Effects of Temperature and Strain Rate on the Tensile Behavior of Unfilled and Talc-Filled Polypropylene. Part I: Experiments

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The tensile behavior of unfilled and 40 w% talc-filled polypropylene has been determined at four different temperatures (21.5, 50, 75 and 100°C) and three different strain rates (0.05, 0.5 and 5 min^{-1}). Experimental results showed that both unfilled and talc-filled polypropylenes were sensitive to strain rate and temperature. Stress-strain curves of both materials were nonlinear even at relatively low strains. The addition of talc to polypropylene increased the elastic modulus, but the yield strength and yield strain were reduced. The temperature and strain rate sensitivities of these materials were also different. An energy-activated, rate sensitive Eyring equation was used to predict the yield strength of both materials. It is shown that both activation volume and activation of energy increased with the addition of talc in polypropylene.

1. INTRODUCTION

During the past several years, both unfilled and mineral-filled polypropylenes have found an increasing number of applications in automotive components, such as instrument panels, radiator fans, heating ducts and housings (1). The principal reason for this increasing attention is the fact that polypropylene provides a balance of strength, modulus and chemical resistance at a relatively low cost. Both polypropylene and mineral-filled polypropylene have many potential applications in automobiles, appliances and other commercial products in which creep resistance, stiffness and some toughness are demanded in addition to weight and cost savings. Most of these applications require good performance over a range of temperatures and deformation rates. Hence it has become important to know the effects of temperature as well as strain rate on the mechanical behavior of polypropylene and its composites.

Mechanical behavior of polypropylenes has been reported by many investigators in the past. Many of these studies involved understanding the structure-property relationships or deformation characteristics of polypropylene. In recent years, Hartmann *et al.* (2) have reported the tensile yield behavior of polypropylene at different temperatures. Duffo *et al.* (3) examined the tensile behavior of polypropylene in a temperature range between 20°C and 150°C. Arruda *et al.* (4) examined the strain rate–dependent, large plastic deformation of polypropylene under uniaxial compressive load at various strain rates. Most of the publications on mineral-filled polypropylenes have addressed the effect of filler volume fraction or filler surface modification on the mechanical properties, such as modulus and impact resistance, or morphology of the material. For example, in one recent study, Stamhuis (5) showed that talc filler can significantly improve the impact resistance of polypropylene if it is physically blended with either an SBS or an EPDM elastomer. Radhakrishnan and Saujanya (6) have considered the effect of $CaSO_4$ filler on crystallization and morphology of polypropylene.

In the present paper, the tensile behavior of an unfilled polypropylene and a talc-filled polypropylene were studied at four different temperatures and three different strain rates. The purpose of this study was to determine the strain rate sensitivity and temperature sensitivity of polypropylene and talc-filled polypropylene. A further purpose of this study was to develop a simple constitutive equation that can describe the stress-strain characteristics of these materials over a range of temperatures and strain rates. The constitutive equation is presented in Part II of this research.

2. EXPERIMENTS

The materials investigated in this study were an unfilled homopolymer polypropylene and a 40 w%

talc-filled homopolymer polypropylene. Pellets of both materials were obtained from Ferro Corporation. The unfilled polypropylene pellets (Ferro NPP00GC16NA) were naturally white, while the talc-filled polypropylene (Ferro TPP40AC52BK) were black, indicating that it contained carbon black as the colorant. The melt flow rates of these two materials were reported as 4 g/10 min and 6.8 g/10 min, respectively (7). The higher melt flow rate for the talc-filled polypropylene is due to the presence of talc fillers. The glass transition temperature and melting point of polypropylene are reported as −8°C and 176°C, respectively (8).

Square plates, 150 mm × 150 mm, were injection molded from the pellets in a single edge-gated mold with a central 25-mm-diameter core (*Fig. 1*). A 90-ton injection molding machine was used to mold the plates. The melt temperature, mold temperature and injection pressure were 230°C, 65°C and 5.5 MPa, respectively, for the unfilled polypropylene and, 230°C, 40°C and 2.7 MPa for the 40 w% talc-filled polypropylene. The plate thickness was 2.5 mm. Dogbone-shaped specimens were prepared from these plates in two different directions—one parallel to the flow direction (L direction) and the other normal to the flow direction (W direction). The specimen dimensions were 100 mm in overall length, 50 mm in gauge length and 12.7 mm in gauge width.

Uniaxial tensile tests were performed on an MTS servohydraulic test machine equipped with a forced air convection oven. A strain gauge extensometer (25-mm gauge length) was used to measure axial strain. The tests were conducted at 1.25, 12.5 and 125 mm/min. Since the extensometer gauge length was 25 mm, it is assumed that the strain rates were 0.05, 0.5 and 5 min^{-1}. The test temperatures were 21.5, 50, 75 and 100°C.

Three parameters were evaluated from each stress-strain curve: elastic modulus (E), yield strength (σ_y) and yield strain (ε_y). Elastic modulus or Young's modulus is the initial slope of the stress-strain curve. Yield strength is assumed to be the maximum stress observed in each stress-strain diagram and the strain corresponding to the yield strength is the yield strain. Necking was observed in unfilled polypropylene specimens at temperatures other than 21.5°C. For talc-filled-filled polypropylene, slight necking was observed at 75 and 100°C, but not at 21.5 and 50°C.

3. RESULTS

Tensile stress-strain curves of unfilled and talc-filled polypropylene in the flow (L) direction under different test conditions are shown in *Figs. 2* and *3*, respectively. Tensile stress-strain curves normal to the flow (W) direction are similar in nature. From *Figs. 2* and *3*, it can be observed that the stress-strain relationships of both unfilled and talc-filled polypropylene were nonlinear even at strains lower than the yield strain. Each curve shows a maximum stress, which is assumed to be the yield strength of the material. After yielding, stress decreased steadily with strain until fracture occurred. The tensile properties of both materials at different test conditions are listed in *Tables 1* and 2.

