Polymer(Korea)·Published by the Polymer Society Korea
Vol. 8, No. 6, 1984

The Effects of Injection Pressure on the Mechanical and Viscoelastic Properties of Glass Fiber-ABS Composite

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(Received September 5, 1984; Accepted November 7, 1984)

Abstract: The effects of glass fiber content and injection pressure on the mechanical and viscoelastic properties of glass fiber-ABS composite were investigated. Fiber orientation, glass transition temperature, tensile modulus and storage modulus increased as glass fiber content increased. Tensile strength had the same propensity as tensile modulus except at high glass fiber content where its increase was negligible. It was found that fiber orientation had the maximum value at certain injection pressure and the pressure shifted toward higher pressure with the increase in glass fiber content. The same trends were observed for tensile and storage modulus. Some semi-empirical equations were proposed to predict the tensile modulus by combining theoretical equations and fiber orientation parameters.

1. Introduction

Glass fiber has been most widely used as reinforcing material to obtain the desirable properties of composite material. In addition to glass fiber, carbon fiber, graphite, aramid fiber and whiskers are used for the same goal. But these materials have a demerit of cost elevation. Therefore many researches have been devoted to the composites reinforced with glass fiber[1,2].

Discontinuous fiber reinforced plastic composites can easily be molded into various complicated shapes. Besides this merit, good mechanical properties could be achieved by controlling the orientation of reinforcing fibers at low cost of production[3,4].

Many authors reported that the mechanical properties of composite were strongly dependent upon fiber and matrix orientation[5~7]. In the course of injection molding, frozen-in orientations of the matrix and the reinforcing fiber are influenced by complex processing variables such as ram pressure, viscosity of the matrix, melt temperature and mold geometry etc[8~11]. It is generally known that injection pressure affects the degree of molecular orientation[11~13]. An increase in injection pressure causes an increase in shear stress, thus results in the decrease in melt viscosity of the polymer at high shear rate. But at relatively low shear rate (500sec^{-1}) in the temperature region of this study, the dependence of viscosity on shear rate is very small [13~15].

Fiber attrition also takes place in the process. It is well known that the length of glass fiber in the composite is much smaller than that of initial fiber. Fiber length and its distribution of the injected composite are mainly affected by injection pressure, screw design, melt viscosity of the polymer and glass fiber content etc[16,17].

Glass fiber reinforced ABS resin has good mechanical properties: high impact strength and good tensile properties etc. The effects of processing variables on the properties of Glass fiber-ABS composite, however, have seldom been reperted. In the present study, the effects of injection pressure and fiber content on the tensile and dynamic mechanical properties of glass fiber-ABS composite were examined.

2. Experimental

2-1. Materials

Commercially available ABS resin (Han Nam ABS-750) and "E" type glass fiber (mean diameter 13μ, length 3mm) were used as matrix and reinforcing fiber respectively.

2-2. Preparation of specimen

ABS resin chips were compounded with glass fiber by corotating twin-screw type compounding machine at various volume fractions of glass fiber:0. 072, 0. 112, 0. 196 and 0. 273. Subsequently, compounded materials were thoroughly dried in oven at 60-70°C for 12-24 hours. They were injected into dumbbell shaped mold for tensile test by injection molding machine (Toshiba IS-60 B)with molding conditions described below. Molded specimens were slowly cooled to the ambient temperature.

Injection pressure	700, 1000, 1300, 1600kg/cm²
Barrel temperature	250(*270)°C
Mold time	13 seconds
Mold temperature	50°C
Injection time	6 seconds

* When the volume fraction of glass fiber is equal to or greater than 0. 196.

2-3. Measurement of the length of glass fiber

Glass fibers, which were separated from ABS matrix by ashing in electric furnace at 600—650°C for more than 2 hours, were dispersed on a slide glass with water. Length and its distribution were determined by optical microscope.

