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AN EXPERIMENTAL STUDY ON CRACK HEALING OF VARIOUS GLASSY POLYMERS

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The validity of a simple tube model proposed by the senior author for crack healing recovery of various polymeric materials such as PMMA(Polymethyl-methacrylate) and Homalite-100(A Polyester) has been tested.

The experimental results of recovery in terms of healing fracture toughness were compared with the theoretically predicted trends of recovered fracture toughness. The agreements between these results were found to be reasonable within the limitation of experimental method.

After healing the PMMA CT (compact tension) specimens with various solvents having different solubility parameters(δ), the healing recovery (R) was measured by both the method of maximum load and the method of caustics. The relationship between R and solubility differences ($\Delta\delta$) between a solven and a glass polymer was found to be $R \propto (\Delta\delta)^{-1/4}$.

Key Words : Simple Tube Model, Crack Healing, Polymeric Materials, Healing Recovery, Healing Fracture Toughness, Maximum Load Methods, Method of Caustics, Solvent, Solubility Parameters.

1. INTRODUCTION

The investigation on various composites utilizing various polymeric materials as matrices has attracted the attention of many researchers and manufacturers because of its high specific strength and the concern with energy saving. The fracture researches for composite materials among other studies have been actively done with the help of both linear and elastic-plastic fracture mechanics theories. However, few researchers have considered the defect healing phenomenon which is opposed to fracturing in the glassy polymeric materials as an important issue among many serious problems.

In the earlier published reports (Outwater, 1969 ; Wool, 1981 ; De Gennes, 1971 ; Kramer and Edward, 1967) relating to the crack healing process, a simple model such as the reptation model under some reasonable assumptions was utilized to investigate the healing toughness and some other healing mechanical and physical properties. Futhermore, a theoretical model for the strength of weldliness was proposed and its good correlation with experimental results was published(Kim and Suh, 1986). It seems, however, that the general acceptance of the proposed modeling analysis is yet open to question.

Recently, Lee has proposed a new simple and intuitive theoretical model for the prediction of healed fracture toughness of cracked or defective polymeric components(Lee, 1986). In this newly proposed modeling analysis, the effects of chain scission and the behavior of chain molecules under healing conditions on the healing process were incorporated. Furthermore, the wetting and interdiffusion processes were assumed to be equivalent through the whole domain of the fractured surfaces and the chain length in the theoretical modeling analysis to have the relationship between the healing recovery (R) and the healing time (t) as $R \propto t^{1/2}$ rather than $R \propto t^{1/4}$ obtained by other investigators (Jud, 1981; Wool, 1981). The detailed theoretical modeling analysis can be found in Lee(1986).

The main objective of this paper is to test the simple healing model proposed by Lee through comparision between the predicted healing trend with the aid of proposed model and experimentally determined results.

2. EXPERIMENTS

Materials used for this experimental work were a typical uncross-linked thermoplastics, PMMA (plymethylmethacrylate) and a polyester cross-linked thermoset, Homalite-100. Figure 1 shows the typical geometry of the specimen. It was found that the Homalite-100 and PMMA contracted during the healing period in the high temperature surrounding. Virgin materials were heat-annealed at the expected healing temperature for about two hours to allow all possible contractions to arise. The crack tip geometry was made by a single through saw blade technique described below. The blunt notch was first made by using a saw cut. The V-shaped groove at this blunt notch was introduced by using an appropriate size of chisel. A specially ground saw blade was finally used to make a very sharp crack tip configuration.

A universal testing machine (Tensilon) was used to apply tensile load in the compact-tension (CT) specimens. Srawley (Srawley and Gross, 1972) and Wessel (Wessel, 1968) estimated the stress intensity factors for this type of CT specimens by using respectively the boundary collocation method and an experimental technique. The fracture toughness of both virgin and healing materials with the specimen geometry of the ASTM standard specification were determined by utilizing the well-Known Wessel's result such as

$$K_I = p\{A_1(a/W)^{1/2+}\ A_2(a/w)^{3/2+}A_3(a/w)^{5/2+}A_4(a/W)^{7/2+}\ A_5(a/W)^{9/2+}\cdots\cdots\}/BW^{1/2} \quad (1)$$