Figures 2 and *3* show the effect of temperature on the stress-strain curves of unfilled polypropylene and talc-filled polypropylene in the L directions. At the same strain rate, the overall stress level decreased with increasing temperature. Both elastic modulus and yield strength decreased with increasing temperature. While the yield strain of talc-filled polypropylene increased with increasing temperature in the temperature range considered, the yield strain of unfilled polypropylene increased as the temperature was increased up to 75°C; however, at 100°C, the yield strain of unfilled polypropylene decreased. This behavior is similar to the results reported by Hartmann *et al.* (2). *Figures 2* and *3* also show the effect of strain rate on the stress-strain curves of unfilled polypropylene and talc-filled polypropylene parallel to the flow (L) direction. It is observed that the elastic modulus and yield strength increased with increasing strain rate for both materials; however, the effect of strain rate on the yield strain was relatively small for the three strain rates investigated.

Figures 4a and *4b* show comparisons of stress-strain curves of unfilled polypropylene and talc-filled polypropylene at a strain rate of 5 min^{-1} and temperatures of 21.5°C and 100°C. As expected, talc-filled polypropylene had a higher elastic modulus than unfilled polypropylene. However, as can be observed in these figures as well as from *Tables 1* and *2*, the difference in yield strengths of unfilled polypropylene and talc-filled polypropylene decreased with increasing temperature. The difference between the yield strains of unfilled polypropylene and talc-filled polypropylene also decreased with increasing temperature. This may be due to relaxation of thermal residual stresses in the polypropylene matrix surrounding the talc fillers as the test temperature was increased. The residual stresses were developed in the injection molding process as the polypropylene matrix cooled down from the melt temperature. This was due to the large difference in the coefficients of thermal expansion of the polypropylene matrix and the talc filler.

Figures 5a and *5b* show comparisons of stress-strain curves in the L and W directions. For unfilled polypropylene (*Fig. 5a*), the yield strength and stress level after yielding were slightly higher in the W direction than in the L direction. For talc-filled polypropylene (*Fig. 5b*), the yield strength and stress levels after yielding were much higher in the L direction than in the W direction. The yield strength in the W direction was lower by almost 15%. The significant difference in properties in the two mutually perpendicular directions indicates inherent anisotropy of the talc-filled polypropylene. From *Fig. 5b*, it can also be observed that with increasing temperature, the difference between the stress-strain curves of talc-filled polypropylene in the L direction and the W direction became smaller. This may also be an effect of relaxation of residual stresses with increasing temperature.

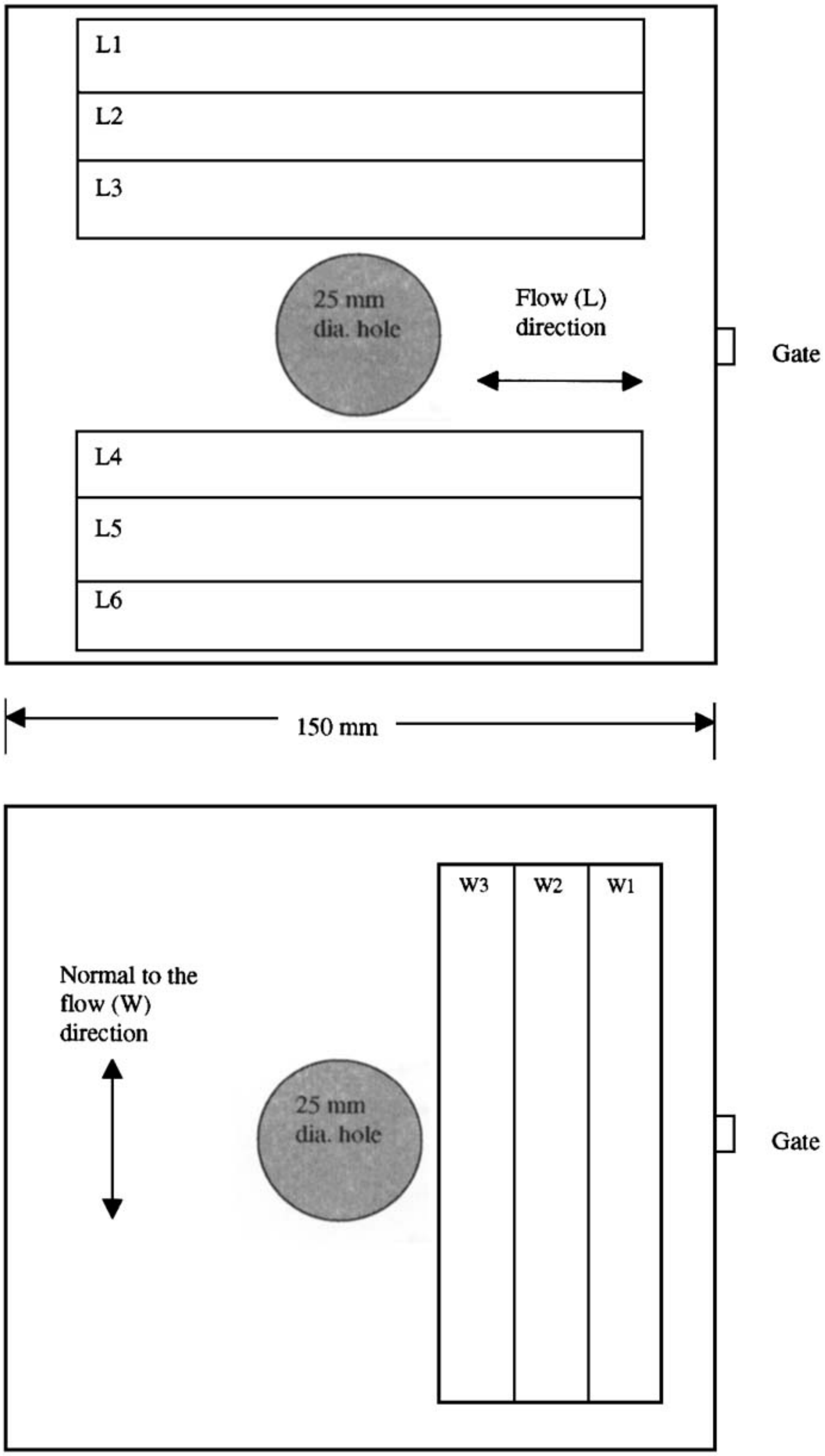


Fig. 1. Tensile specimen locations in injection molded plates.