2-4. Determination of fiber orientation

Orientation of glass fi ers were measured from the scanning electron micrograph (Jeol JSM-35, Fig. 1) of the specimen drawn from the central part in the longitudinal direction, which was etched by mixed solvent (Toluene: Ethanol=2 : 5) for three minutes and stained with gold. Orientation function was calculated by the equation(1).

$$f=(1/n)\sum_{i=1}^{n}\frac{1}{2}(3\cos^2\theta_i-1)\cdots\cdots(1)$$

where, n: number of measurement

θ_i: orientation angle of individual-fiber

f: orientation function

2-5. Tensile and dynamic mechanical test

Tensile moduli were determined from stress-strain curves by universal tensile tester(Aut-

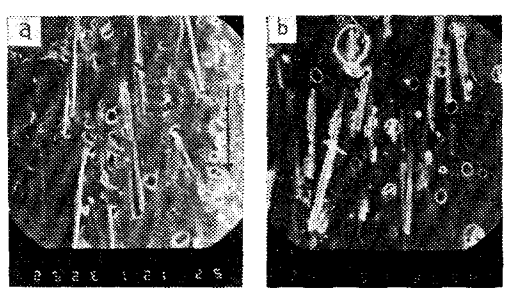

Fig. 1. Scanning electron micrograph of glass fiber-ABS composite etched by mixed solvent and stained with gold
(a) $V_2=0.273$, Inj. press. 1000kg/cm²
(b) $V_2=0.273$, Inj. press. 1600kg/cm²

ograph DCS-500, Shimadzu) at the cross head speed of 10mm/minute. Dynamic mechanical analyzer(DMA, Du Pont 981) was used for tanδ, storage and loss moduli. In the test, temperature range was 60-160°C, heating rate 10°C/minute and the amplitude of resonance vibration 0.2mm.

3. Results and Discussion

3-1. Fiber length distribution and its Orientation

There were many reports on the attrition of glass fibers in the injection molding process. Takada et al[18] reported that the mean length of glass fibers in the injected thermoplastic composites were only 0.3mm due to the attrition by screw, when its initial length was 3-6mm. According to Schweizer[17] fiber attrition was only slightly dependent upon the initial length of glass fibers in the case of polypropylene and polybutyleneterephthalate(PBT). It, however, was strongly dependent on the shear strength of the polymer matrix. Another factors that affect the fiber attrition are melt viscosity of the polymer, glass fiber content and shear rate[16], which exert negative effects on fiber length distribution. Fiber length distribution is shown in Fig. 2, when the glass fiber-ABS composite was injected at the pressure of 1600 kg/cm². Fiber length distributions for other molding conditions(not shown) were similar to that.

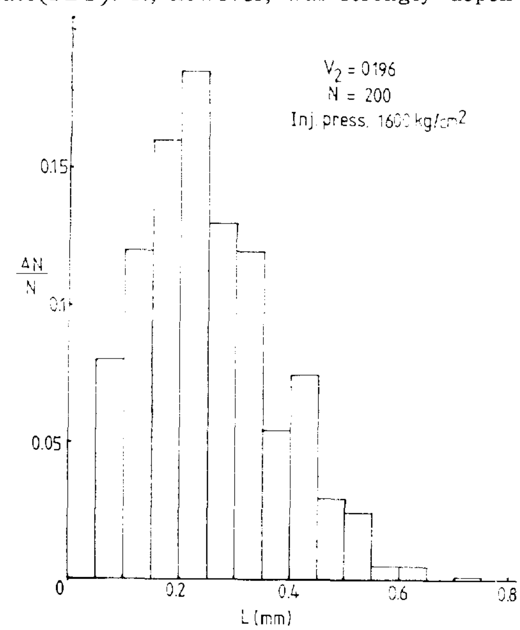

Fig. 2. Distribution of glass fiber length in the injection molded glass fiber-ABS composite.

The mean length of glass fibers at various injection pressures and glass fiber contents are listed in Table 1. It indicates that fiber attrition increases as the volume fraction of glass fiber increases. The phenomenon is due to the fiber-fiber interaction and severe shearing conditions owing to the elevation of the melt viscosity of the ABS matrix[16].