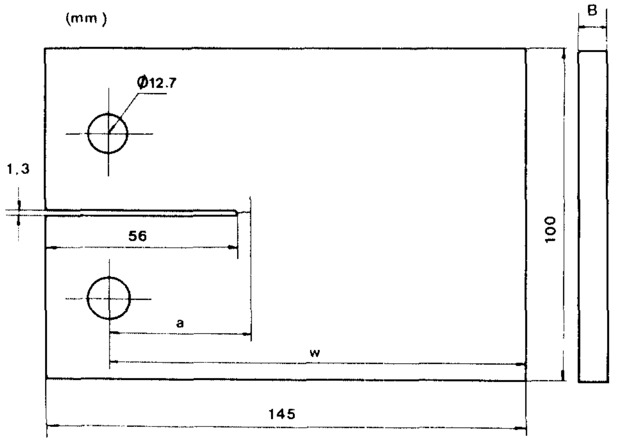

Fig. 1 Dimension of compact-tension specimen

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P is the applied load

W is the width of a specimen

B is the thickness of a specimen

a is the crack length

$A_1=29.6$, $A_2=-185.5$, $A_4=-1017.0$, $A_5=638.9$

For the different fracture specimen with various crack and ligament lengths, Srawley's numerical results were utilized to estimate the stress intensity factors for both virgin materials and healing specimens. The values of fracture toughness for the virgin materials determined by using Eq. (1) were compared to those obtained by the method of caustics. It was noticed, however, that keen attention should be paid to the method of caustics. It was necessary to predetermine the reference distance, which is the interval between the specimen mid-plane and the reference plane (image plane), that would provide the appropriate initial curve with respect to the thickness of the specimens. Figure 2 shows the appropriate ratios of the radius of initial curves, which is located at the vicinity of the crack tip and corresponds to the caustics on the image plane, to the thickness of specimens for Homalite-100 and PMMA. A preliminary result should be referred to reference (Lee, et al., 1986) and the details will be published elsewhere.

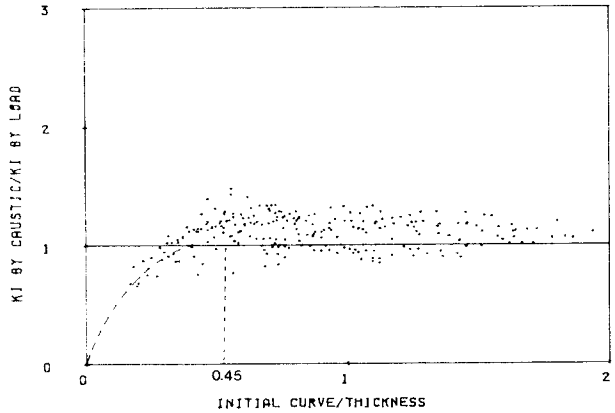


Fig. 2(a) r_0/B vs. K_I by caustics/K_I by load (PMMA)
(r_0=initial curve, B=thickness)

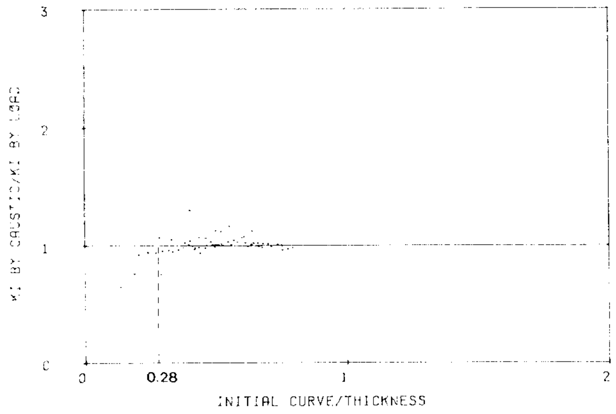


Fig. 2(b) r_0/B vs. K_I by caustics/K_I by load (Homalite-100)
(r_0=initial curve, B=thickness)

