = -\frac{8\eta}{r_c^2 \rho g} h - \frac{8\eta P_c}{r_c^2 \rho^2 g^2} \ln\left(1 - \frac{\rho g h}{P_c}\right) \qquad (2)$$

where h is the wicking height at time t, r_c is the equivalent capillary radius of the unidirectional fiber bundles, P_c is the capillary pressure, η and ρ are the viscosity and density of liquid, respectively. Thereby, we can evaluate the value of P_c, which is the only driving force in wicking experiments, by fitting the height-time data. Fig.3 presents excellent agreement.

The contact angle can then be estimated according to Young-Laplace equation.

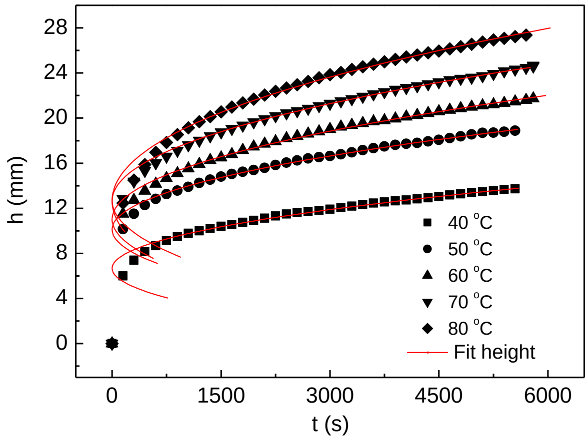
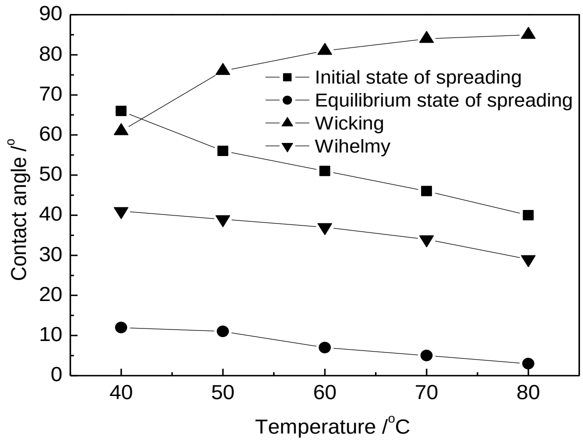
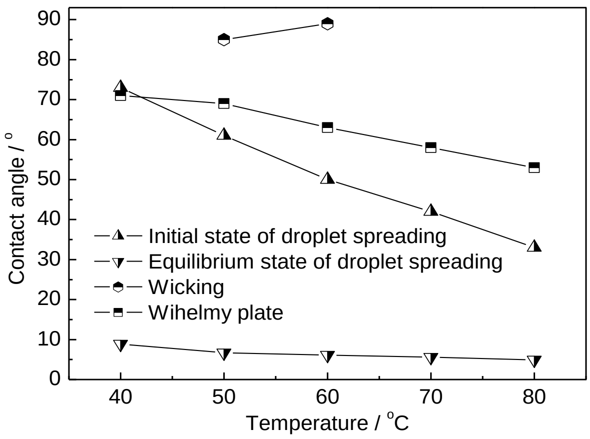


Fig.3. Wicking height-time curves for epoxy into carbon fiber bundles with 53% fiber volume fraction.

The contact angles between epoxy and carbon fiber, which are measured by Wihelmy plate, wicking, and droplet spreading respectively, are plotted versus temperature in Fig.4a. We can see that the four types of CA present quite different values ranging from 0 to 90°. The plot slope of the Wilhelmy CA is similar to that of the equilibrium state CA of spreading, both decreasing with increasing temperatures, but the contact angle values for the latter are much smaller than the former ones. Comparing with the equilibrium state, the initial state CA reveals slightly different tendency with the temperature particularly at lower temperatures (e.g. 40-50 °C), which should be ascribed to viscosity effect on the CA measurement. In comparison with the Wilhelmy method, the wicking CA increases with increasing temperature, which is contrary with the quasi-static results, indicating that the penetration dynamics takes a predominant role in LCM process. Moreover, Fig.4b shows the same variation tendency of contact angles between the glass fiber and epoxy, which are measured by different methods and at different temperatures. Furthermore, the values of Wihelmy CA for carbon fiber/epoxy are 30-40° over the experimental temperatures, while those for glass fiber/epoxy are from 53-70°, suggesting poorer surface interaction for the glass fiber than the carbon fiber with the epoxy resin.



(a) Carbon fiber/epoxy



(b) Glass fiber/epoxy

Fig.4. Plots of contact angles versus temperature, which were measured by Wihelmy plate, wicking and droplet spreading methods, respectively.

3.2 Capillary effect on impregnation dynamics

In order to further investigate the capillary effect on the impregnation dynamics, penetration experiments were conducted for aligned fiber bundles driven by external constant pressures, i.e. vacuum or compressed air. According to Darcy's law, square of the impregnation height is proportional to the time as follow [9]:

$$\psi^2 = \frac{h^2}{t} = \frac{2K}{\eta(1-V_{\rm f})}P_{\rm app} + \frac{2KP_{\rm c}}{\eta(1-V_{\rm f})} \tag{3}$$

where Ψ^2 stands for relative velocity of penetration flow, P_{app} is the applied external pressure, K is permeability of the unidirectional fiber bundle, V_f is the fiber volume fraction.

Fig.5 gives the penetration height-square root of time plots for silicone oil into aligned glass fiber bundles, which produce good linear relationships as expected by equation 3. The capillary pressure can then be obtained from the intercepts and slopes of the straight lines in Fig.5. We can find that the velocity for impregnation under vacuum assistance is relatively larger than the impregnation driven by compressed air with the same conditions.