Orientation is also a very important parameter as well as aspect ratio to estimate the mechanical behaviors of fiber reinforced composites[6~9]. Orientation of glass fiber is listed in Table 2. It has increasing trend with the increase in glass fiber content, which is probably due to the increase in melt viscosity and fiber-fiber interaction. In closer look at the Table 2, one can find that at low glass fiber content (V_2=0.072) fiber orientation shows decreasing trends with the increase in injection pressure and that at the glass fiber content of 0.112 the highest orientation occurs around the injection pressure of 1300 kg/cm² Above this glass fiber content, orientations show increasing trends. From the phenomena described above, it is considered that injection pressure favors the fiber orientation till certain pressure, which depends on the processing conditions. Additional pressure, however, seems to decrease the orientation. And injection pressure with highest orientation is considered to shift toward higher position as

Table 1. Mean Length of Glass Fiber in the Glass Fiber-ABS Composite. (Glass Fibers Shorter than 0.05mm were Ruled out for Measurements and the Figures in Parenthesis are Corresponding Aspect Ratios)

unit: mm

Vol. Fraction of Glass Fiber	Injection Pressure(kg/cm²)			
	700	1,000	1,300	1,600
0.072	0.385 (29.6)	0.321 (24.7)	0.324 (24.9)	0.298 (22.9)
0.112	0.316 (24.3)	0.332 (25.5)	0.324 (24.9)	0.289 (22.3)
0.196	0.291 (22.4)	0.304 (23.4)	0.303 (23.3)	0.262 (20.2)
0.273	—	0.216 (16.6)	0.203 (15.6)	0.213 (15.6)

Table 2. Orientation Angle of Glass Fiber in the Glass Fiber-ABS Composite. (Figures in Parenthesis are Corresponding Orientation Functions)

Unit: degree

Vol. Fraction of Glass Fiber	Injection Pressure (kg/cm²)			
	700	1,000	1,300	1,600
0.072	12.6 (0.915)	16.2 (0.870)	16.8 (0.859)	13.2 (0.909)
0.112	15.7 (0.876)	12.5 (0.891)	10.3 (0.935)	11.8 (0.929)
0.196	*7.4 (0.959)	12.3 (0.894)	11.0 (0.935)	10.7 (0.923)
0.273	—	*8.4 (0.950)	10.6 (0.933)	7.5 (0.969)

*Unsatisfactorily filled specimen in the mold

glass fiber content increases.

Generally high pressure induces high shear rate, which results in high level of orientation. On the other hand, the increase in shear rate reduces the melt viscosity of the ABS matrix, which results in rapid mold filling and high level of relaxation unless there exist instantaneous cooling. Hence, the net results of these two compensating effects of shear rate give the maximum orientation at certain injection pressure[13~15]. (Unsatisfactorily filled specimen in the mold nearly without relaxation (Table 2) shows higher orientation than expected.) Addition of glass fiber to the ABS matrix brings an increase in melt viscosity and a decrease in melt elasticity[11]. In other words, substantial relaxation effect appears at higher pressure in the case of higher glass fiber content.

3-2. Mechanical properties

There are many ways to predict the elastic modulus of composite materials. One of those is "Rule of mixture"(eq.2). It is applicable

$$E_U = E_1V_1 + E_2V_2 \quad \cdots\cdots(2)$$

where, E:elastic modulus

V: volume fraction

(subscript 1, 2 hereafter denotes matrix and glass fiber respectively)

to the continuous fiber composite in which the fiber is perfectly uniaxially oriented. Nielsen proposed another relation(eq.(3)) applicable to randomly oriented three dimensional composite.

$$\log E_G = V_1 \log E_1 + V_2 \log E_2 \quad \cdots\cdots(3)$$

On the other hand, modified Halpin-Tsai equation (eq.(4)) was proposed to estimate the moduli of composites with various types of filler. Equation(4) may be applicable to the tensile and shear moduli of bead or fiber filled composites in the longitudinal and transverse directions. For uniaxially oriented fiber filled composites, A reduces to 2L/D[5a].

