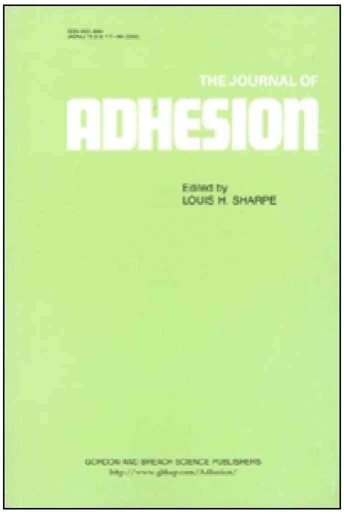

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

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To cite this Article Lin, C. B. , Lee, Sanboh and Liu, K. S.(1991) 'The Microstructure of Solvent-Welding of PMMA', The Journal of Adhesion, 34: 1, 221 — 240
To link to this Article: DOI: 10.1080/00218469108026516
URL: http://dx.doi.org/10.1080/00218469108026516

J. Adhesion, 1991, Vol. 34, pp. 221–240
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Printed in the United Kingdom

The Microstructure of Solvent-Welding of PMMA

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(Received September 17, 1990; in final form February 26, 1991)

The microstructure of solvent-welding of PMMA has been investigated. The morphologies of shear, tensile, tear and cleavage fracture surfaces are observed. Effects of solubility parameter, elevated temperature, and cosolvent on the mechanical strength are studied. The solvents used are methanol, ethanol and DMF. The feature sizes of shear, tensile and tear fracture surfaces and the angle of the cleavage curve at the interface of PMMA can be employed to determine the mechanical strength qualitatively. Constituents of shear, tensile and tear fracture surfaces are tongues, long and narrow hackles, and equiaxed hackles, respectively. PMMA treated at high temperature showed enhanced mechanical strength. PMMA treated with 95% by volume ethanol yielded the best quality of adhesion. The solvent affected zone and the residual solvent are also discussed.

KEY WORDS PMMA; solvent-welding; shear; tensile; tear; cleavage.

1. INTRODUCTION

The phenomenon of structural adhesion in polymeric materials is well known but not well understood. Wake[1] and Allen[2] categorized four mechanisms of adhesion as mechanical interlocking, diffusion theory, electronic theory, and adsorption. Gauthier[3] sorted out the structural adhesives based on supplier, chemical reactivity properties, etc., and defined a structural adhesive as a material used to transfer load between adherends in the service environment. Structure adhesives are not the only media used to promote the adhesion of polymeric components. An alternative approach for inducing adhesion is solvent-welding. When both surfaces of a polymeric material are treated with a suitable solvent in the absence of an extraneous adhesive and when, after a short period, the two surfaces are brought together under small pressure, the assembled adherends can be stressed to a large proportion of the maximum failure load for a long time without failure. The purpose of the solvent is to cause the polymer chains of the two adherends to interdiffuse with each other, not to transfer load from one adherend to the other. Titow[4] joined two

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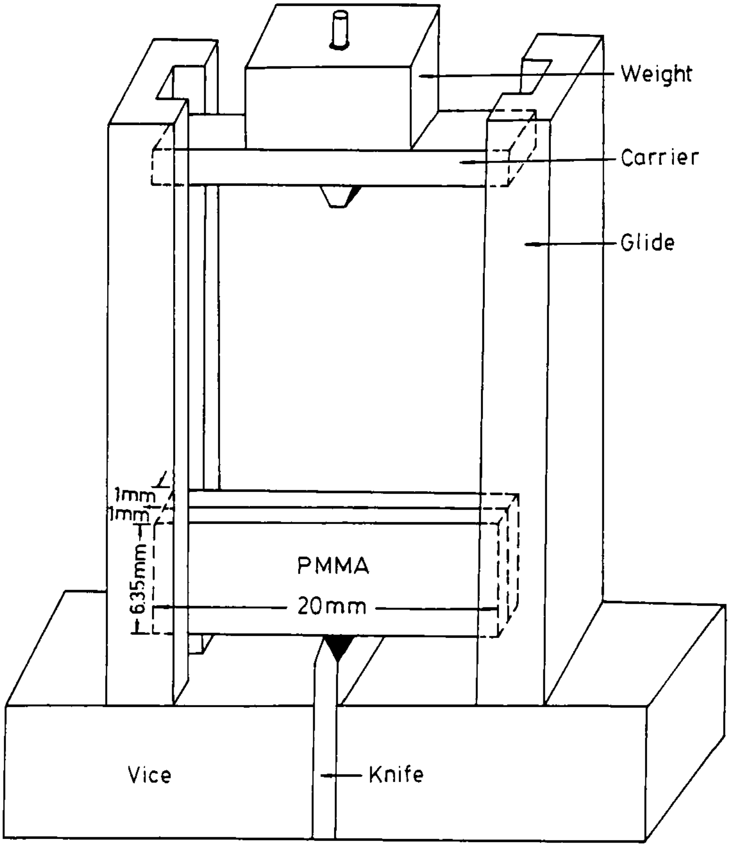
*To whom correspondence should be addressed polycarbonates or polysulphones using solvent-welding and observed that the residual solvent concentrated within the bonding zone. Menges *et al.*[5] and Yue and Cherry[6] found that, in addition to the solvent affected zone (SAZ) observed by Titow *et al.*,[7] a soft residual final adhesive layer (FAL) was sandwiched between two SAZ. Yue[8] also found that the extent of solvent-welding depends on the ability of solvent to dissolve the polymer but is not related to the solvent affected zone in the polymer. Based on the model of critical principal strain,[9] Yue and Chui[10] concluded that a large critical principal strain implied a high intrinsic weld strength. Yue[11] also studied the structure and strength of solvent welds between dissimilar amorphous thermoplastics. In order to promote the adhesion, the solvent is required to plasticize the surface of polymer strongly, *i.e.*, to increase the rate and the extent of inter-diffusion of the polymer chains. The implication is that only good solvent be employed for this purpose, at temperatures above the glass transition temperature of polymers. Although good solvents are suitable for solvent-welding of polymeric materials, they may be noxious and harmful.

On the other hand, on the basis of the reptation model of chain dynamics proposed by de Gennes[12] and Doi and Edwards,[13] Wool and coworkers[14–17] found that when polymeric materials with a crack or a craze were placed inside a furnace at a temperature above the glass transition point for some period of time, the mechanical strength recovered to a high proportion of the original strength. Instead of employing a temperature above the glass transition point, Lin *et al.*[18] observed healing in a crack in PMMA using methanol at temperatures of 40–60°C. Methanol is a non-solvent and the temperature range is below the glass transition point, 104°C. These observations stimulated us to study the solvent-welding of PMMA. The purpose of this study is to understand the mechanism of solvent-welding from the point of view of microstructure such as the morphologies associated with cleavage, tear, tensile and shear fracture. The effects of type and concentration of solvent and solvent-welding temperature are included in this study.

