Indentation Responses of Viscoelastic Materials

I. M. LOW, G. PAGLIA, C. SHI

Materials Research Group, Department of Applied Physics, Curtin University of Technology, GPO Box U1987, WA, Australia 6845

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ABSTRACT: Vickers indentation is arguably one of the most widely used techniques for characterizing the mechanical properties of materials because it is easy, inexpensive, and nondestructive. However, its popularity has so far been limited to ceramics and metals, and very little literature information is available on the Vickers indentation properties of high or rigid polymers. In this article, the Vickers indentation responses of an epoxy and acrylic polymer have been studied. The hardness of these materials is found to be time-dependent as a result of viscoelastic flow and relaxation processes. Unlike ductile metals, the microhardness is not dependent on the indentation load. The elastic recovery in the Vickers impression takes place only along the side faces but not along the diagonals. Thus, the use of Vickers indentation as a convenient tool for evaluating the hardness and viscoelastic responses of rigid polymers is justified. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 2349–2352, 1998

Key words: Vickers indentation; viscoelastic; hardness; epoxy; acrylic; elastic recovery

INTRODUCTION

Vickers indentation has been a standard method for material characterization for a long time as it provides an easy, inexpensive, nondestructive, and objective method of evaluating basic properties from small volumes of materials. Besides hardness and toughness, it has also recently been used to characterize residual stresses,[1,2] yielding stress,[3] Young's modulus,[4] thermal shock resistance,[5,6] and subsurface damage.[7]

Hitherto, Vickers indentation has been primarily used to characterize properties of metals and ceramics. Very little literature information is available on the Vickers indentation properties of high or rigid polymers. The reluctance of researchers in using Vickers indentation as a tool for studying the properties of these materials may be due to the problem of pronounced elastic recovery or time-dependent behavior, which is absent in most metals and ceramics. Also, the conventional hardness testing methods for plastics Rockwell (ASTM D785) and Shore (ASTM D676) are limited by their load ranges, indenter shapes, and hardness ranges.

During the loading of polymers, a small amount of elastic deformation occurs and is followed by viscoelastic flow. When unloading, spontaneous elastic recovery takes place and is followed by a time-dependent recovery of the deformation. The recovery of viscoelastic polymers depends on the material itself, temperature, and the state of internal stresses.[8] The elastic recovery usually results in a pyramidal indent having sides that are concaved inwards or star-shaped. However, the elastic recovery in the direction of the diagonals is generally negligible or very small. Therefore, the measurement of diagonal lengths should give valid hardness values for viscoelastic rigid polymers.

In this article, we present the responses of 2 rigid polymers under Vickers indentation. The

Correspondence to: I. M. Low

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variations of hardness as a function of load and time are investigated. Results show that the hardness of these materials is time-dependent but load-independent. The effect of viscoelastic flow on the hardness response is discussed.

EXPERIMENTAL PROCEDURE

Rigid polymers used for the study were based on an epoxy resin and an acrylic resin. The epoxy resin employed was a commercial product KIT 36 supplied by Fibreglass and Resin Sales, Perth, Western Australia. Details of preparation of the epoxy polymer have been described elsewhere.[9,10] In brief, a sheet of 100 mm × 50 mm × 6 mm was prepared by casting a mixture of epoxy (2 parts) and hardener (1 part) in a greased metal mold, which was then cured at room temperature for at least 24 h. The acrylic used was a moulding Transoptic powder supplied by Buehler, Lake Bluff, USA. Acrylic discs of 25 mm diameter and 10 mm height were compression-molded in a Struers Prestopress at 2.5 tons pressure. To facilitate the process of sample polishing for Vickers indentation measurements, short bars (10 mm × 10 mm × 6 mm) of epoxy were cut and mounted in acrylic resin. Both epoxy and acrylic disc samples were then polished to 1 μm surface finish using a Struers Pedemat auto-polisher. The Vickers hardness measurements were performed using a Zwick microhardness tester to evaluate the resistance to deformation or hardness of the polymer. In general, hardness values can be related to other materials properties, such as strength and elastic modulus. The lengths of the diagonals (2a) were used to calculate the hardness, determined here as

$$H_v = P/2a^2 \tag{1}$$

where P is the load used. The variations of hardness as a function of load were performed over the range 0–100N at an indentation time of 20 s. The effect of indentation time (0–600 s) on the variations of hardness at 50N load was performed to ascertain the viscoelastic nature of these materials. The hardness values were measured immediately after indentation and also after 72 h to detect if elastic recovery had occurred. Photomicrographs of the morphology of indents and their elastic recovery were taken using a Nikon optical microscope.