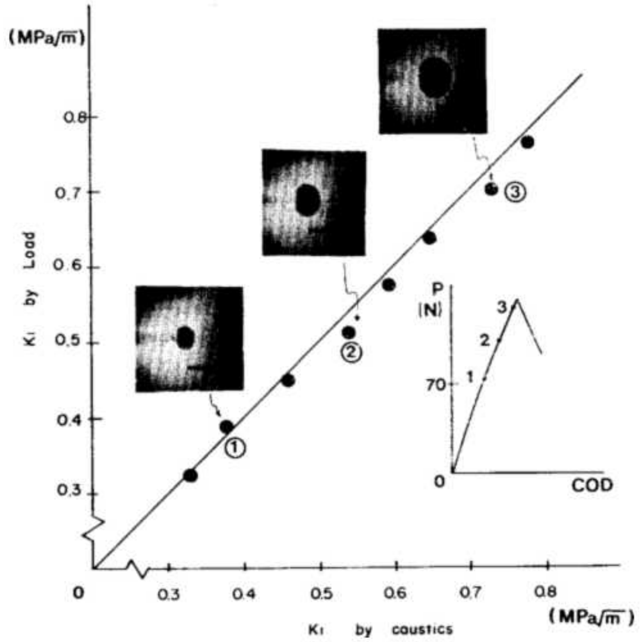


Fig. 3 K_I by load vs. K_I by caustics

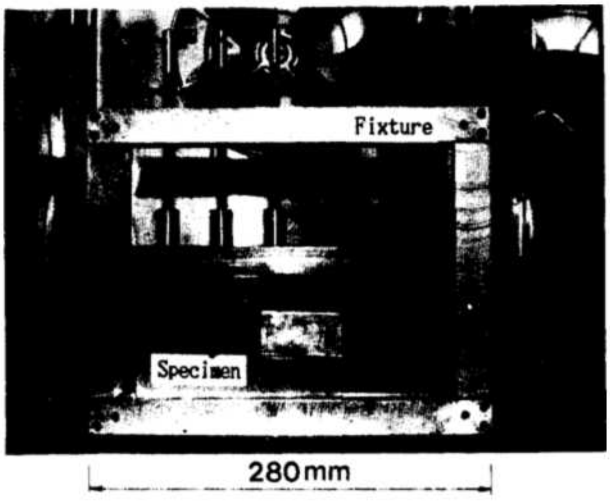


Fig. 4 fixture for healing experiments in the furnace

The stress intensity factors determined from the two distinct procedures described above were compared with each other as shown in Fig. 3. It was found that the differences were within 5%. A special crack healing frame was constructed to provide uniform constant compressive loads along the healing surface as shown in Fig. 4. This contact pressure which was needed to help instantaneous wetting along the fractured surfaces was to be adjustable through the push rods mounted on springs. The fractured specimen was sandwiched between two flat smooth aluminum plates to prevent possible warping and buckling during the healing process. The wetting occurred between the fractured surfaces was observed to be the surface mode and it spread out in the radial direction with constant velocity. The wetting covered the whole interface in a short time, say t=30sec. It was also noted that the specimen temperature when the fractured surfaces were made contact should be lower than the glass transition temperature, Tg, of the specimen material. If we raise the specimen temperature above Tg in the first place and then place the fractured surfaces in contact, the surface molecule rearrangement would occur before