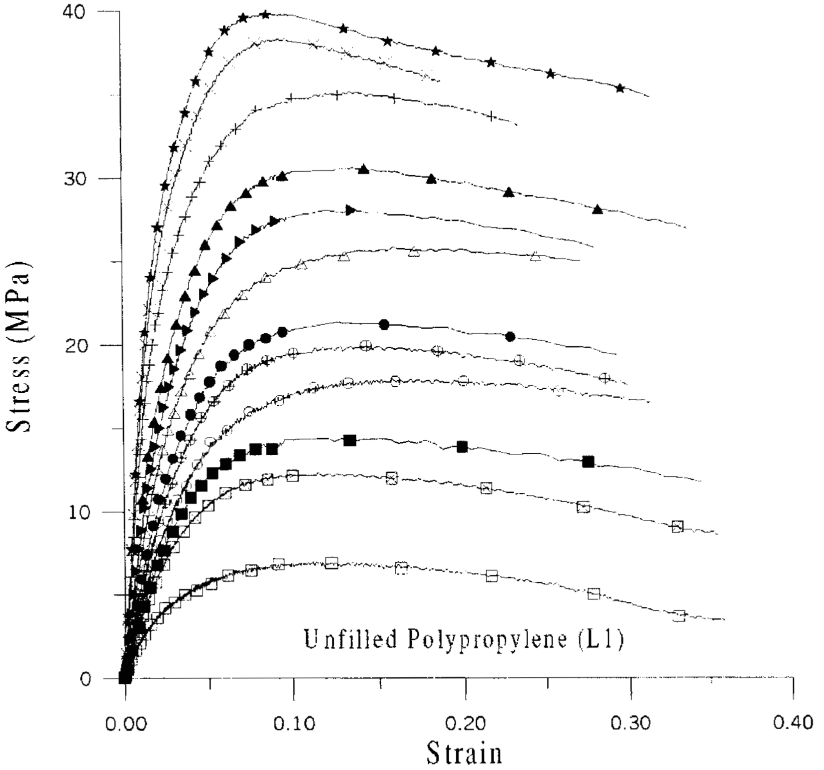


Fig. 2. Stress-strain curves of unfilled polypropylene in the flow (L) direction at various strain rates and temperatures. + 21.5°C, 0.05 min^{-1}; × 21.5°C, 0.5 min^{-1}; ★ 21.5°C, 5 min^{-1}; △ 50°C, 0.05 min^{-1}; ▶ 50°C, 0.5 min^{-1}; ▲ 50°C, 5 min^{-1}; ○ 75°C, 0.05 min^{-1}; ⊕ 75°C, 0.5 min^{-1}; ● 75°C, 5 min^{-1}; □ 100°C, 0.05 min^{-1}; ⊠ 100°C, 0.5 min^{-1}, ■ 100°C, 5 min^{-1}.

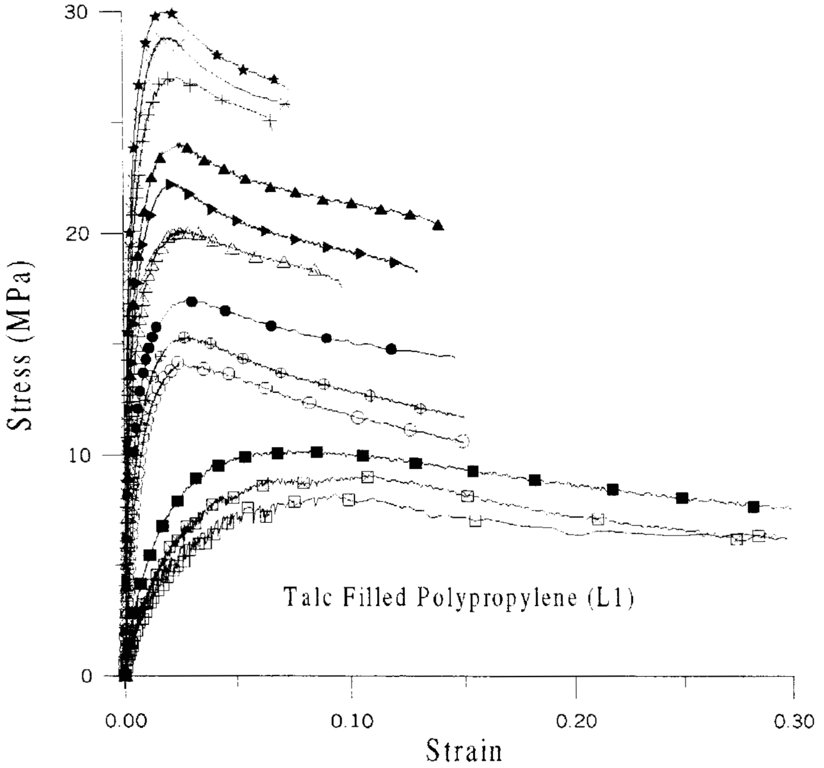


Fig. 3. Stress-strain curves of 40 w% talc-filled polypropylene in the flow (L) direction at various strain rates and temperatures. Symbols are shown in Fig. 2.

Table 1. Tensile Properties of Unfilled Polypropylene.

		Parallel to the Flow Direction (L Direction)			Normal to the Flow Direction (W Direction)		
Strain Rate (min^{-1})	Temp (°C)	Modulus (GPa)	Yield Strength (MPa)	Yield Strain (%)	Modulus (GPa)	Yield Strength (MPa)	Yield Strain (%)
0.05	21.5	1.61	35.16	13.49	1.70	36.91	11.92
0.5		1.76	38.23	9.76	1.85	38.82	9.23
5		1.89	39.81	8.58	1.97	41.24	8.32
0.05	50	0.76	25.81	15.95	0.91	26.74	16.71
0.5		0.88	28.02	12.10	1.01	29.32	13.65
5		0.99	30.60	13.61	1.28	31.42	12.77
0.05	75	0.44	18.43	17.04	0.33	20.40	17.62
0.5		0.51	20.46	14.46	0.40	22.14	15.07
5		0.62	22.11	12.77	0.62	24.42	13.87
0.05	100	0.25	14.77	14.60	0.33	16.84	13.84
0.5		0.38	16.29	12.55	0.41	18.35	12.20
5		0.46	17.38	12.82	0.45	19.41	14.33

Table 2. Tensile Properties of 40 w% Talc-Filled Polypropylene.