2. EXPERIMENTAL

PMMA with inherent viscosity 0.237 dl/g was obtained from DuPont in the form of a 6.35mm thick Lucite®L type cast acrylic sheet. Pieces of dimensions 20mm × 6.35mm × 1.0mm for cleavage and tear tests, 20mm × 6.35mm × 2.0mm and 60mm × 6.35mm × 2.0mm for tensile tests, and 80mm × 6.35mm × 2.0mm for shear tests were cut from the sheet. They were polished with 600 and 1000 grit Carbimet papers and with 1 μm and 0.05 μm alumina slurries. Finally, samples were annealed in air for 24 hr at 120°C and cooled in the furnace to an approximate temperature of 26°C.

The three solvents used were methanol, ethanol and N,N-Dimethyl-foramide (DMF). All were of analytical grade. The properties of these solvents are listed in Table I. The solvent was applied to the surfaces of the PMMA. The two wetted surfaces were then brought together and the specimen was mounted, under compression, in one of the welding instruments for cleavage, tear, tensile, and shear tests as shown in Figs. 1(a)–(d). The specimen with the welding instrument was put



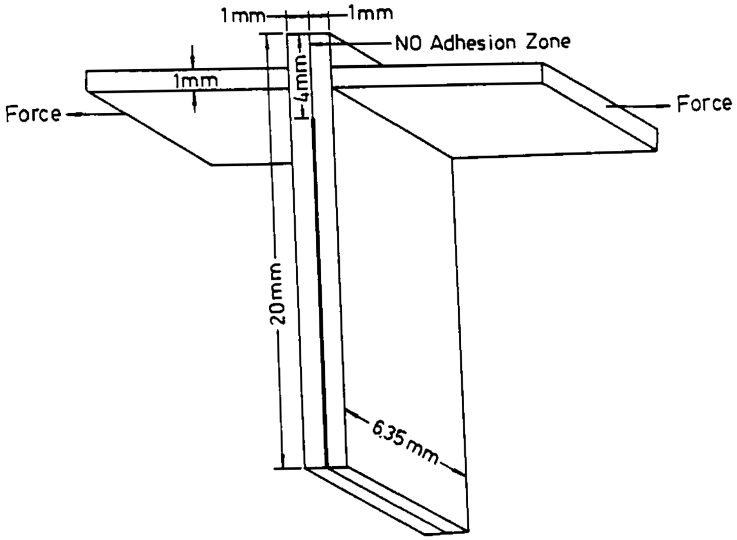
(a)

See Figure 1 Caption on page 225.

into a thermostatted furnace for two minutes and then removed from the furnace. Then the specimen was taken out of the welding instrument and aged for one day at room temperature.

Effects of temperature and solvent concentration on adhesion were also studied. Ethanol was used as a solvent for welding because it is the least harmful solvent in this study. Since the boiling point of ethanol is 78°C, the solvent-welding was undertaken at temperatures of 80°C–100°C, under a compression load of 1.5 kg, for 2 minutes. The purpose of the compression load is to put both surfaces firmly in contact so that the molecules of PMMA will penetrate adjacent interfaces. Water, which does not penetrate PMMA well, was used to dilute the ethanol in order to study the effect of ethanol concentration on solvent-welding. The volume fraction of ethanol in the cosolvent was in the range 80%–100% by volume. Solvent-welding was undertaken at a temperature of 90°C.

Specimens for tear, tensile and shear tests were mounted on a universal tensile



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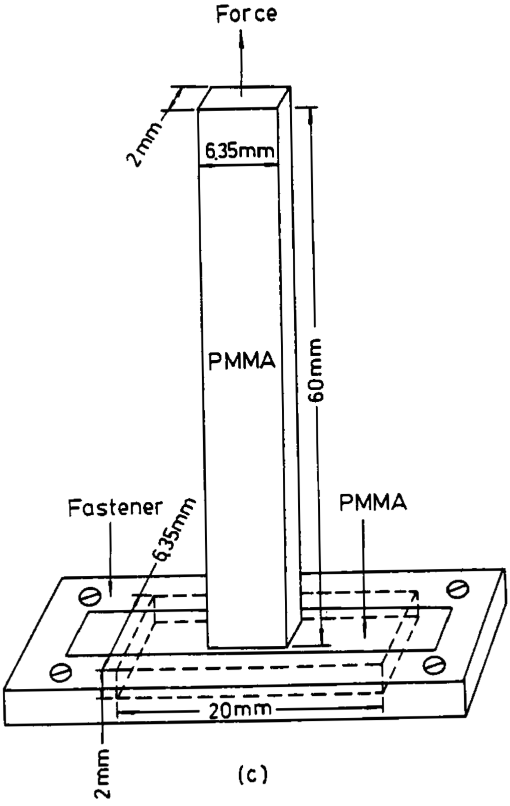

(c)

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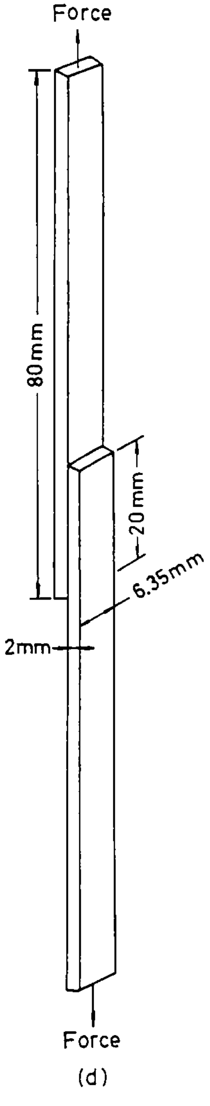


FIGURE 1 Schematic diagrams of various solvent-welding testing instruments: (a) cleavage test; (b) tear test; (c) tensile test; and (d) shear test.

testing machine with a cross head speed of 0.05 mm/sec. Tests were run at room temperature. The cleavage specimen was mounted on the cleavage testing machine as shown in Fig. 1(a). The fracture surface morphologies of the different modes were observed using a Leitz Metallux 3 optical microscope.

3. RESULTS AND DISCUSSION

3.1 Solubility Parameter Effect

The solubility parameters and the hydrogen bonding parameters for methanol, ethanol and DMF as listed in Table I were obtained from Vincent and Raha.[19] Comparing the solubility parameter of PMMA (9.2) obtained from Vincent and Raha[19] with those of the three solvents (Table I), we find that the solubility parameter of DMF is closest to that of PMMA.

It is known that methanol and ethanol are non-solvents for PMMA at room temperature. However, when the temperature is increased to 87°C, both become good solvents.[20] Thus, in order to assure that all the solvents act as good solvents, the temperature used for solvent-welding was 90°C. At 90°C the solvents can easily interact with the PMMA and generate a tacky layer on the surface. For the purpose of understanding the adhesion, the solvent-welding was undertaken using one of the welding instruments for the shear, tensile, tear and cleavage tests.

Figures 2(a)–(c) show the morphologies of the shear fracture surfaces of the PMMA treated with methanol, ethanol and DMF, respectively. It is found that the common feature of these shear fracture surfaces is a tongue. This feature is shown enlarged in the inset of Fig. 2(c). The size of the tongue follows the sequence from small to large for joints made with methanol, ethanol and DMF, respectively. The shear strengths of the PMMA treated with methanol, ethanol and DMF are listed in Table II. In comparing the morphology and the shear strength, it is seen that a larger tongue corresponds to a higher shear strength.