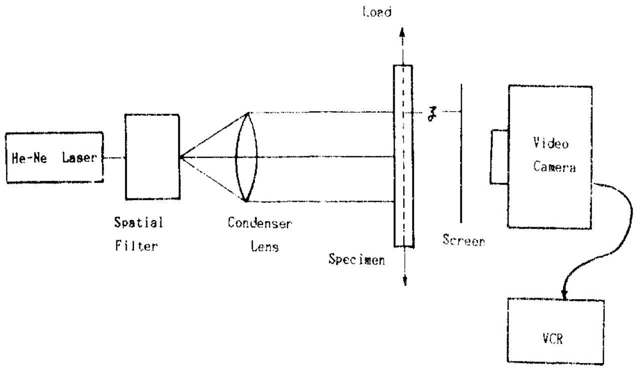


Fig. 5(a) Schematic diagram of caustics measurement

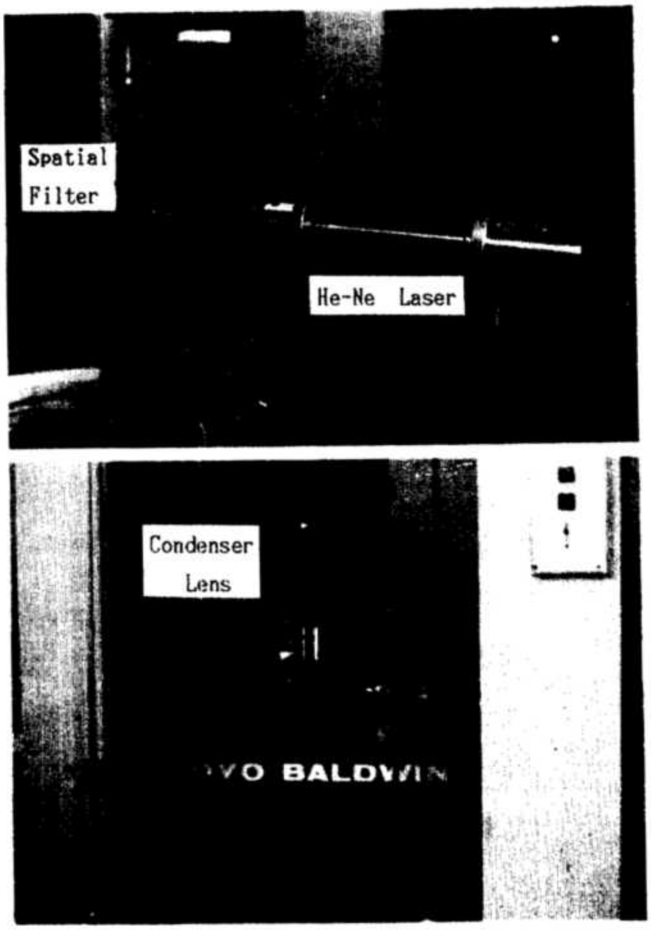


Fig. 5(b) Photographs of caustics measurement setups

wetting and healing process so that the crack healing could be inhibited. Therefor, it is suggested that the fractured surfaces should be placed in contact first and then temperature of the specimen be raised above Tg of the material. The schematic diagram and experimental setup for caustices measurement are shown in Fig. 5.

Investigating the solvent healing phenomena in terms of stress intensity factor in PMMA CT specimens, various solvents which have different solubility parameters from that of PMMA as shown in Table 1 were used as healing agents.

Table 1 Solubility parameter, δ, of various solvents

Solvent	Ethyl -Acetate	Toluene	Acetone	Pyridine	Ethanol	Meth -anol
δ	9.1	8.9	10.0	10.9	12.7	14.5

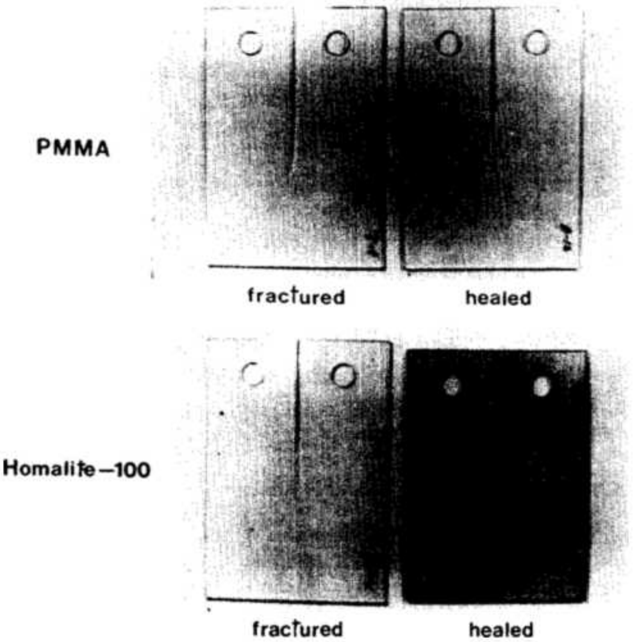


Fig. 6 Configuration of fractured and healed specimen (thickness : 3.8mm)

The experimental procedures were the same as the above described earlier in this section. However, the deformed crack tip configuration which may arise during the healing period was noticed. The accurate healing recovery (R) in terms of fracture toughness cannot be determined by using the healed specimen with the blunted crack tip.