		Parallel to the Flow Direction (L Direction)			Normal to the Flow Direction (W Direction)		
Strain Rate (min^{-1})	Temp (°C)	Modulus (GPa)	Yield Strength (MPa)	Yield Strain (%)	Modulus (GPa)	Yield Strength (MPa)	Yield Strain (%)
0.05	21.5	7.69	27.32	2.30	7.43	22.18	1.71
0.5		7.71	28.48	2.11	8.00	24.10	1.88
5		9.30	29.66	2.02	8.23	25.22	1.67
0.05	50	5.05	20.50	2.65	4.42	18.48	2.83
0.5		6.29	22.12	2.26	5.64	19.91	1.98
5		7.10	23.40	2.33	6.45	20.68	1.99
0.05	75	2.98	15.27	3.14	0.82	13.30	8.83
0.5		3.21	15.71	2.87	0.85	14.82	7.64
5		3.38	16.80	2.94	0.92	15.79	6.95
0.05	100	0.45	11.65	10.64	0.61	11.55	8.39
0.5		0.51	13.00	11.98	0.72	12.40	8.33
5		0.81	13.52	8.64	0.73	13.05	8.72

4. STRAIN RATE SENSITIVITY AND TEMPERATURE SENSITIVITY OF UNFILLED AND TALC-FILLED POLYPROPYLENE

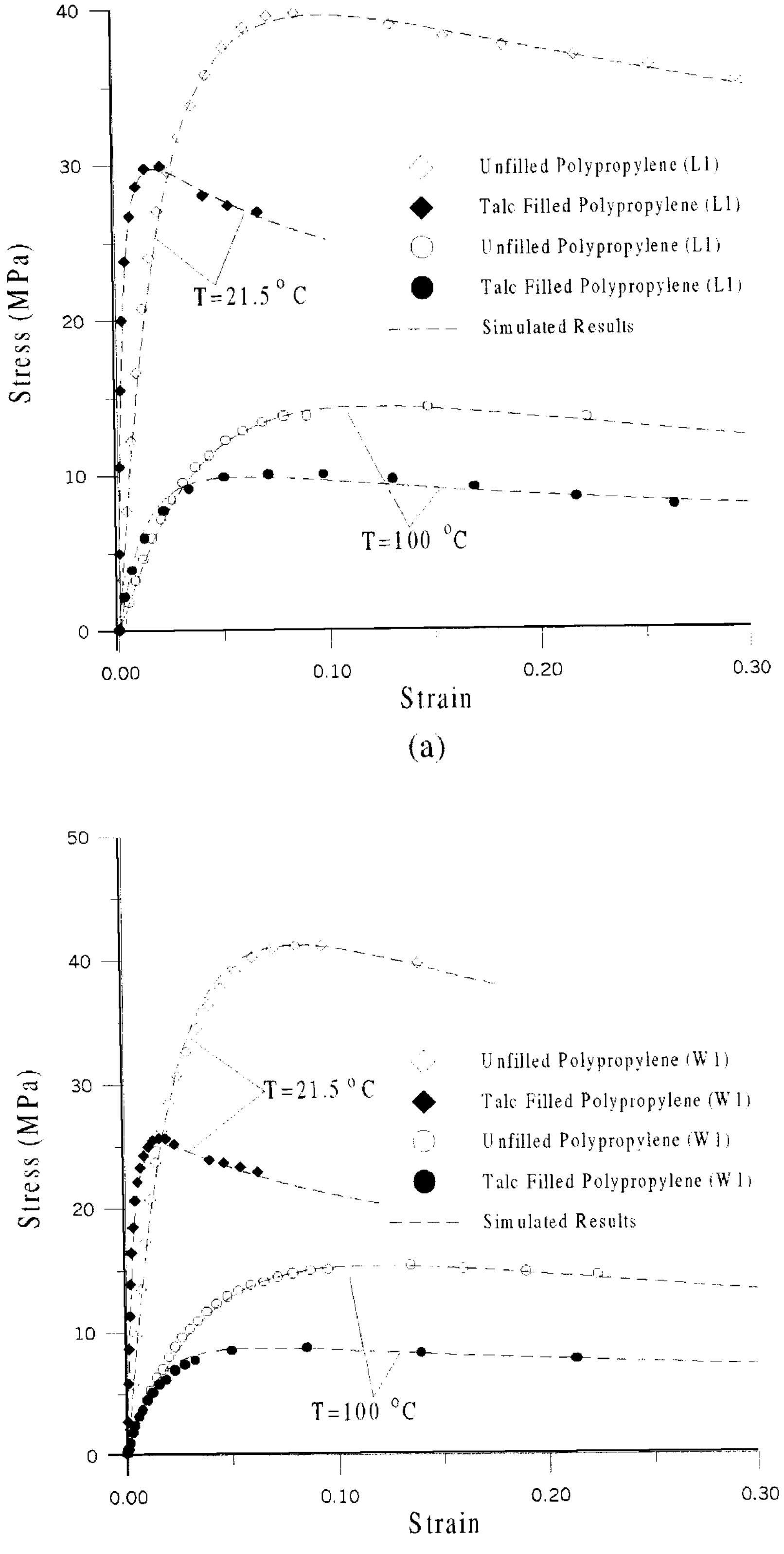
From *Figs. 2* and *3* and *Tables 1* and *2*, it can be concluded that both unfilled polypropylene and 40 wt% talc-filled polypropylene were temperature and strain rate sensitive materials. *Figure 6* shows the variation of modulus E and yield strength σ_Y with $\ln\dot{\varepsilon}$ at various temperatures for unfilled polypropylene and talc-filled polypropylene in the flow (L) direction. Similar variation was observed normal to the flow (W) direction. *Figure 6* shows that the relationships between E and $\ln\dot{\varepsilon}$ as well as σ_Y and $\ln\dot{\varepsilon}$ at the temperatures investigated can be represented by single straight lines, the slopes of which give information about the strain rate sensitivities of modulus and yield strength, respectively. The manner in which modulus and yield strength decreased with increasing temperature can be seen more clearly by plotting E and σ_Y directly as a function of test temperature (*Fig. 7*). The following relationships were found to fit the modulus and yield strength data and represent the temperature and strain rate sensitivities of these two materials.