Figures 3(a) and (b) show the morphologies of tensile fracture surfaces of the PMMA treated with methanol and ethanol, respectively. The shape of the hackles corresponding to the methanol- and ethanol-treated samples is narrow and long. The size of the hackle corresponding to the ethanol-treated sample is larger than that corresponding to the methanol-treated sample. The morphology of the tensile fracture surface of the PMMA sample treated with DMF shown in Fig. 4(c) has

TABLE I
Specifications of three solvents

Solvent	Methanol	Ethanol	N,N-Dimethylformamide
Molecular weight (g/mole)	32	46	73
Solubility parameter $(\mathrm{cal/cm^3})^{1/2}$	14.3	12.9	12.1
Hydrogen bonding parameter $(\mathrm{cal/m^3})^{1/2}$	19.8	17.7	11.7
Boiling point (°C)	64.7	78.2	151–156
Density (g/cc) at 20°C	0.801	0.797	0.949–0.954

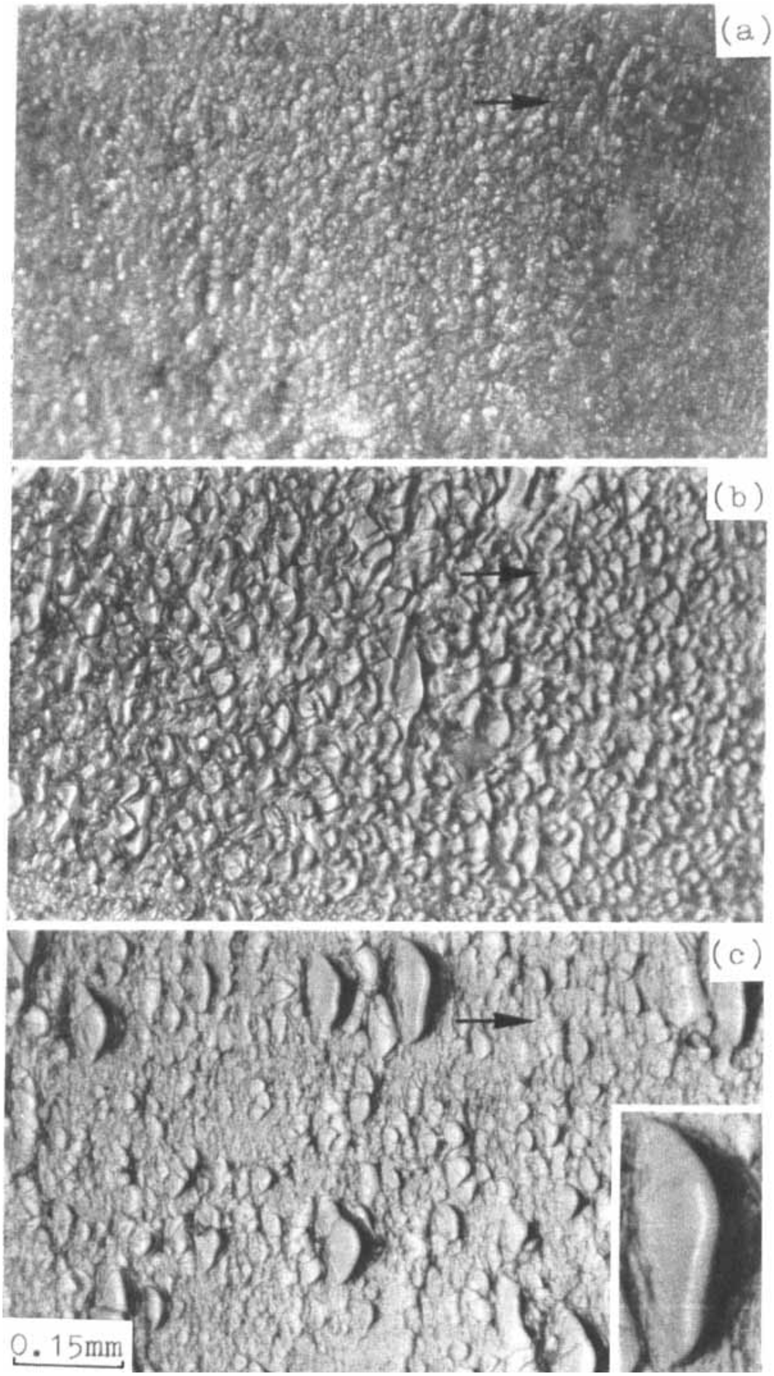


FIGURE 2 The morphologies of different shear fracture surfaces for solvent (a) methanol; (b) ethanol; and (c) DMF, where the arrow indicates the shear direction.

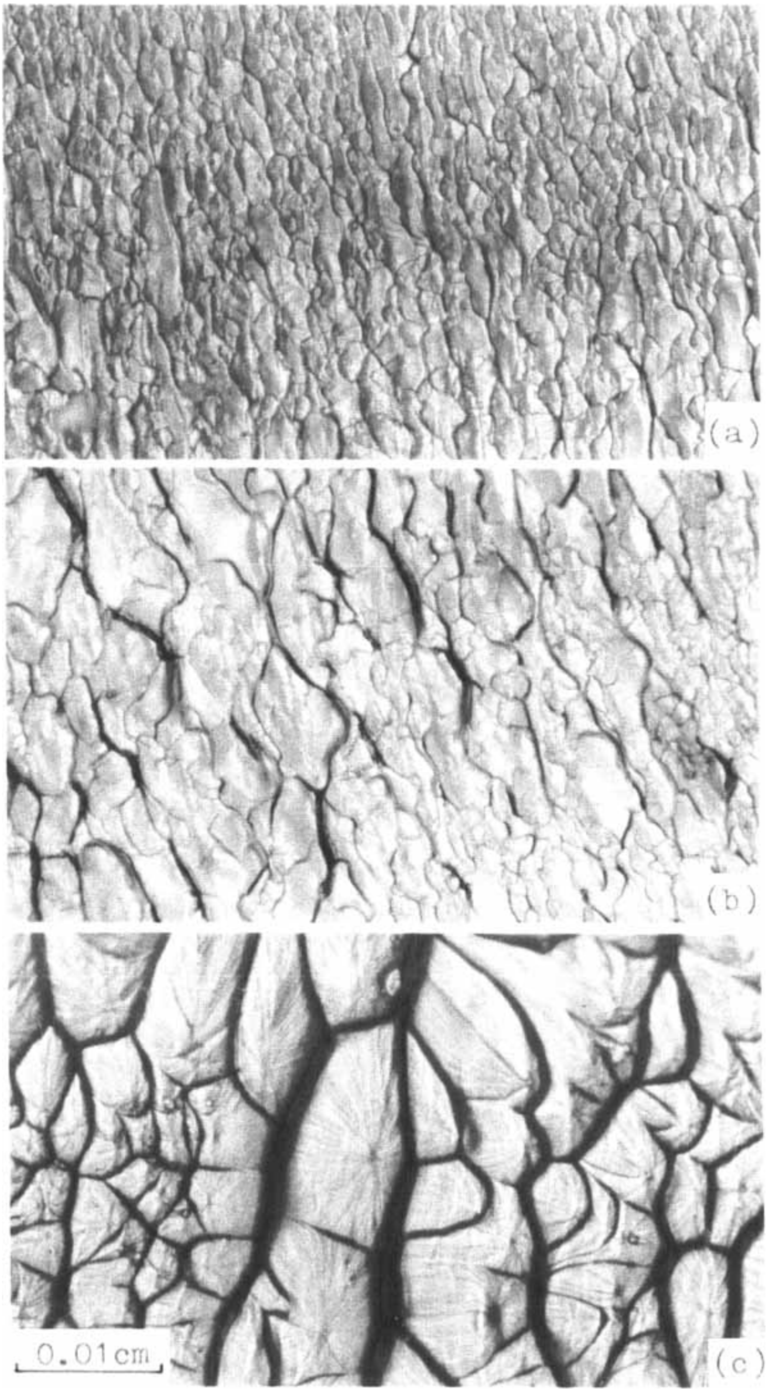


FIGURE 3 The morphologies of different tensile fracture surfaces for solvent (a) methanol; (b) ethanol; and (c) DMF.