$$E_H = E_1 \frac{1+ABV_2}{1-B\Psi V_2} \quad \cdots\cdots(4)$$

where, A: constant

$$B = \frac{E_2/E_1 - 1}{E_2/E_1 + A}$$

$$\Psi = 1 + \left(\frac{1-\phi_m}{\phi_m^2}\right)V_2$$

ϕ_m:maximum packing fraction

For the composites with random parallel

packing of fiber, ϕ_m reduces to 0.82[5b]. Equation (4), however, is valid only when reinforcing fibers are perfectly oriented along the direction of load and adhesion between fibers and matrix is perfect. According to Patel et al.[7], orientation angle dependence of the elastic modulus of the fiber reinforced composite can be given as eq.(5). The equation, however, is applicable only when the

$$E_\theta^{-1}=\frac{1-V_2}{E_1}\sin^4\theta+\frac{1-V_2}{E_1(1-V_2)+E_2V_2}\cos^4\theta+2\frac{(1-\nu_1)(1-V_2)}{E_1}\sin^2\theta\cos^2\theta \quad \cdots\cdots(5)$$

where, E_θ: elastic modulus of composite with an orientation angle θ

ν_1: poisson's ratio for the matrix

modulus lies in between the calculated value by "Rule of mixture" and the real value of matrix. Therefore the equation may not be used for the chopped fiber composites unless some corrections, which render the modulus to lie between practical values, are made.

Tensile moduli of glass fiber-ABS composites with various processing conditions are summarized in Fig.3, 4, 5 and 6. Fig.3 shows that tensile modulus is linearly dependent on the volume fraction of glass fiber. At high volume fraction, the trend slightly deviates from the linearity. The phenomenon is supposed to stem from the reduction in aspect ratio and irregular stress transfer from the ABS matrix to glass fiber due to poor mold filling. (Actually samples of high glass fiber content (V_2=0.273) could not be molded satisfactorily at the injection pressure of 700 kg/cm^2.)

Injection pressure dependence of tensile modulus is shown in Fig.4. Tensile modulus of ABS matrix slightly decreased as injection

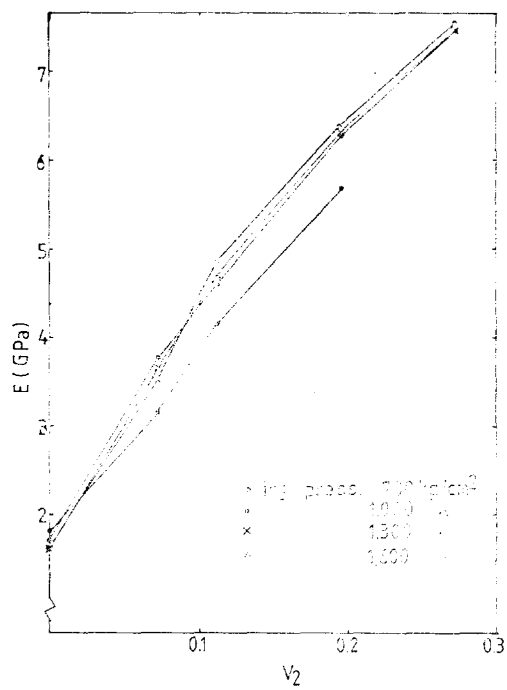

Fig. 3. Effect of glass fiber content on the tensile modulus of glass fiber-ABS composite.