New crack tips about 5mm ahead of the blunted crack tips were produced in the healed surface with the application of carefully induced impact loads using a sharp-edge blade. The virgin and healed specimens are shown in Fig. 6.

3. RESULT AND DISCUSSION

Figure 7 shows a typical response between the applied load

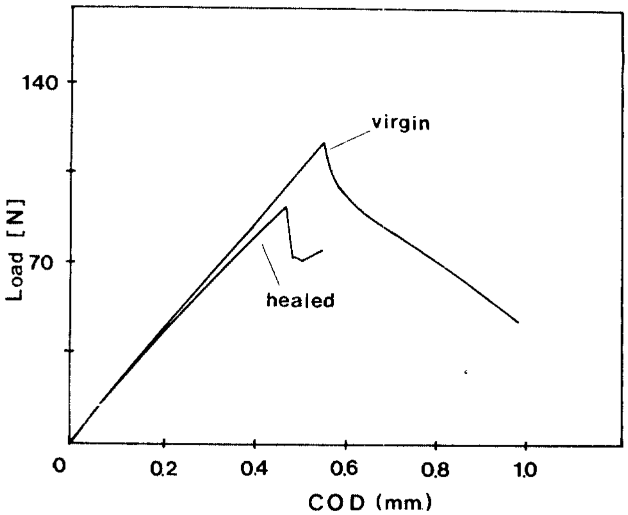


Fig. 7(a) Load vs. crack opening displacement(COD) (PMMA)

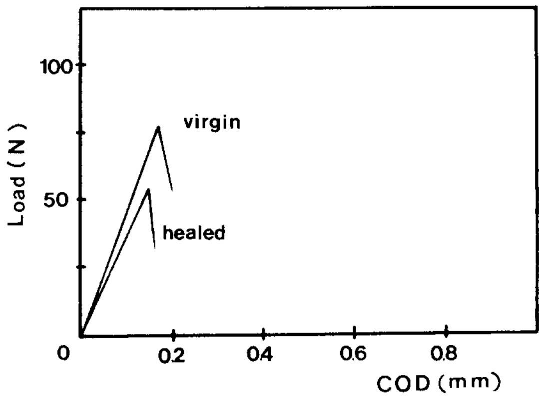
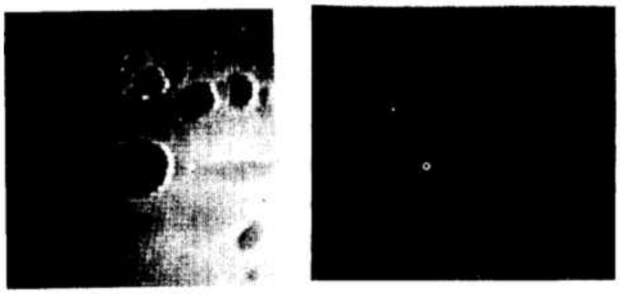
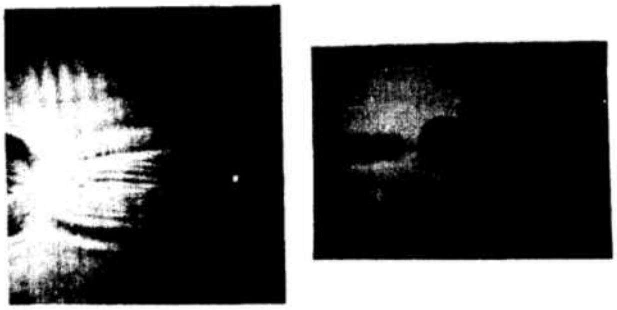


Fig. 7(b) Load vs. crack opening displacement(COD) (Homalite-100)