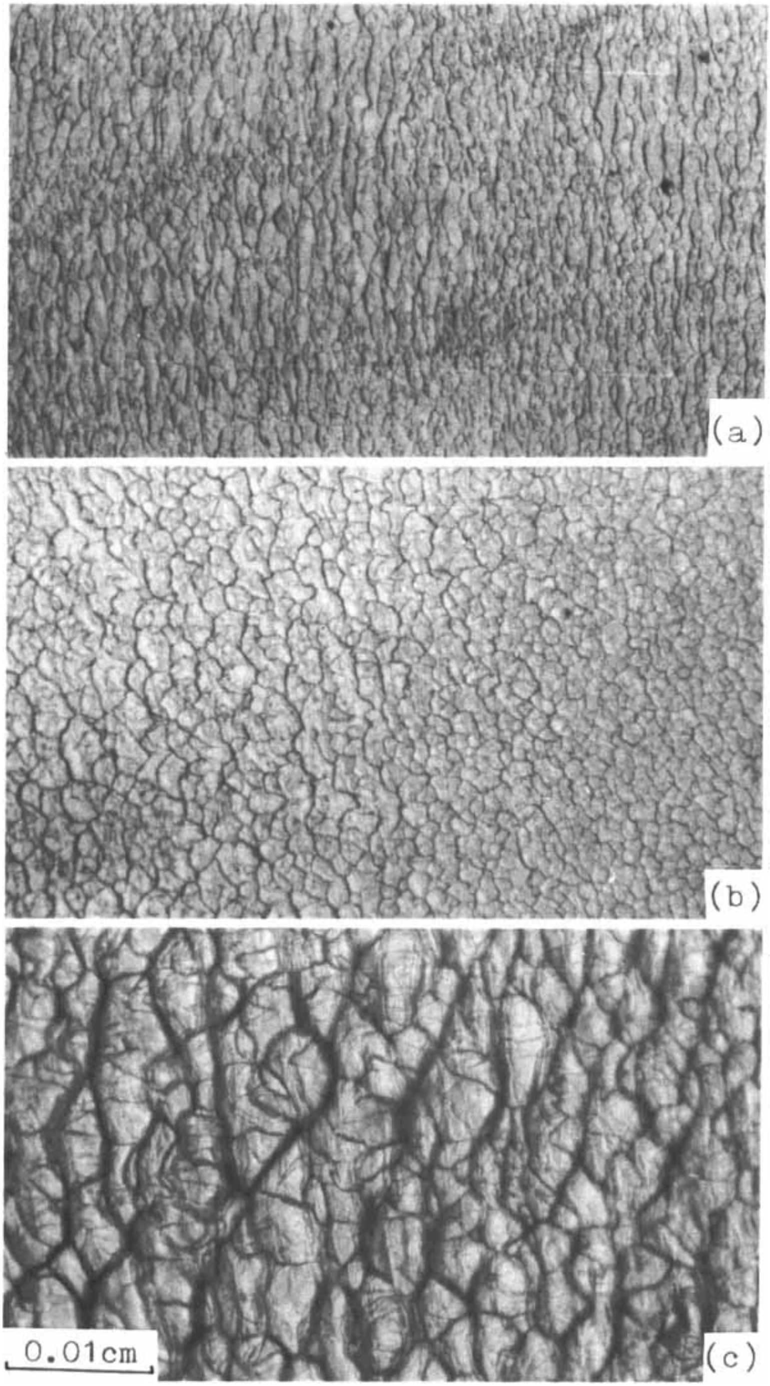


FIGURE 4 The morphologies of different tear fracture surfaces for solvent (a) methanol; (b) ethanol; and (c) DMF.

TABLE II
Effect of solubility parameter

The shear fracture stress τ, tensile fracture stress σ_{tn}, tear fracture stress σ_{tr} and the angle θ of cleavage curve at the interface of PMMA with and without solvent treatment at 90°C under a compression load 1.5 Kg.

Solvent	Methanol	Ethanol	DMF	Virgin
τ (MPa)	9.75	13.61	22.41	61.8*
σ_{tn} (MPa)	2.4	4.9	9.7	74.5**
σ_{tr} (MPa)	3.67	4.17	7.04	25.2
θ	26°	12°	0°	

Note: * and ** were obtained by manufacturer based on ASTM standards D732 and D638, respectively.

many elliptical paraboloids, with striations emanating from the vertex of each paraboloid. The tensile strengths of the PMMA samples treated with methanol, ethanol, and DMF are tabulated in Table II. Comparing the tensile strength with the morphology, we find that the feature size of the tensile fracture morphology increases with the tensile strength.

Figures 4(a)–(c) illustrate the morphologies of the tear fracture surfaces of PMMA treated with methanol, ethanol, and DMF, respectively. The common features are the hackles which have shapes which are narrow and long, small equiaxed, and large equiaxed, corresponding to those treated with methanol, ethanol and DMF, respectively. The tear fracture stresses of PMMA treated with methanol, ethanol, and DMF are listed in Table II. Comparing the morphology of the tear fracture surface with the corresponding tear fracture stress, we find that the larger size of hackle corresponds to the higher tear fracture stress. The morphology of the tear fracture surface of virgin PMMA is illustrated in Fig. 5. Figures 4 and 5, when compared, illustrate that the tear fracture surface of PMMA treated with solvents is completely different from that without solvent treatment. The mechanical strengths of virgin PMMA are also listed in Table II. Noted that the tensile and shear strengths of virgin PMMA were obtained from DuPont based on ASTM standards D638 and D732, respectively. In summary, the mechanical strengths of PMMA samples treated with solvents are lower than those of virgin PMMA.

The morphologies of cleavage fracture surfaces of PMMA untreated and solvent-treated are illustrated in Figs. 6(a)–(d). When two pieces of the virgin PMMA are pressed together and then cleaved, the cleavage curve or chevron shape of one piece is discontinuous at the interface to that of the other as shown in Fig. 6(a). On the other hand, when both surfaces of PMMA are treated with solvent, brought together and cleaved, the cleavage curve of one PMMA piece is continuous at the interface with that of the other, but their derivatives are not the same at the interface as shown in Figs. 6(b)–(d). The angles,θ, jumped at the interfaces measured from the Figs. 6(b), (c) and (d) are 26°, 12°, and 0° (listed in Table II), respectively. Comparing the mechanical strengths (shear, tensile and tear) and morphologies of the cleavage fracture surfaces, we find that the specimen showing the smallest angle has the highest mechanical strength.