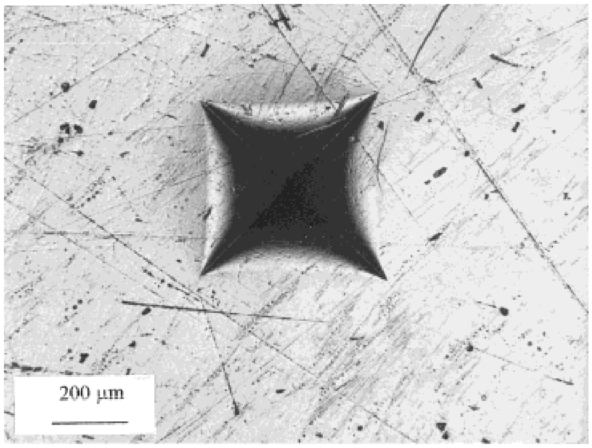
RESULTS AND DISCUSSION

Indentation Responses

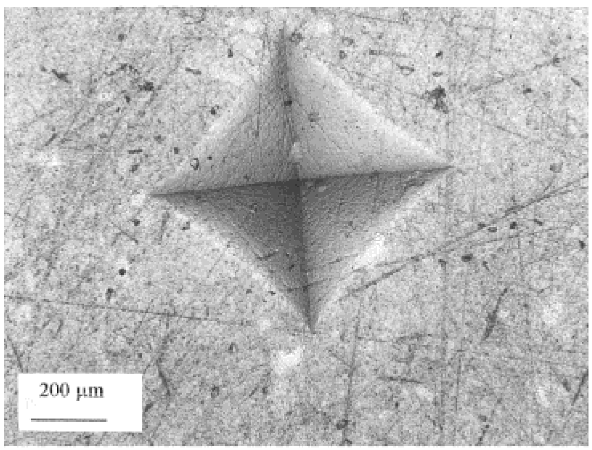
The size of Vickers indents increased with an increase in the load. However, no indentation cracks were observed in both polymers, even at the maximum load (100N) used. This is interesting because these materials have a very low fracture toughness (K_{ic}) of typically $\sim$ 1.0 MPa m$^{1/2}$, which is comparable to that of silica glass. The latter is well-known to be very brittle and forms indentation cracks readily.[11] The absence of indentation cracks in both epoxy and acrylic samples can be ascribed to (1) low hardness (H_v) and (2) high critical load (P_c) to initiate cracks, where P_c is proportional to (K_{ic}^4/H_v^3).[11–13] Hence, polymeric materials and metals hardly ever show radial cracks, except at very high loads. For instance, it would need a critical load of 800 kN to form indentation cracks in a mild steel.[11] Similarly, the critical load for epoxy and acrylic can be calculated to be approximately 800N, which is well beyond the loading capacity of the Zwick tester.

The acrylic sample displayed a star-shaped Vickers impression [Fig. 1(a)], whereas the epoxy sample displayed a pyramidal impression [Fig. 1(b)], which is commonly observed in ceramics and metals. The former appears to exhibit a much greater extent of elastic recovery along the faces but not along the diagonals of the impression. In contrast, it appears that near permanent plastic deformation had occurred below the contact during indentation of epoxy.

Figure 2 shows the variations of hardness as a function of load for epoxy and acrylic with the latter being the much harder polymer. Clearly, the hardness of these polymers is virtually independent of load, a characteristic similar to that of brittle materials, such as silica glass and alumina.[14] In contrast, materials with a ductile behavior such as metals normally display microhardness, which decreases with load. This load-dependent hardness characteristic has also recently been observed for metal-like Ti_3SiC_2[15,16] and can be attributed to the effect of large grain size. At small loads, the contact diagonal (2a) of Vickers impression is less than the grain size, and the hardness measures properties of single grains; when 2a becomes much larger than the grain size at high loads, the hardness measures polycrystalline properties, with more grains oriented for deformation by slip. It follows that the absence of



(a)



(b)

Figure 1 Morphology of Vickers indent in (a) acrylic and (b) epoxy.

load dependency of hardness in amorphous epoxy and acrylic may be due to the lack of a grain size effect.

Viscoelastic Responses

The viscoelastic nature of both epoxy and acrylic during indentation is clearly revealed in Figure 3, which shows the variations of hardness as a function of indentation time. The hardness of these materials decreased with an increase in time. This suggests that during loading, the size of indent increases with time by virtue of viscoelastic flow and relaxation processes. This time-dependent behavior of hardness was more pronounced in acrylic than in epoxy, which may be attributed to the difference in their chemical structure, glass transition temperature (T_g), and hardness. The presence of crosslinking in epoxy may serve to reduce the extent of viscoelasticity.

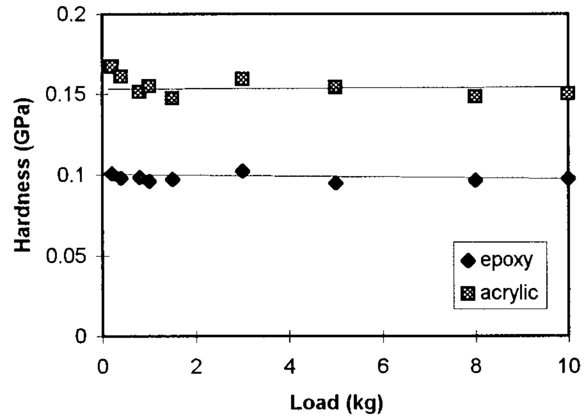


Figure 2 Variations of hardness as a function of indentation load.