$$E = E_0\left(1 + m_1 \ln\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)\exp[-\lambda_1(T - T_0)] \quad (1)$$

$$\sigma_Y = \sigma_{Y0}\left(1 + m_2 \ln\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)\exp[-\lambda_2(T - T_0)] \quad (2)$$

where, E_0, σ_{Y0}, $\dot{\varepsilon}_0$ and T_0 are reference elastic modulus, reference yield strength, reference strain rate and reference temperature, respectively. Two other parameters, $m_{1,2}$ and $\lambda_{1,2}$, appearing in *Eqs 1* and *2* are defined as strain rate strengthening coefficient and thermal softening coefficient, respectively. Mathematically, they are defined as

$$m_{1,2} = \frac{\partial(E, \sigma_y)}{\partial \ln \dot{\varepsilon}} \quad (3)$$



(a)

(b)

Fig. 4. Comparison of stress-strain curves of unfilled polypropylene and 40 w% talc-filled polypropylene at 21.5 and 100°C and strain rate of 5 min^{-1} (a) in the flow (L) direction and (b) normal to the flow direction.

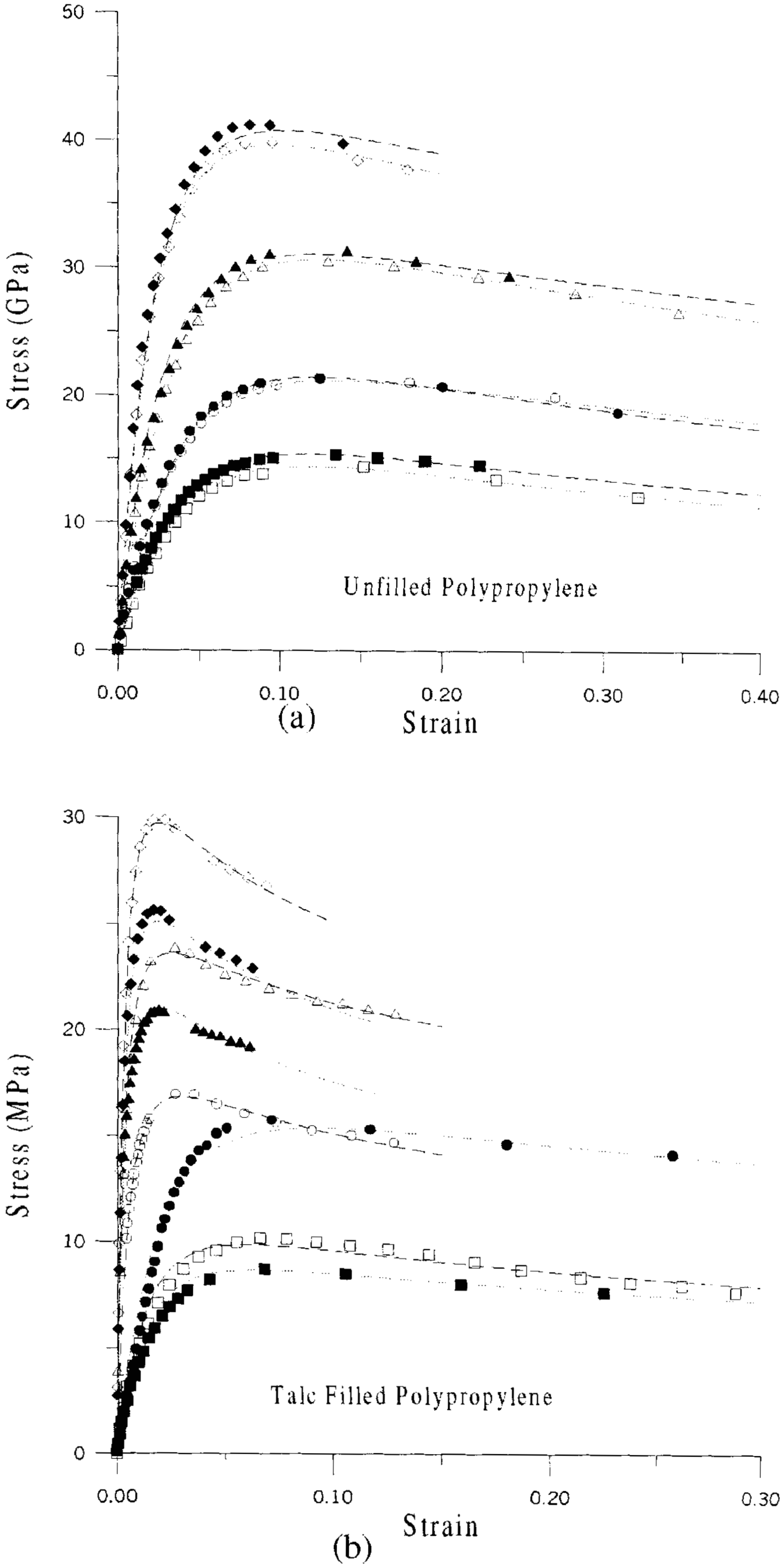


Fig. 5. Comparison of stress-strain curves in the flow (L) direction and normal to the flow (W) direction at various temperatures and strain rate of 5 min^{-1} (a) unfilled polypropylene and (b) 40 w% talc-filled polypropylene. ◇ 21.5°C (L direction), ◆ 21.5°C (W direction), △ 50°C (L direction), ▲ 50°C (W direction), ○ 75°C (L direction), ● 75°C (W direction), □ 100°C (L direction), ■ 100°C (W direction).