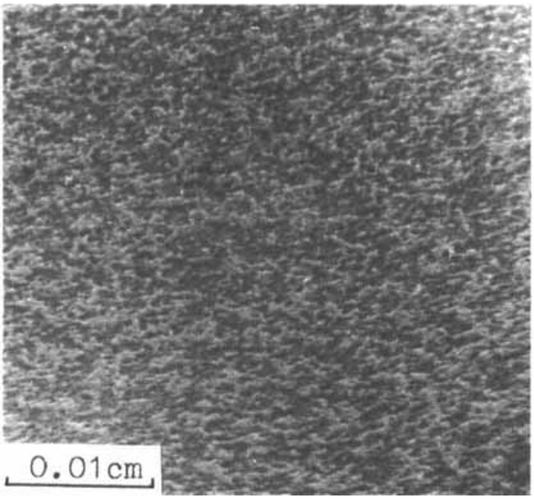


FIGURE 5 The morphology of tear fracture surface of virgin PMMA.

The solvent affected zone arises from the swelling of PMMA so that the boundary between the adherend and solvent affected zone is clearly marked by the depth of solvent penetration into the polymer. The sizes of the solvent affected zones of PMMA treated with methanol, ethanol and DMF are 165 μm, 52 μm, and 20 μm, respectively. It is found that the solvent affected zone increases with decreasing mechanical strengths. On the other hand, it can also be seen from Fig. 6 that the original interface of PMMA treated with solvent is destroyed and an array of irregular holes form along the central line. Note that the region near the interface is magnified and shown on the right hand side of each picture. The region containing the irregular holes is the tacky layer. However, the boundary between the tacky layer and the solvent affected zone cannot be clearly identified. The tacky layer arises from the polymer dissolving in the solvent so that the residual solvent is possibly found inside it. The boiling points of methanol, ethanol and DMF are 64.7°C, 78.2°C, and 151–156°C respectively, as listed in Table I. The sequence of volatility from large to small is methanol, ethanol and DMF. Increasing volatility is associated with decreasing residual solvent in PMMA. Thus the amount of residual methanol in PMMA is less than 0.1 mg, which is the minimum weight measured by our electronic digital balance. The amounts of residual ethanol and DMF in PMMA are 5 wt% and 18 wt%, respectively. Note that the weight percent is defined as the ratio of residual solvent to the original amount which wetted the surface of the PMMA. We conclude that the residual solvent in PMMA increases with the decreasing difference of solubility parameters between solvent and PMMA.

Lin *et al.*[18] stated that the introduction of methanol into PMMA at temperatures of 40–60°C reduced the glass transition temperature of PMMA and produced crack healing. Their mechanism came from the wetting and the interdiffusion of PMMA chains. Wool and coworkers[14–17] have also investigated the crack healing of PMMA

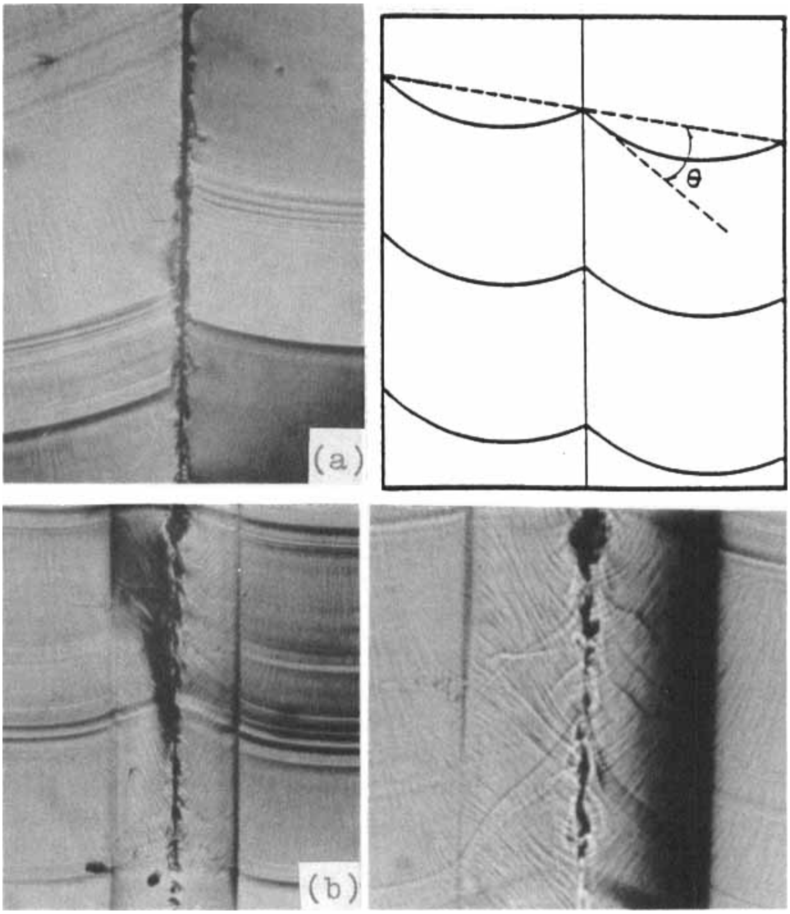
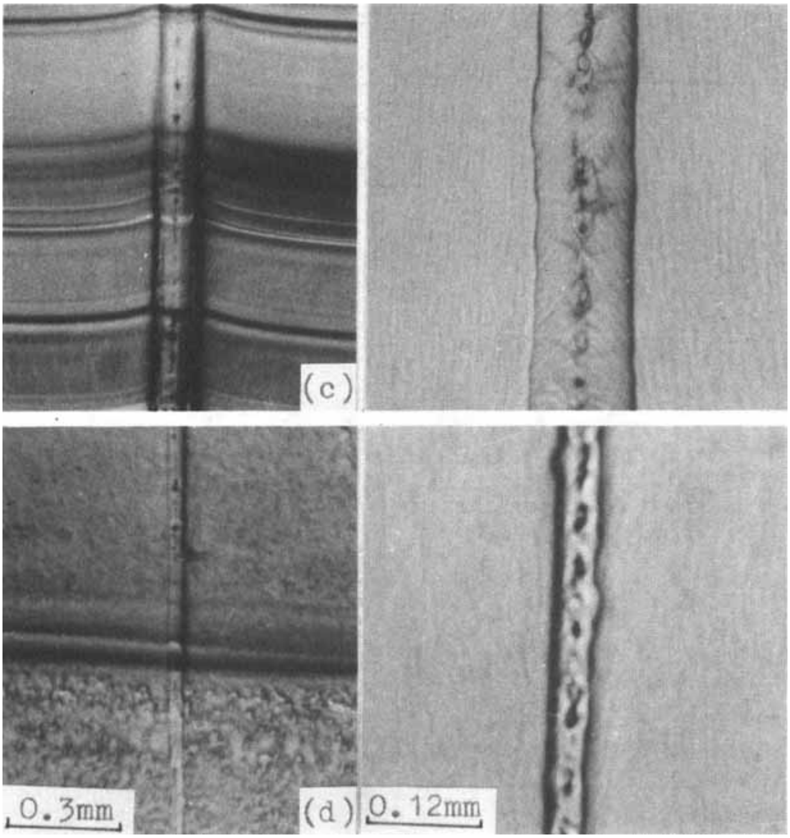