PMMA (T=130℃, t=30 min)   Solvent healing (Acetone)



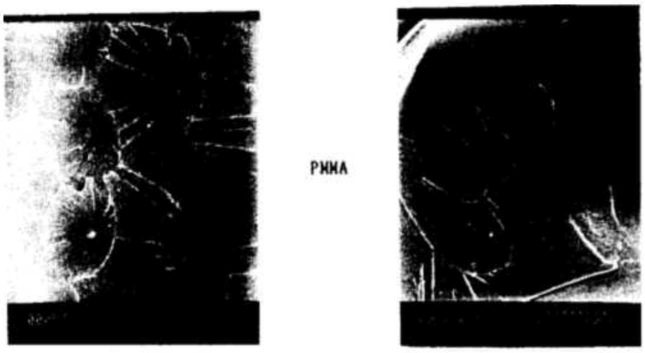
Homalite-100 (T=140℃, t=10 hr.)   Solvent healing (toluene)

Fig. 8 Typical caustics under various healing conditions

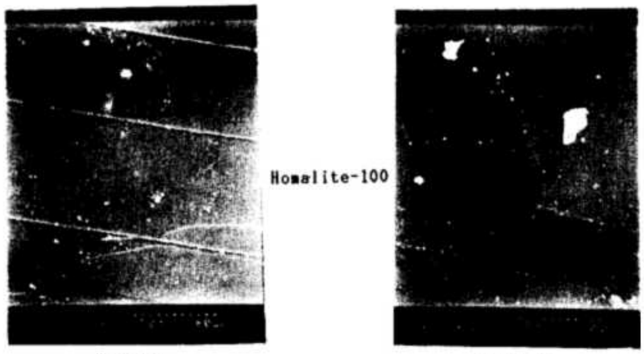
and the crack opening displacement (COD) in CT specimens of PMMA and Homalite-100. It is notable that the compliance of healed specimens increases slightly as is expected and the crack initiatioin points are easily distinguishable asin the case of virgin specimens.

Typical shapes of caustics in various healed specimens are shown in Fig. 8. The shape of caustics shows a little irregularity as was expected because of the deformation which took place along the healed line. However, the irregularity was not so pronounced as shown in Figure 8 so as to make measurement of stress intensity factors impossible.

Fracture surface morphology of the virgin and the healed specimen was investigated by making comparison of micrographs of fracture surfaces using the scanning electron microscope. Figure 9 shows microfractographs of the fracture surface in the virgin and healed CT specimens. The changes in the fracture surface morphology were found to be not noticeable within this experimental limit. It may be due to the unchanged molecular structure even though the molecular chains moved across the healing surface above the transition temperature.



virgin   (×500)   healed (120℃, 50 min.)



virgin   healed (140℃, 14 hr.)

Fig. 9 Microfractographs of the fractured surface of compact tension specimens

Healing recovery (R) in terms of ratio of healing K_I to virgin K_I was plotted against the healing time for various healing temperature such as 120℃, 130℃, and 140℃ in PMMA specimen as shown in Fig. 10. The similar plot for Homalite-100 specimens healed under various temperatures like 140℃ and 160℃ is shown in Fig. 11. Typical caustics and load (P)-loading point displacement corresponding to some experimental data are also shown in both Fig. 10 and Fig. 11.