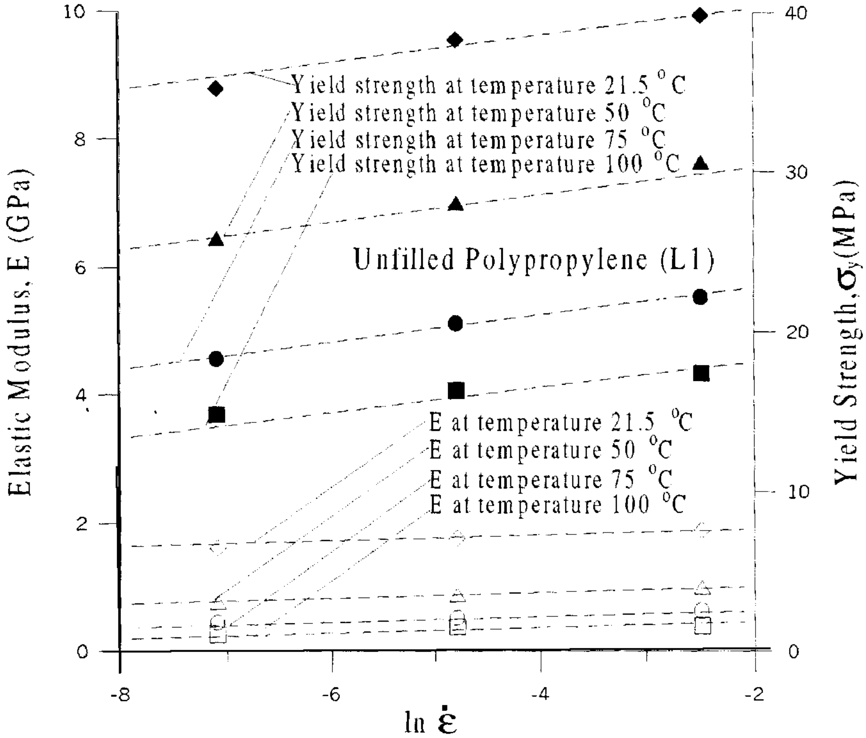


Fig. 6a. Effect of strain rate on elastic modulus and yield strength of unfilled polypropylene in the flow (L) direction.

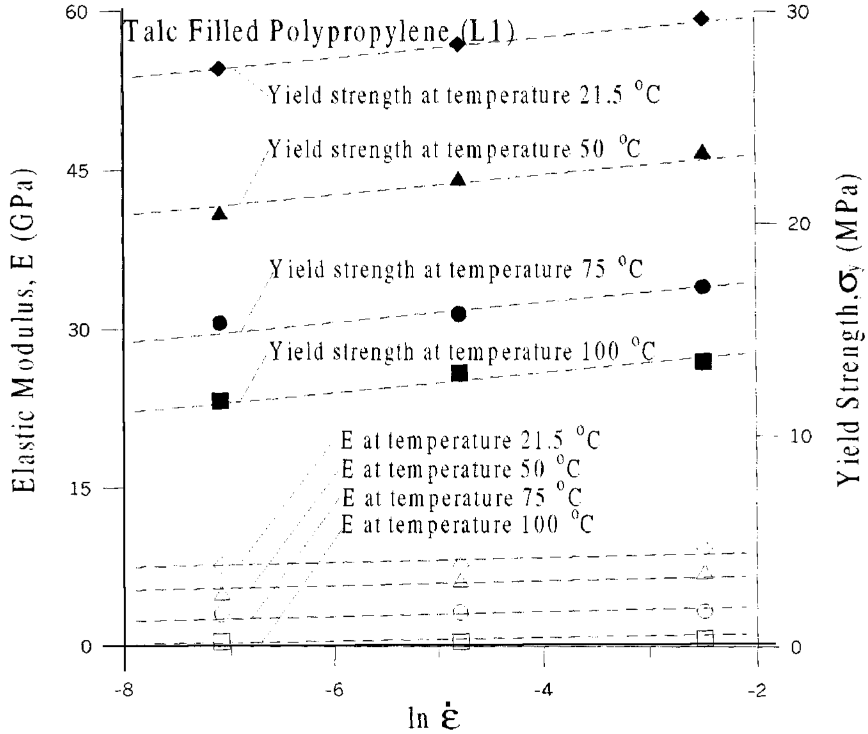


Fig. 6b. Effect of strain rate on elastic modulus and yield strength of 40 w% talc-filled polypropylene in the flow (L) direction.

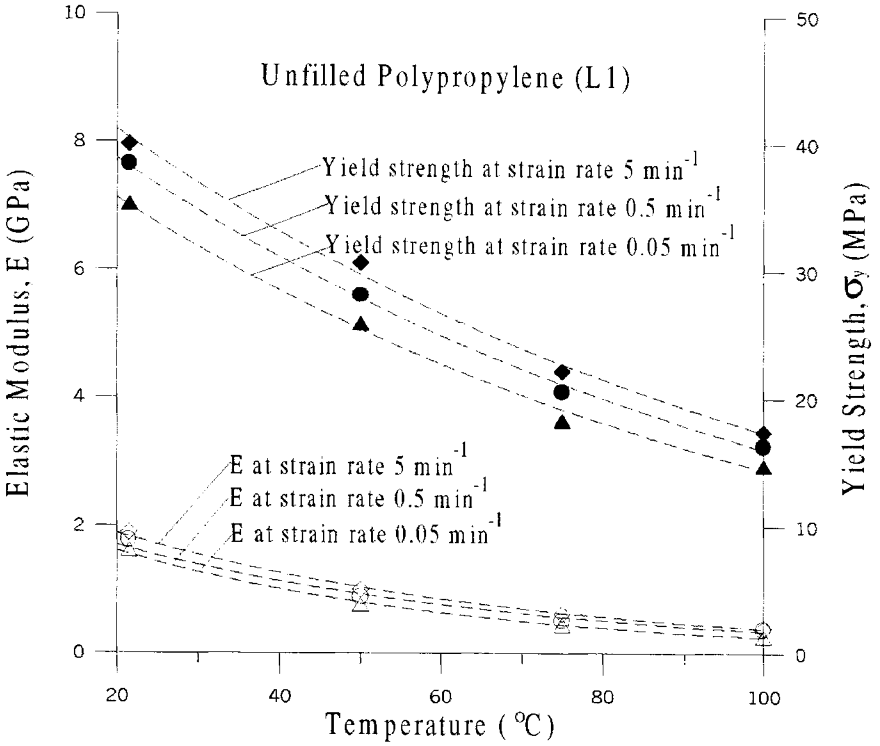


Fig. 7a. Effect of temperature on elastic modulus and yield strength of unfilled polypropylene in the flow (L) direction.