FIGURE 6 The morphologies of different cleavage fracture surfaces: (a) virgin PMMA; and PMMA treated with solvent (b) methanol; (c) ethanol; and (d) DMF. The upper right figure illustrates the angle,θ, of the cleavage curve at the interface of PMMA.

at temperatures above its glass transition point. The common mechanism of their studies can be explained by the reptation model of the polymer chain.[12-13] The major difference between solvent-welding and crack healing is that the mechanical strength of the polymer for solvent-welding relies on two surface layers of polymer dissolved in the solvent forming a tacky layer, and that for crack healing is on the basis of the penetration depth of polymer chains. For the same solvent treatment, the required elevated temperature for the movement of polymer chains is lower than that for production of the tacky layer. Thus, under the same conditions, the elevated temperature for solvent-welding is higher than that for crack healing with the same solvent treatment, but lower than that for crack healing without solvent treatment.



See Figure 6 Caption on page 232.

3.2 Temperature Effect

The solvent welding of PMMA treated with ethanol was undertaken at temperatures of 80–100°C. For temperatures below 80°C the PMMA needs quite a long time to adhere using solvent-welding whereas for temperatures above 100°C the PMMA can be healed without solvent treatment[14–17] because the glass transition point of PMMA is 104°C. The effect of temperature on shear strength is listed in Table III. It can be seen from Table III that the shear strength of PMMA increases with temperature. The common feature of the shear fracture surface is many tongues, the sizes of which increase with temperature. Figure 7(a) shows an example of the morphology of the shear fracture surface of PMMA treated with ethanol at 100°C. The overlap of tongues aligns along the shear direction indicated by the arrow. The tensile strengths of PMMA treated with ethanol at different temperatures are also listed in Table III. It is found that the tensile strength increases with temperature.

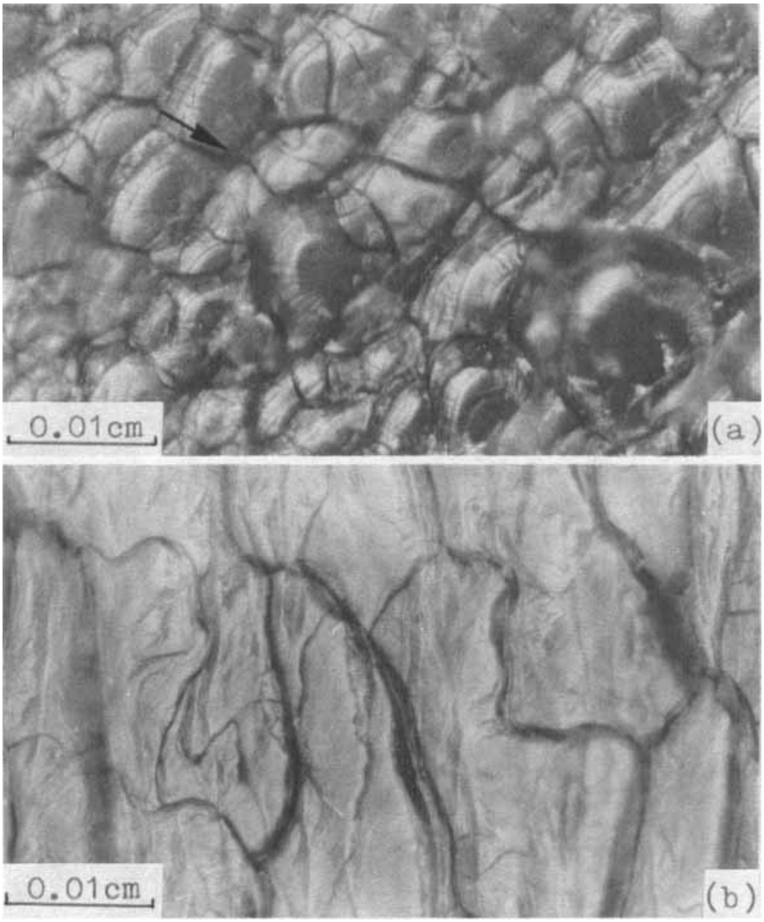
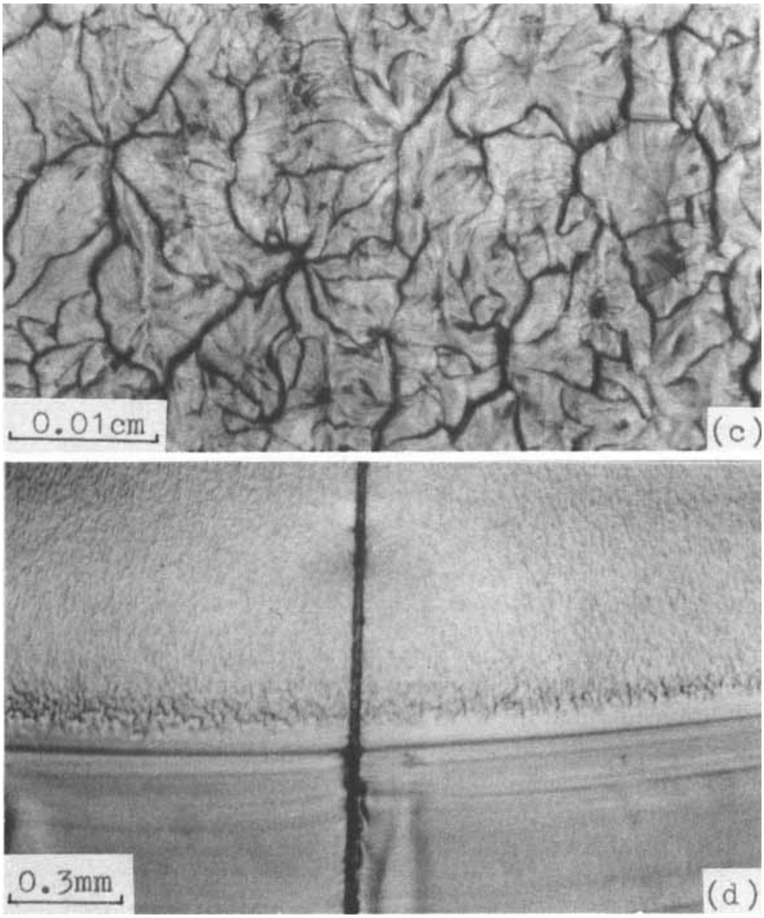


FIGURE 7 The morpholgies of different fracture surfaces of PMMA treated with ethanol at a temperature of 100°C for (a) shear; (b) tensile; (c) tear; and (d) cleavage tests.