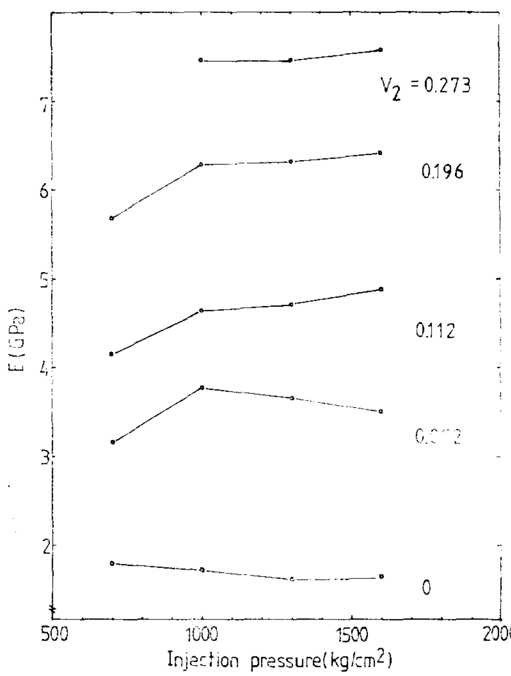

Fig. 4. Effect of injection pressure on the tensile modulus of glass fiber-ABS composite.

pressure increased from 700 to 1,600 kg/cm². At low glass fiber content (V_2=0.072), tensile modulus had a maximum at an injection pressure of 1,000 kg/cm², above which the modulus slightly decreased. At high glas s fiber contents ($V_2 \geq 0.112$), the modulus of the composite, however, increased with the injection pressure. These results are explained by considering the fact that the modulus of composite is influenced by the orientation of glass fiber and the glass fiber content.

Relationship between tensile modulus and glass fiber orientation function is shown in Fig.5. The higher the orientation function, the higher the modulus. Reversed trend, however, was observed at low glass fiber content (V_2=0.072). This is due to the fact that the relaxation of the polymer molecular chain is easier than that of glass fiber and that the matrix properties contributes considerably to the composite modulus at low glass fiber content.

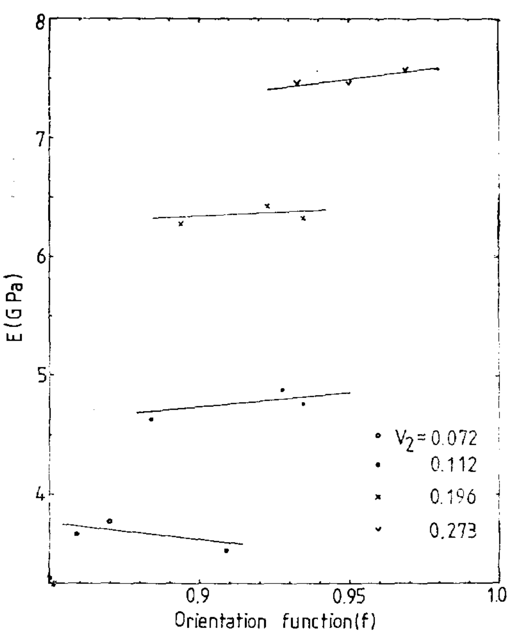

Fig. 5. Tensile modulus of glass fiber-ABS composite vs. orientation function.

Table 3. Comparison Between Experimental Moduli and Calculated Values (Injection Pressure: 1,600kg/cm²)

Vol. Fraction of Glass Fiber	Experimental Modulus (GPa)	Calculated Modulus (GPa)		
		E_U	E_G	E_H
0	1.71	—	—	—
0.072	3.52	6.55	2.22	4.42
0.112	4.88	9.23	2.58	5.97
0.196	6.33	14.9	3.52	9.22
0.273	7.58	20.0	4.68	11.7

Experimental moduli and calculated values from the eqs. (2), (3) and (4) are listed in Table 3. Calculated values differ from the experimental value to a considerable amount. The reason for the difference is that orientation of the reinforcing fiber is neglected in the calculations. Therefore it is attempted to make a semi-empirical relation(eq.(6)) for predicting the modulus of composite more precisely by combining eq.(4) and(5).