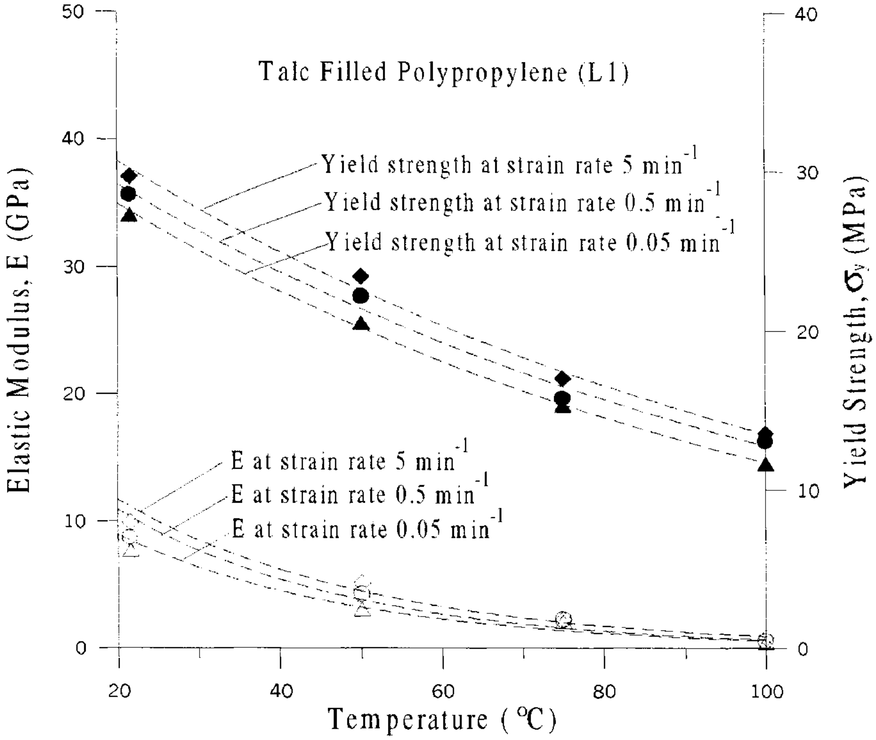


Fig. 7b. Effect of temperature on elastic modulus and yield strength of 40 w% talc-filled polypropylene in the flow (L) direction.

$$\lambda_{1,2} = -\frac{\partial \ln(E, \sigma_Y)}{\partial T} \qquad (4)$$

The values of $m_{1,2}$ and $\lambda_{1,2}$ are calculated from the experimental results using the least square method and are given in *Table 3*. The strain rate sensitivity and temperature sensitivity values are given over the strain rates and temperature range considered here. From *Table 3*, it can be observed that talc filler not only increased the tensile modulus, but also increased the strain rate sensitivity and temperature sensitivity of modulus. In both L and W directions, the strain rate strengthening coefficient, m_1, and thermal softening coefficient, λ_1, for the elastic modulus of talc-filled polypropylene were much higher than those of unfilled polypropylene. On the other hand, talc filler not only reduced the yield strength, but also reduced the strain rate sensitivity and temperature sensitivity of yield strength. The strain rate strengthening coefficient, m_2, of talc-filled polypropylene was lower by 44.4% in the L direction and by 40.8% in the W direction. The thermal softening coefficient, λ_2, of the talc-filled polypropylene was lower by 4.89% in the L direction and by 11.3% in the W direction. The dashed lines in *Figs. 6* and *7* are simulated results, which fit the experimental data well.

5. DISCUSSION

Yielding in polymers is considered an energy activated rate dependent phenomenon (9). The yield strength of a polymer is represented by the Eyring equation:

$$\sigma_y = \frac{Q}{V} + \frac{RT}{V}\ln\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right) \qquad (5)$$

where, σ_y = yield strength

Q = activation energy

V = activation volume

R = Gas constant

T = temperature

$\dot{\varepsilon}$ = strain rate

$\dot{\varepsilon}_0$ = reference strain rate

According to *Eq 5*, plots of yield strength σ_y vs. $\ln\dot{\varepsilon}$ at different temperatures should produce parallel straight lines, which is the case as seen in *Fig. 6*. Each of these lines has the same slope, which means that (RT/V) is constant.

To determine the activation volume, activation energy and reference strain rate at each temperature, yield strength data at that temperature are first plotted against $\ln\dot{\varepsilon}$. The slope of this plot is (RT/V). The activation volume V is determined from this slope. The intercept of this plot at $\dot{\varepsilon} = 1$ is equal to $\left(\frac{Q}{V} - \frac{RT}{V}\ln\dot{\varepsilon}_0\right)$.

Since (RT/V) is constant, a plot of $\left(\frac{Q}{V} - \frac{RT}{V}\ln\dot{\varepsilon}_0\right)$ vs. $(1/V)$, if linear, gives the value of the activation energy Q. *Figure 8* shows that these plots for unfilled and talc-filled polypropylenes in the L and W directions are indeed linear.