The typical morphology of the tensile fracture surface is illustrated in Fig. 7(b) where the PMMA was treated with ethanol at 100°C. Many long and narrow hackles appear in Fig. 7(b). Their size increases with temperature. The tear strengths of PMMA treated with ethanol at different temperatures are listed in Table III. The tear strength increases with the temperature. The common feature of the tear fracture surface is many equiaxed hackles, the sizes of which increase with temperature. Figure 7(c) shows an example of the morphology of the tear fracture surface of PMMA treated with ethanol at 100°C.

The effect of temperature on the morphology of the cleavage fracture surface of PMMA is similar to that of the solubility parameter. The typical morphology of the cleavage fracture surface is illustrated in Fig. 7(d) where the PMMA is treated with ethanol at 100°C. The angles,θ, of the cleavage curves at interface of PMMA treated



See Figure 7 Caption on page 234.

TABLE III
Effect of temperature

The shear fracture stress τ, tensile fracture stress σ_{tn}, tear fracture stress σ_{tr} and the angle θ of cleavage curve at the interface of PMMA treated with ethanol at different temperatures.

T (°C)	80	85	90	95	100
τ (MPa)	8.9	11.7	13.6	16.0	21.4
σ_{tn} (MPa)	3.7	4.2	4.9	5.5	6.4
σ_{tr} (MPa)	3.0	4.1	4.2	5.4	6.1
θ	16°	13°	12°	4°	0°

with ethanol at different temperatures are tabulated in Table III. It is found that the angle decreases with increasing temperature. Comparing the morphology of the cleavage fracture surface and the mechanical strengths, we find that the smaller angles correspond to higher mechanical strengths. The sizes of the solvent affected zones of PMMA treated with ethanol at 80°C, 85°C, 90°C, 95°C, and 100°C are 105 μm, 55 μm, 52 μm, 15 μm and 12 μm, respectively. The size of the solvent affected zone, therefore, increases with decreasing temperature. This implies that the smallest solvent affected zone corresponds to the highest mechanical strength. Since the extent of volatility increases with the elevated temperature for the same boiling point, the PMMA dissolved in the ethanol to form a tacky layer increases with decreasing temperature. The amounts of residual ethanol in the PMMA treated with the ethanol at 80°C, 85°C, 90°C, 95°C and 100°C are 9 wt%, 7 wt%, 5 wt%, 3 wt% and <1 wt%, respectively. The residual ethanol in PMMA decreases with increasing effective glass transition point of PMMA so that the residual ethanol decreases with increasing mechanical strengths as listed in Table III. We infer that the solvent-welding process at higher temperatures corresponds to better quality of adhesion.

3.3 Cosolvent Effect

Based on the study of Cowie *et al.*,[21] mixtures of water and alcohols containing 0.4 volume fraction water have been found to be good solvents for PMMA at room temperature. We infer that the concentration of alcohol would affect the quality of adhesion. Thus, concentrations of ethanol varying from 80% to 100% by volume were studied. The solvent welding of PMMA took place at 90°C. Note that no adhesion was observed for concentrations of ethanol below 80% because so much water appeared on the wetted surface as to resist the bonding of the adherends.

The shear strengths of PMMA treated with different concentrations of ethanol at 90°C are listed in Table IV. The shear strength of PMMA increases with increasing concentration of ethanol up to 95% and then decreases. The shear fracture surface for 80% and 85% ethanol looks like a forest and the size of tree for the former is smaller than that for the latter. The typical morphology of the shear fracture surface of PMMA is shown in Fig. 8(a), where the concentration of ethanol is 85%. The shear fracture surfaces for 90%, 95%, and 100% ethanol looks like many tongues, the size of which is largest for 95% ethanol. The morphology of the shear fracture surface of PMMA treated with 95% ethanol is illustrated in Fig. 8(b) where the arrow indicates the shear direction. Comparing the shear strength and morphology of the shear fracture surface, we find that the largest tongue corresponds to the highest shear strength. The tensile strengths of PMMA treated with different concentration of ethanol of 90°C are tabulated in Table IV. The maximum tensile fracture stress is located at ethanol concentrations of 90% and 95%. The common feature of the tensile fracture surface of PMMA treated with different concentrations of ethanol at 90°C is narrow and long hackles, the sizes of which increase with the concentration of ethanol until 90–95% and then decrease. The typical morphology of a tensile fracture surface is illustrated in Fig. 8(c) where the concentration of ethanol is 95%. The tear strengths of PMMA treated with different

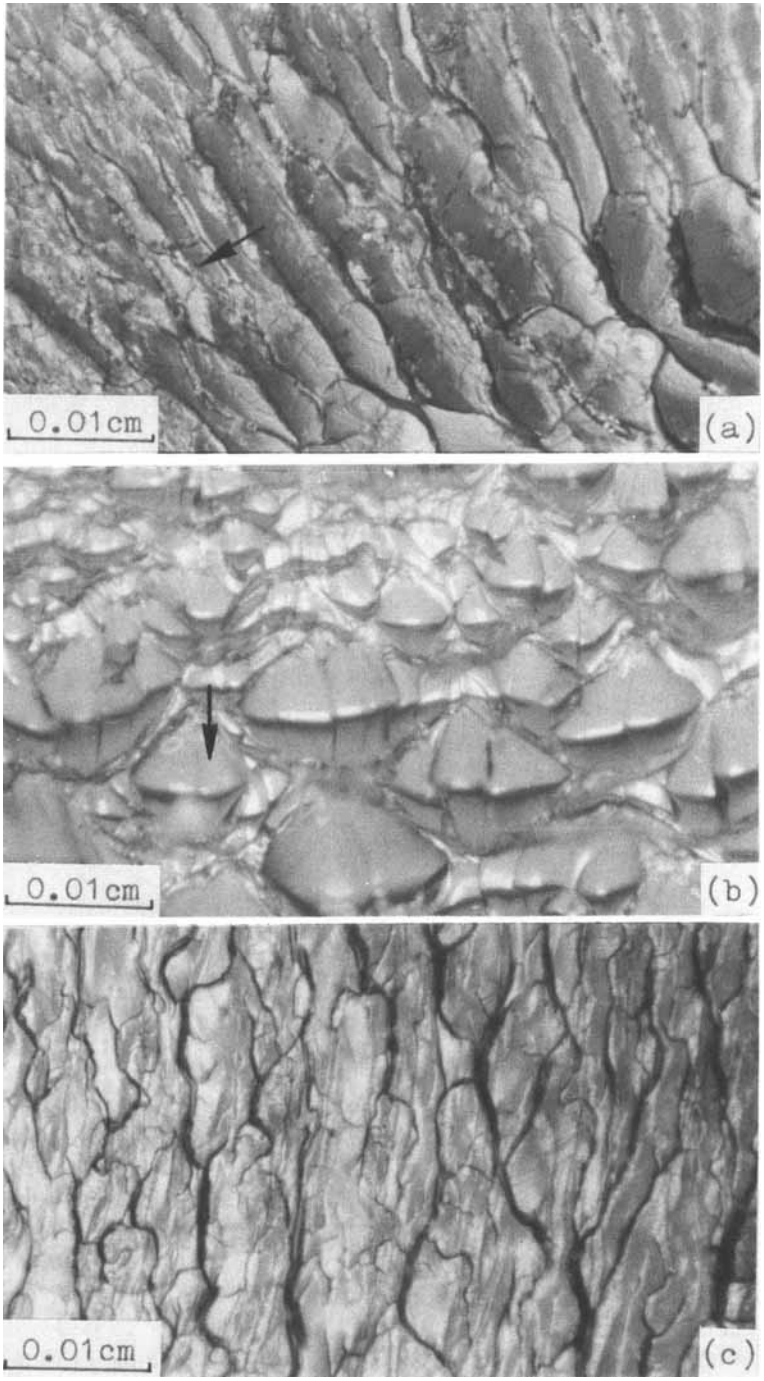
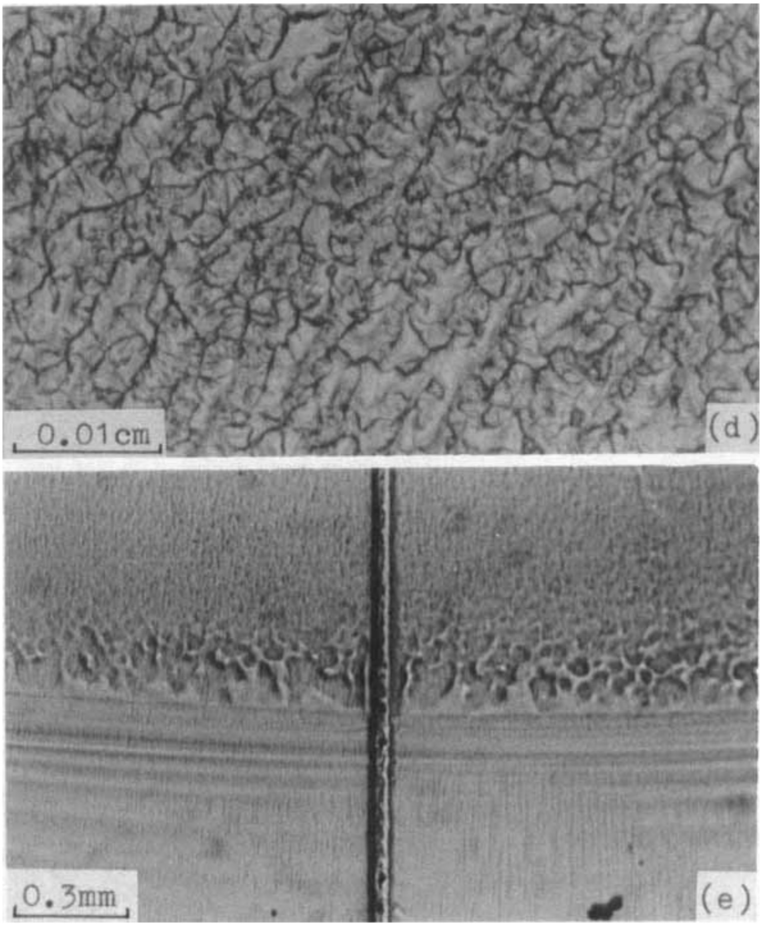