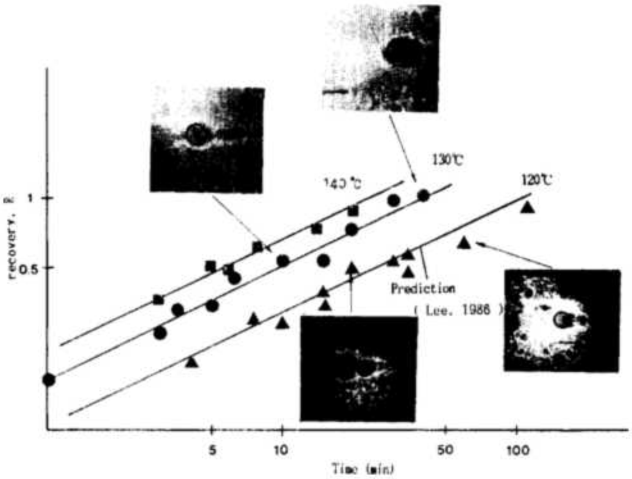


Fig. 10 Healing recovery vs. healing time for PMMA

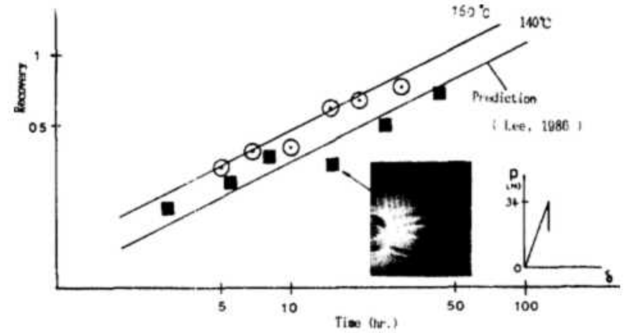


Fig. 11 Healing recovery vs. healing time for homalite-100

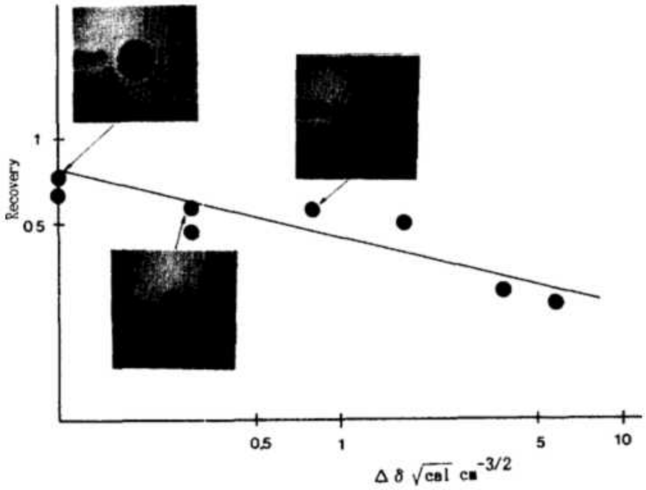


Fig. 12 Healing recovery vs. solubility parameter for PMMA

It is notable that the experimental results shows good agreement with the theoretically predicted healing trends as appeared in both Fig. 10 and Fig. 11. It is also interesting to note that the healing trends are identical for either thermosets or thermoplastics as far as the relation between recovery R, and healing time t, is concerned although the basic structure of the molecular chain entanglement are different.

The higher the healing temperature was the shorter the healings time due to the more active movement of the molcular chain at higher temperatures. The precise relationship between the healing temperature and healing time in terms of healing recovery, say fracture toughness, is yet to be answered.

Figure 12 shows the relationship of the recovery, r, to the solubility differences, $\Delta\delta$, between PMMA and various solvents. The shape of caustics and load-loading point displacement are also shown in this figure for some experimental data and the empirical relation of $R \propto (\Delta\delta)^{-1/4}$ could be determined without doing further physical investigations.

4. CONCLUSIONS

During the course of this investigation the experimental limitation of the method of caustics in terms of the ratios of the initial curve to the specimen thickness were determined for PMMA and Homalite-100. The appropriate ratios of the radius of initial curves to the specimen thickness were found to be 0.28 and 0.45 for PMMA and Homalite-100 compact-tension specimens respectively.

The simple tube model for crack healing in the polymeric materials proposed by Lee was examined by comparing the theoretically predicted healing trends with the experimentally determined healing recover, R. The agreement between the predicted values and the experimental results were found to be good within the limitation of experiments. The relationship between the recovery, R, and the healing time, t, was turned out to be $R \propto t^{1/2}$ rather than $R \propto t^{1/4}$ which was proposed by some other investigators.

For the healing of PMMA specimens through the use of solvents as healing agents, an empirical relation between recovery, R, and the solubility differences ($\Delta\delta$) such as $R \propto (\Delta\delta)^{-1/4}$ was determined.

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