On the basis of σ_y vs. $\ln\dot{\varepsilon}$ curves in *Fig. 6*, the activation volumes of unfilled and talc-filled polypropylenes were first calculated. Then, following the procedure mentioned above, the activation energies were calculated from the slopes of the straight lines in *Fig. 8*. The activation volumes and activation energies are listed in *Tables 4* and *5*, respectively. In these tables, one can observe that the activation volumes of both unfilled and talc-filled polypropylenes increased with increasing temperature and the activation volumes of talc-filled polypropylene were significantly higher than those of unfilled polypropylene. Note that the activation volume of unfilled polypropylene at 21.5°C is twice the value reported Hartmann *et al.* (2). The activation energy was, as expected, independent of temperature; however, the activation energy of talc-filled polypropylene was greater than that of unfilled polypropylene. Furthermore, the activation energies of both unfilled and talc-filled polypropylenes were greater in the flow or L direction than normal to the flow or W direction. The activation volumes, on the other hand, did not change much from the L-direction to the W-direction and, therefore, can be considered direction independent.

6. CONCLUSIONS

Uniaxial tensile tests were conducted on unfilled polypropylene and 40 w% talc-filled polypropylene at four different temperatures and three different strain rates. Based on the analysis of the experimental data, the following conclusions are reached:

Table 3. Strain Rate Strengthening Coefficients (m_1 and m_2) and Thermal Softening Coefficients (λ_1 and λ_2) of Unfilled Polypropylene and Talc-Filled Polypropylene.

	Unfilled Polypropylene (L)	40 w% Talc-Filled Polypropylene (L)	Unfilled Polypropylene (W)	40% Talc-Filled Polypropylene (W)
m_1 (for elastic modulus)	0.0452	0.2403	0.0566	0.1656
λ_1 (for elastic modulus)	0.0202	0.0328	0.0212	0.03503
m_2 (for yield strength)	0.8638	0.4800	0.8471	0.5011
λ_2 (for yield strength)	0.0110	0.0105	0.0098	0.00871

Note: L and W represent parallel to the flow and normal to the flow directions, respectively.

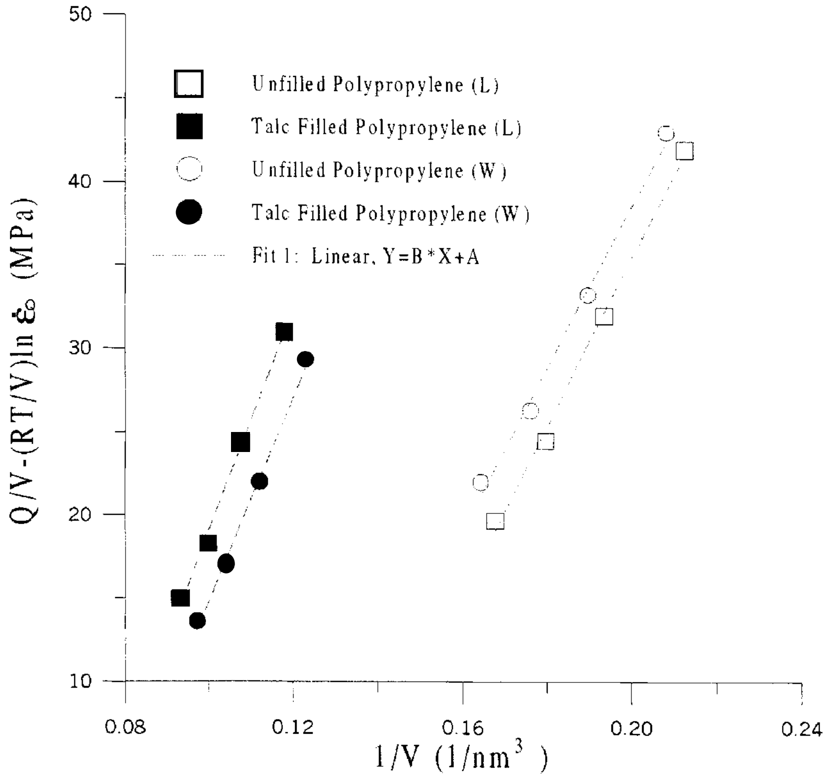


Fig. 8. Plots to determine activation energies.

Table 4. Activation Volumes in (*nm*)^3 at Different Temperatures.

Material	Temperature (°C) 21.5	50	75	100
Unfilled Polypropylene (L)	4.706	5.162	5.563	5.961
40 w% Talc-Filled Polypropylene (L)	8.469	9.289	10.01	10.73
Unfilled Polypropylene (W)	4.799	5.263	5.671	6.078
40 w% Talc-Filled Polypropylene (W)	8.113	8.898	9.586	10.28

Note: L and W represent parallel to the flow and normal to the flow directions, respectively.

Table 5. Activation Energies and Reference Strain Rates.

Material	Unfilled Polypropylene (L)	40 w% Talc-Filled Polypropylene (L)	Unfilled Polypropylene (W)	40 w% Talc-Filled Polypropylene (W)
Activation Energy (kJ/mol)	301.4	396.1	291.4	365.2
Reference Strain Rate (min^{-1})	1.159×10^8	2.147×10^{10}	2.028×10^7	6.001×10^9

Note: L and W represent parallel to the flow and normal to the flow directions, respectively.

1. Unfilled polypropylene and talc-filled polypropylene are typical strain rate and temperature dependent materials. Both elastic modulus and yield strength of unfilled polypropylene and talc-filled polypropylene decreased with increasing temperature and increased with increasing strain rate.
2. Talc-filled polypropylene exhibited significantly different tensile properties in the flow direction of injection molding than normal to the flow direction. The difference in tensile properties of unfilled polypropylene in these two directions was relatively small.
3. The talc filler increased the tensile modulus of polypropylene, but decreased both yield strength and yield strain. Talc filler increased the strain rate and temperature sensitivities of tensile modulus and decreased the strain rate and temperature sensitivities of yield strength.
4. Yield strength of both unfilled polypropylene and talc-filled polypropylene can be predicted using an energy activated rate dependent Eyring equation. Both activation volume and activation energy of talc-filled polypropylene were higher than those of unfilled polypropylene. Also, activation energy showed direction dependence, while activation volumes were direction independent.

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