FIGURE 8 (a) The morphologies of shear fracture surface of PMMA treated with the concentration of ethanol 85% at 90°C and the morphologies of different fracture surfaces of PMMA treated with the concentration of ethanol 95% for (b) shear; (c) tensile; (d) tear; and (e) cleavage tests.



See Figure 8 Caption on page 237.

concentrations of ethanol at 90°C are also listed in Table IV. The tear strength of PMMA increases with the increasing concentration of ethanol until 95% and then decreases. The common feature of the tear fracture surface of PMMA treated with different concentrations of ethanol at 90°C is equiaxed hackles, the size of which increases with the increasing concentration of ethanol until 95% and then decreases. Figure 8(d) shows the typical morphology of a tear fracture surface of PMMA where the concentration of ethanol is 95%.

The angles,θ, of the cleavage curves at the interface of PMMA treated with different concentrations of ethanol at 90°C are tabulated in Table IV. It is found that the angle decreases with increasing concentration of ethanol until 95% and then increases. Comparing the angle of the cleavage curves and the mechanical strengths, as listed in Table IV, we find that the smaller angles correspond to the large mechanical strengths of PMMA. The sizes of the solvent affected zones of

TABLE IV
Effect of cosolvent

The shear fracture stress τ, tensile fracture stress σ_{tn}, tear fracture stress σ_{tr} and the angle θ of cleavage curve at the interface of PMMA treated with different concentrations of ethanol at 90°C.

Concentration (vol%)	80	85	90	95	100
τ (MPa)	8.7	10.2	14.3	15.1	13.6
σ_{tn} (MPa)	0.7	2.4	5.7	5.7	4.9
σ_{tr} (Mpa)	3.7	4.2	5.6	6.0	4.2
θ	22°	15°	6°	0°	12°

PMMA treated with ethanol concentration of 80%, 85%, 90%, 95%, and 100%, are 52 μm, 50 μm, 30 μm, 27 μm and 52 μm, respectively. The size of the solvent affected zone decreases with increasing concentration of ethanol until 95% and then increases. Because the solvent-welding temperature, 90°C, is higher than the boiling point of ethanol and lower than the boiling point of water, the ethanol in PMMA can easily evaporate but water cannot. Thus the amounts of residual cosolvent (including water and ethanol) in PMMA treated with ethanol of concentration 80%, 85%, 90%, 95%, and 100% by volume are 14.9 wt%, 8.9 wt%, 6.8 wt%, 6.6 wt%, and 5.0 wt%, respectively. The residual cosolvent decreases with the increasing concentration of ethanol. The mechanical strength of PMMA treated with different concentrations of ethanol does not correspond either to maximum or to minimum residual solvent. Comparing the mechanical strengths as listed in Table IV with morphologies of fracture surfaces, we find that the maximum mechanical strength is located at the 95% concentration of ethanol which is a good solvent as explained by Cowie *et al.*[21] using the hydrogen-bonding model.

4. SUMMARY AND CONCLUSIONS

The microstructure of solvent-welding of PMMA and related mechanical strengths have been investigated. Morphologies of cleavage, tear, tensile and shear fracture surfaces have been examined. Three solvents, methanol, ethanol and DMF were employed. The effects of solubility parameter, elevated temperature and cosolvent on mechanical strength were also studied. Some important results are summarized as follows.

1. The features of tear, tensile and shear fracture surfaces are equiaxed hackles, long and narrow hackles, and tongues, respectively. The size of their features increases with the mechanical strength. Also, the angle of the cleavage curve at the interface decreases with increasing mechanical strength.
2. PMMA solvent-welded at higher temperature recovers a larger fraction of its original strength.
3. Water, as a cosolvent with ethanol, changes the extent of adhesion of PMMA.

PMMA treated with 95% by volume of ethanol corresponds to the best quality of adhesion.

4. The size of the solvent affected zone decreases with increasing mechanical strength.

5. As expected from the solubility parameter, PMMA solvent-welded with a good solvent has a large amount of residual solvent. For the same solvent, solvent-welding at high temperature leads to a small amount of residual solvent. For mixtures of water with ethanol, PMMA solvent-welded with ethanol in a high concentration has a small amount of residual cosolvent.

Acknowledgement

The authors would like to thank Dr. C. T. Hu for valuable comments. This work was supported by the National Science Council, Taiwan, Republic of China.

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