EXPERIMENTAL STUDY OF MICROMECHANISMS OF BRITTLE-TO-DUCTILE TRANSITION IN *Si* SINGLE CRYSTALS

K.J. HSIA
University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA
A.S. ARGON
Massachusetts Institute of Technology, Cambridge, MA 02139, USA

ABSTRACT

The micromechanisms of brittle-to-ductile transition (BDT) in *Si* single crystals are investigated using a novel experimental technique recently developed by Brede et al. (1991) and Hsia et al. (1992). The crack arrest tests are performed by propagating a cleavage crack with a quasi-steady state velocity against a temperature gradient. Laser imaging technique is used to measure the crack velocity. The crack arrest temperature (BDT temperature) is determined as a function of crack velocity. The results indicate that high dislocation mobility and a high dislocation density are needed to arrest a running cleavage crack. For *Si* crystals, the rate-limiting mechanism is dislocation motion rather than dislocation nucleation.

KEYWORDS

Brittle-to-ductile transition, cleavage crack, dislocation nucleation and mobility.

INTRODUCTION

The phenomenon of brittle-to-ductile transition (BDT) in fracture has been studied by many researchers for decades. But the fundamental mechanisms during the BDT have been investigated only recently. Recent theoretical developments have indicated that the fundamental phenomenon of BDT is governed at the tips of propagating cleavage cracks by basic atomistic processes. Two distinct processes, brittle fracture by breaking bonds between atoms along cleavage plane and ductile fracture by generating dislocations in slip planes, compete against each other at propagating crack tips. While the cleavage fracture process is fairly well understood by means of an energetic consideration, dislocation generation and its effects on a propagating cleavage crack are not yet fully understood mechanistically.

Dislocation generation at crack tips involves two successive stages, *dislocation nucleation* at and *dislocation motion* away from the crack tip region. Two different groups of materials exist: the one in which dislocation mobility is relatively high compared to the barrier to dislocation nucleation, and the one in which dislocation nucleation is relatively easy but mobility is low. In the former group the rate-limiting phenomenon which determines the BDT is the competition between dislocation nucleation and cleavage crack propagation, whereas in the latter it is the competition between dislocation motion induced crack tip shielding or blunting and cleavage fracture. It is expected that the BDT behavior of these two groups of materials have different characteristics.

In a poineer attempt to model the competition between cleavage crack growth and dislocation emission from the crack tip, Rice and Thomson (1974) performed an activation analysis in which the total energy change due to the formation of a dislocation half loop at crack tip was calculated as a function of loop radius and dislocation core cut-off radius. In their analysis, a linear elastic crack tip stress field was used, and the materials were assumed to behave as a continuum up to the crack tip. Their model was very successful in distinguishing intrinsically brittle and intrinsically ductile materials, but the predicted critical activation energies for the intrinsically brittle materials to initiate plastic deformation were two orders of magnitude too high than that achieveable by thermal activation. Argon (1987) pointed out that the linear elastic solution was no longer applicable at the crack tip where a dislocation loop was formed. A Peierls (1940) type periodic resistance to the dislocation emission must then be used in a more accurate analysis. Chueng et al. (1991) made a full field analysis using the molecular dynamics approach, and successfully reduced the critical activation energy for α-iron by a factor of 15. But due to the limitation of the molecular dynamics approach and the unrealistic configuration, the predicted energy barrier for dislocation emission was still about one order of magnitude too high. The biggest uncertainty in these models is the dislocation core cut-off radius which is often chosen arbitrarily. Following the suggestion by Argon (1987), Rice (1992) recently developed a model which took into account the nonlinear interaction of atoms based on the Peierls concept. This model avoided the ambiguity of core cut-off radius, and identified the critical energy barrier to dislocation emission as the peak energy level a partially nucleated dislocation encountered when it reached the instability configuration.

The above models are applicable to the material systems in which dislocation nucleation is the rate-controlling mechanism for the BDT. Except for the model by Argon (1987), none of these models is capable of predicting the BDT temperature for different crack velocities or different strain rates. However, if the BDT is indeed dislocation nucleation controlled (i.e., dislocation motion is substantially easier than dislocation nucleation at temperatures near the BDT), the influence of strain rate on the BDT in these material systems may not be as profound as in dislocation mobility controlled material systems.

The BDT due to dislocation motion induced crack tip shielding and blunting has also been studied by a number of researchers. Ashby and Embury (1985) developed a model by studying the dislocation behavior adjacent to a crack tip, and discussed the effect of crack tip dislocation density on the BDT. Argon (1987) considered a steady state propagating crack which drags a plume of dislocations with it. By taking into account of the shielding effects, he obtained a relationship of the BDT temperature as a function of crack velocity. Hirsch et al. (1989), on the other hand, studied the interaction between the stress field of individual dislocations and that of a crack tip. For a stationary crack under different loading rates, their model was capable of predicting the BDT temperature in *Si* single crystals.