$$E=E_H \cdot F(\theta,V_2) \quad \cdots\cdots(6)$$

where, $F(\theta,V_2)=E_\theta(V_2)/E_0(V_2)$

$$E_\theta(V_2)=\frac{1-V_2}{E_1}\sin^4\theta + \frac{1-V_2}{E_H}\cos^4\theta + 2\frac{1-V_2}{E_1}(1+\nu_1)\sin^2\theta \cdot \cos^2\theta$$

$$E_0(V_2)=(1-V_2)/E_H$$

Considering that experimental modulus lies between E_H (perfect orientation) and E_G(random orientation), one can propose another semi-empirical relation (eq.(7)) that comprises E_H,E_G and f.

$$\log E=\log E_G+\text{kf}\cdot\log E_H/E_G \quad \cdots\cdots(7)$$

where, k: constant

Calculated moduli from semi-empirical eqs. (6) and (7) are listed in Table 4. Calculated values coincide with experimental values well

Table 4. Comparison Between Experimental Modulus and Calc. Value from Semi-Empirical Equation. (inj. press.;1,600 kg/cm²)

Vol. Fraction of Glass Fiber	Experimental Modulus(GPa)	Calc. Modulus (GPa)	
		Eq.(6)	Eq.(7) k=0.7
0	1.71	—	—
0.072	3.52	3.51	3.44
0.112	4.88	4.48	4.45
0.196	6.33	6.43	6.56
0.273	7.58	9.11	8.77

at low glass fiber content. At higher glass fiber content, however, the differences between both values are approximately 15—20%. They are considered to originate from some factors ruled out for theoretical calculations. Firstly, uncounted small fiber particles reduce the actual aspect ratio of reinforcing fiber and the effects of reinforcing fiber is remarkable at high glass fiber content. Secondly, aggregation of glass fiber and ineffective mold filling due to high melt viscosity at high glass fiber content yield unsatisfactory stress transfer from the ABS matrix to glass fiber.

3-3. Viscoelastic properties

Lots of work were carried out to investigate the glass transition temperature(T_g) of various composites. Kolarik et al[19]. reported that the character of molecular motion remained unchanged because the temperature position of loss maximum was independent upon the filler content in poly (2-hydroxy ethyl methacrylate)-glass fiber composites. Other authors[20,21], however, reported that the filler decreased damping and increased T_g since the adsorption of polymer segments onto the filler surfaces restricts the molecular motion of the segment and its surronding segments; modify the orientation and chain conformation of polymer segments and modify

Table 5. Glass Transition Temperature of ABS Resin Reinforced with Glass Fibers.

Vol. Fraction of Glass Fiber	Injection Pressure(kg/cm²)			
	700	1,000	1,300	1,600
0	127	129	133	129
0.072	130	129	132	132
0.112	130	133	134	133
0.196	133	133	133	132
0.273	—	134	134	134

the density of packing of polymer chains near the filler surfaces, On the contrary, Seto[22] reported that T_g of α-hematite filled vinyl chloride-vinyl acetate copolymer composite decreased as the content of α-hematite increased. He explained that the breakage of inter-chain bonding between polymer molecules due to the introduction of filler enhanced the mobility of the polymer chains and that thus decreased T_g of the composite.

In this study, T_g of glass fiber-ABS composite obtained from loss tangent increased slightly as the volume fraction of glass fiber increased (Table 5). The increase in T_g of the composite is attributable to the adsorption of ABS resin onto the surface of glass fiber. Therefore the amount of increase in T_g is influenced by the total surface area of the fibers[20]. On the other hand, T_g increased as injection pressure at low glass fiber content. The phenomenon is considered to be due to the increase in the area of interface by the attrition of glass fibers (see Table 1). But at high glass fiber content, it is nearly constant (Fig. 6).

storage modulus and loss modulus are shown in Fig. 7. Both of these moduli increased as the volume fraction of glass fiber increased. The maximum position of loss modulus was shifted to higher temperature, which is the same phenomenon as that of tanδ in relation to the glass transition temperature. Dependence of storage modulus on injection pressure ,shown in Fig.8, was similar to that of tensile modulus.