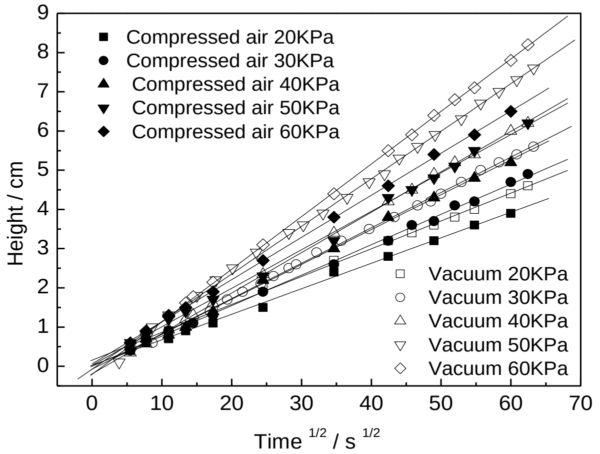
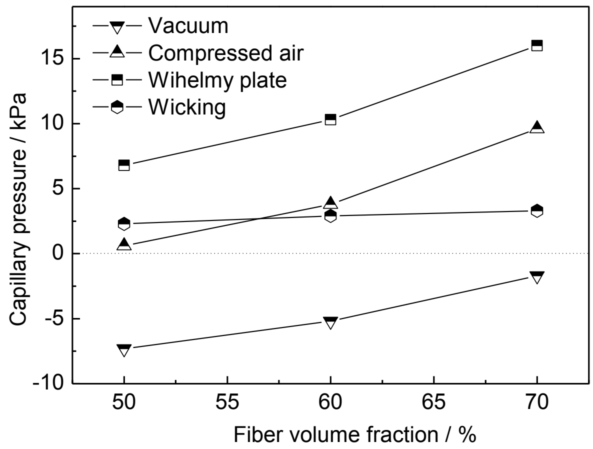


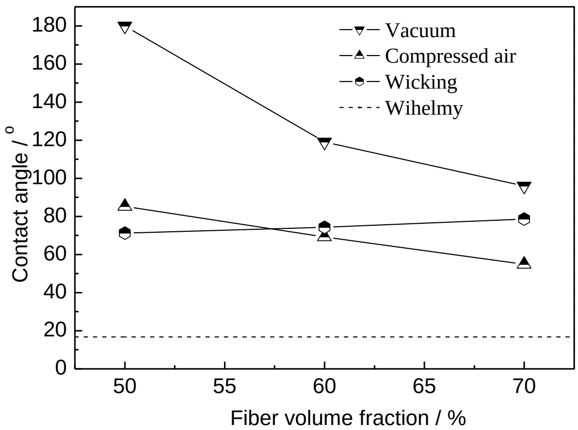
Fig.5. Penetration height of silicone oil in aligned glass fiber bundles (60% fiber volume fraction) under the assistance of external pressures, versus the square root of penetration time.

Fig.6a presents the resultant capillary pressures for silicone oil penetration into glass fiber bundles with different fiber volume fractions. Moreover, the values of P_c, which are theoretically calculated from Wihelmy CA, as well as from wicking experiments, are also plotted in the graph. One can see that the vacuum and compressed air assistant impregnation measurements result in same increasing tendency of the capillary pressure versus the fiber volume fraction, also similar to the tendency for the Wihelmy method. P_c values from the Wihelmy method are the relatively bigger than the values from compressed air assistant impregnation, however, the P_cs from vacuum assistant impregnation are much smaller. It should be noted that all the vacuum assistant impregnation P_cs are negative, suggesting a unique drag effect of capillary force, which is inconsistent with other experiments. Generally speaking, the capillary pressure always acts as promotive synergistic effect on impregnation dynamics, e.g. the compressed air driving penetration, which can be explained by quasi-static surface interactions according to Young-Laplace equation. In addition, the wicking P_c values show no significant change with the fiber volume fraction, indicating that the capillary force has considerable influence on the infiltration velocity under external pressure.

The contact angles are also calculated and shown in Fig.6b. We can find that the CA values from three type penetration experiments are all larger than the quasi-static CA from Wihelmy measurement, which is indicative of dynamic capillary effect on the penetration velocity. Moreover, the vacuum driving penetration CAs are larger than 90°, illustrating negative effect of the capillary force on impregnation. Our analysis suggests that the capillary effect is associated with the capillary number of the impregnation. These results are helpful to elucidate the wettability characterization and the capillary behavior, as well as voids formation and micro-flow simulation in LCM.



(a) Capillary pressure



(b) Contact angle

Fig.6. Capillary pressure and contact angle between glass fiber and silicone oil from Wihelmy plate, wicking and infiltration experiments driven by compressed air or vacuum, respectively.

4 Conslusions

In this paper, contact angle and capillary pressure obtained by different characterizations were compared, including Wihelmy plate, droplet spreading, wicking, and impregnation driven by compressed air pressure and vacuum, respectively. The resultant contact angles and capillary pressures are quite different from the five different measurements. The droplet spreading method is similar to Wihelmy method for characterizing the quasi-static contact angle, but they give different CA values. The wicking experiments can illustrate the effect of resin viscosity and surface tension on the capillary flow, but cannot characterize the thermodynamic wettability directly. The capillary pressures, estimated by the penetration velocity, show increasing tendency with the fiber content of aligned fiber bundles. Significant different capillary effects are observed for the silicone oil impregnation in unidirectional fiber bundles, and particularly for the vacuum driving penetration, the capillary pressure acts as a drag force on the flow velocity.

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