The question still remains whether the BDT is controlled by dislocation nucleation or by dislocaiton motion for a specific material. The effects of crack velocity or strain rate on the BDT are not well understood either. To answer these questions, definitive experimental information is needed. In this communication, we will first briefly describe an experimental technique developed by Brede et al. (1991) and modified by Hsia et al. (1992), and then present preliminary results of our experiments.

EXPERIMENTAL PROCEDURE

Since the cleavage fracture process is nearly temperature-independent, whereas the dislocation generation from a crack tip is strongly temperature-dependent, a definitive way to study the mechanisms of the BDT is to build up a temperature gradient on a sample of cleaveable material, and to propagate a cleavage crack with steady state velocity from the low temperature, brittle side towards the high temperature,ductile side. The crack arrest temperature (the BDT temperature) can then be measured as a function of crack velocity. This experiment not only enables us to obtain accurate information on the BDT temperature for different crack velocities, but also permits us to determine the rate-limiting mechanism through studying of the dislocation structures on fracture surfaces.

For *Si* single crystals, some experimental evidence that the BDT is controlled by dislocation motion have been provided by Brede and Haasen (1988) and by Samuels and Roberts (1989). They observed separately that near the BDT temperature, dislocations can nucleate from crack tips at a stress intensity substantially lower than the critical stress intensity for cleavage fracture. But yet cleavage fracture could occur when the applied loading rate is high. The deficiency of their experimental technique is that the samples are in a uniform temperature field and are loaded from a stationary position so that during loading, the crack tip experiences a full range of stress intensity from zero to the critical level.

Experimental Setup. The experimental technique was developed by Brede et al. (1991) and modified by Hsia et al. (1992). The basic experimental setup is shown in Fig. 1. The fracture sample is put in a vacuum chamber mounted on an Instron loading frame (more detailed

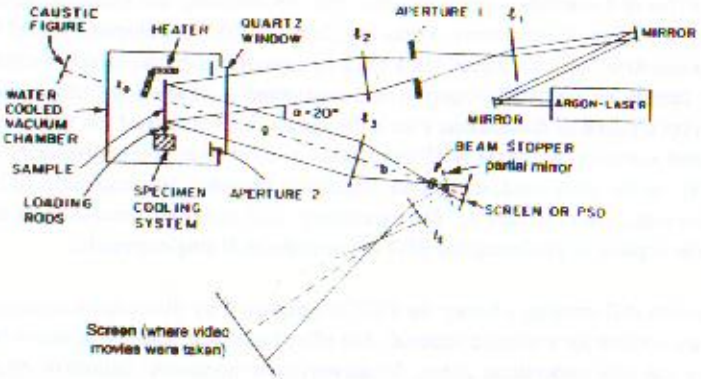

Fig. 1 Basic exeperimental setup for heating/cooling system and crack velocity measurement

description can be found in Brede et al. (1991) and Hsia et al. (1992)). The sample is heated from one side and cooled from the other to achieve the required temperature gradient. Thermo-couples are glued on the sample surface to measure the temperature distribution. The sample is loaded with constant displacement rate at the loading pins so that a nearly steady state crack velocity could be obtained in the constant K samples. Different crack velocity can be achieved by applying different loading rate.

A laser imaging technique has been adopted to measure the crack velocity. The sample surface was optically polished to reflect the laser beam collimated by lenses l_1 and l_2. Through the optical system shown in Fig. 1, a real image of the crack is formed on the screen behind the lens l_3. To obtain a dark field image (bright crack image on a dark background), a beam stopper is placed at the focal point of l_3 so that the reflected parallel light from the sample surface is completely blocked, and only the scattered light from the crack faces could go around the stopper and form a bright image. A position sensing detector (PSD), which converts the optical crack length information into electrical signals, was used to measure the crack length in real time. The data were then recorded by a storage oscilloscope and processed with a PC. The PSD was calibrated by a high speed video camera with a speed of 1000 frames/sec. Although a very low level of noise and a slight distortion of the crack length vs. time curve were observed, the overall performance of the PSD was satisfactory (see Hsia et al. (1992).