Figure 4 shows the hardness response of both polymers immediately and 72 h after the indentation. The hardness values have remained almost unchanged with time, which indicates the near absence of elastic recovery along the diagonals of the impression. This suggests that only the material along the diagonals had undergone permanent plastic deformation because of intense stress concentration. Most, if not all, of the recovery took place along the sides, causing the faces of the impression to curve inwards and form a star-shape [Fig. 1(a)]. A partial plastic deformation had taken place in other regions of the impression, which allowed some elastic recovery to take place. The absence of elastic recovery along the diagonals thus justifies the use of Vickers indentation as a convenient tool for evaluating the hardness, viscoelastic, and other responses of rigid polymers.

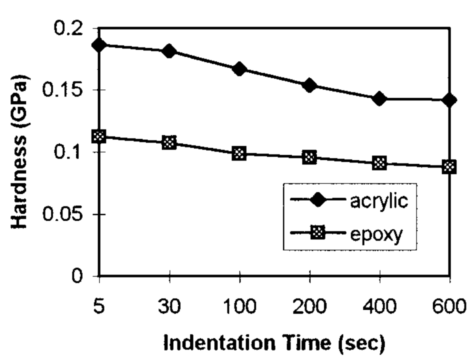
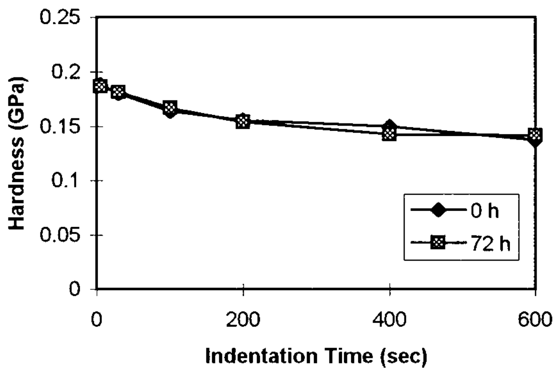
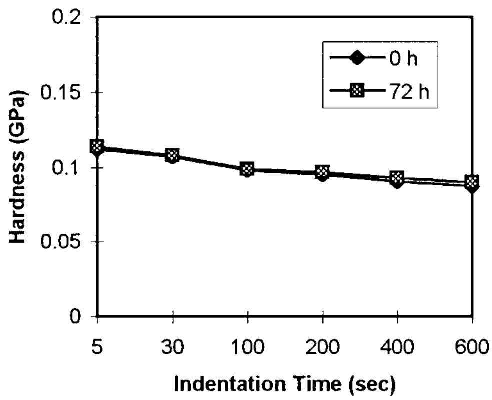


Figure 3 Variations of hardness as a function of indentation time.



(a)



(b)

Figure 4 Variations of hardness as a function of indentation time for (a) acrylic and (b) epoxy immediately and 72 h after the test.

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REFERENCES

1. A. Pajares, F. Guiberteau, R. W. Steinbrech, and A. Dominguezrodriguez, *Acta Metall. Mater.,* **43,** 3649 (1995).
2. V. M. Radhakrishnan, *Trans. Ind. Inst. Metals,* **46,** 241 (1993).
3. K. Zeng, E. Soderlund, A. E. Giannakopoulos, and D. J. Rowcliffe, *Acta Mater.,* **44,** 1127 (1996).
4. D. Chicot, I. Hage, and J. Lesage, *Rev. Metall. Cahiers d'Inform. Techniques,* **92,** 635 (1995).
5. F. Osterstock and B. Legendre, *J. Physique III,* **7,** 561 (1997).
6. F. Tancret and F. Osterstock, *Scripta Materialia,* **7,** 443 (1997).
7. I. M. Low, *J. Europ. Ceram. Soc.,* **18,** 709 (1998).
8. K. Muller, in *The Science of Hardness Testing and Its Research Applications,* J. H. Westerbrook and H. Conrad, Eds., American Society for Metals, Metals Park, Ohio, 1973, pp. 291–299.
9. I. M. Low, P. Schmidt, and J. Lane, *J. Mater. Sci. Lett.,* **14,** 170 (1995).
10. I. M. Low, P. Schmidt, J. Lane, and M. McGrath, *J. Appl. Polym. Sci.,* **54,** 2191 (1994).
11. B. R. Lawn, *Fracture of Brittle Solids,* Cambridge University Press, Cambridge, UK, 1993.
12. B. R. Lawn, T. Jensen, and A. Arora, *J. Mater. Sci.,* **11,** 573 (1976).
13. B. R. Lawn and D. B. Marshall, *J. Am. Ceram. Soc.,* **62,** 347 (1979).
14. I. J. McColm, *Ceramic Hardness,* Plenum Press, New York, 1990.
15. T. El-Raghy, A. Zavaliangos, M. Barsoum, and S. R. Kalidindi, *J. Am. Ceram. Soc.,* **80,** 513 (1997).
16. I. M. Low, S. K. Lee, M. Barsoum, and B. R. Lawn, *J. Am. Ceram. Soc.,* **81,** 225 (1998).