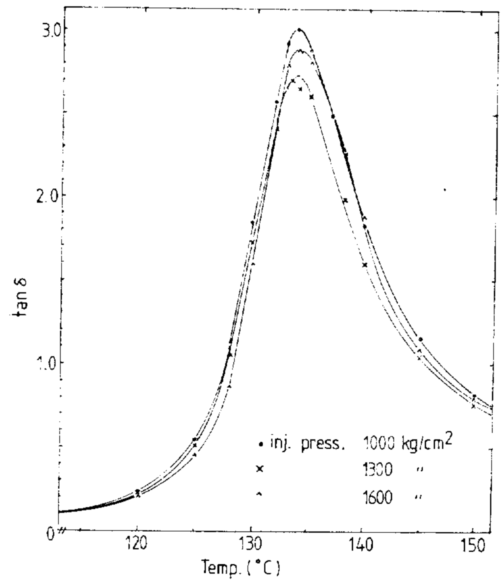

Fig. 6. Dampings of glass fiber-ABS composite (V_2=0.273)

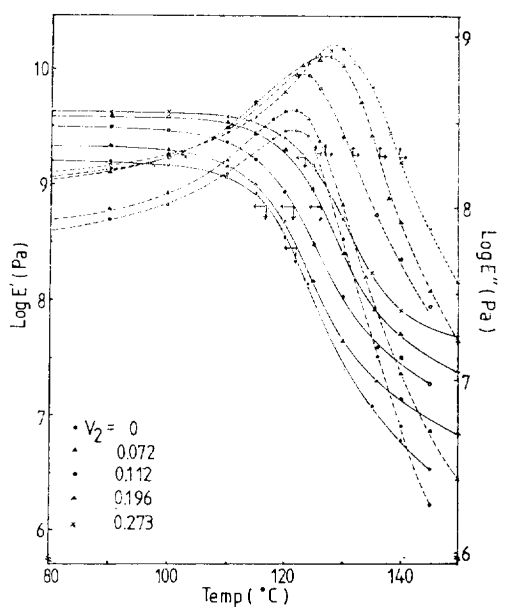

Fig. 7. Storage and loss moduli of glass fiber-ABS composite (inj.press 1600kg/cm²)

4. Conclusions

The higher the glass fiber content, the more the attrition of glass fiber. The degree of attrition due to the increase in injection pressure at fixed glass fiber content, however, was greater at low glass fiber content

Orientation of glass fiber in the ABS composite had its maximum value at certain injection pressure, which increased with glass fiber content.

Tensile moduli slightly decreased above some injection pressure at low glass fiber content, but they increased with the injection pressure at high glass fiber content.

T_g of the composite increased with glass fiber content. Increase in injection pressure, however, exerted slight effects on T_g of the composite only at relatively low glass fiber content.

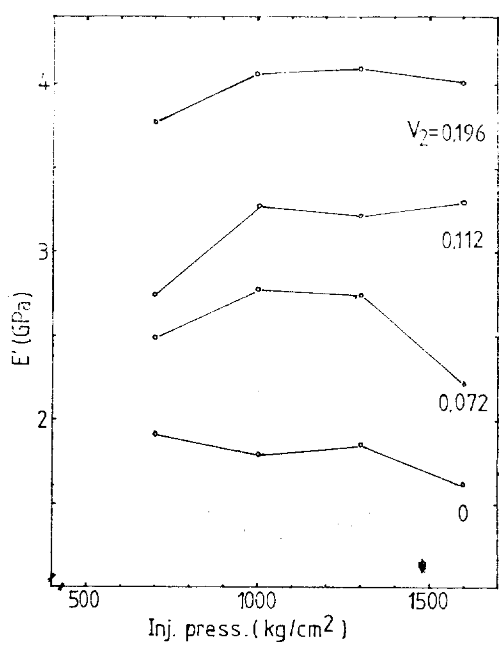

Fig. 8. Relation between dynamic storage modulus and injection pressure.

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