Material Studied and Sample Preparation. The material studied in this work was *Si* single crystals, although the technique can be used on other materials as well. *Si* was chosen to be studied first because it has a well defined, sharp transition region, and it has also been studied extensively in the past, so good information exists about its properties. Most of the samples were cut from a heavily doped n-type crystal by electric discharge machining (EDM). A few samples were cut from a heavily doped p-type crystal. Heavily doped crystals were used

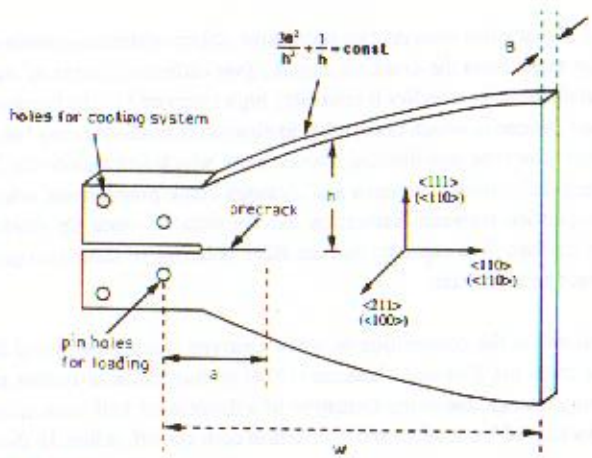

Fig. 2 Constant stress intensity double cantilever beam sample.

because of the conductivity requirement by EDM. Two different sample orientations were tested: one with {111} crack plane and <110> crack growth direction, and the other with {110} crack plane and <110> crack growth direction.

To achieve a well controlled cleavage crack growth, constant stress intensity samples shown in Fig. 2 were used. A serious problem encountered in experiments was that the cleavage cracks would not grow smoothly, but in a jerky manner (i.e., alternative jumps and stops). This behavior is associated with the slight excess of the crack initiation fracture toughness over the crack growth fracture toughness. Since it will be shown later that the crack velocity is far below the elastic wave velocity in the material, no dynamic effect should be present. Therefore, this jerky behavior can not be overcome by changing loading conditions or specimen geometry. To eliminate the crack jumps, some damping was introduced by coating a thin film of ductile metal on the sample surface. This technique has proven to be very effective in stablizing the crack velocity (see details in Hsia et al. (1992)).

EXPERIMENTAL RESULTS

The experimental result of a typical crack arrest test at elevated temperatures with a temperature gradient is shown in Fig. 3. In processing the data acquired by the PSD, a proper smoothing algorithm was employed to filter out undesireable noise. Fig. 3 shows the crack length as a function of time. Initially, the crack was stationary. Then upon loading, the crack started to propagate in the low temperature region, until the crack tip reached a higher temperature and was arrested. Although the loading rate (displacement rate) was kept constant after the crack was arrested, the crack would stay at the arrest location for a while due to plastic flow at the

crack tip. When the plastic deformation could not shield the crack tip fast enough, an unstable cleavage fracture occurred which broke the sample. The data for the final fast fracture is not shown on the plot. The whole process can in fact be viewed as first a transition from the brittle, cleavage fracture mode to the ductile mode, and then a transition from the ductile mode to the cleavage mode by re-initiating a cleavage fracture, although the main interest of the present work is the first transition.

The data of Fig. 3 can be differentiated to obtain the crack velocity as a function of time or crack length. Since there is a one-to-one correspondence between the termperature and crack tip location, a relation between crack velocity and temperature can be obtained. The result is plotted in Fig. 4, which shows that the cleavage crack first accelerated at a temperature level below the BDT, and then decelerated until the crack tip reached the temperature of 900K, where it came to a complete stall (zero velocity). This temperature was thus the BDT temperature for this specific crack velocity. It is noted that if the crack arrest were caused purely by plastic flow induced crack tip shielding, the propagating velocity drop should have been more steep. Therefore it is likely that the deceleration of crack velocity is due partly to the decrease of load caused by the initial crack acceleration, although the final arrest of the crack must be due to generation of plastic flow.

To trace the dislocation activities at crack tips around the crack arrest location, the fracture surface was etched and examined by means of optical and electron microscopy. Fig. 5a shows an SEM picture of etched {110} fracture surface around the crack arrest location. A distinct crack arrest front is clearly seen. On one side of the front is the cleavage fracture surface which is smooth and featureless. On the other side is the roughened, wavy surface representing substantial amount of dislocation activities. In fact, most of the dislocation induced plastic flow occurred right on the arrest front where, although we can not identify individual dislocation etch pits, some dimples are seen which are indications of clusters of dislocations upon etching. It can be expected that the dislocation density at the arrest location is rather high. So it is possible that before the final unstable fast fracture occurred, the crack also experienced some ductile crack propagation by void growth mechanism.

It is interesting to note that a line of dislocations formed before the crack reached the crack arrest front. This can be seen more clearly in Fig. 5b taken with an optical microscope, which shows the same fracture surface with a lower magnification. It should be mentioned that this phenomenon is not always repeatable. A more typical fracture surface would have the distinct crack arrest front with few individual dislocation etch-pits around it. Nevertheless, this line of dislocations demonstrated that even though dislocation nucleation conditions can be met at a propagating crack tip, it may not be able to result in the BDT unless a critical crack tip dislocation configuration or dislocation density is reached. Thus in *Si* single crystals, dislocation mobility is likely to be the rate-limiting mechanism which controls the BDT, as has already been demonstrated by Brede and Haasen (1988) and Hirsch et al. (1989).

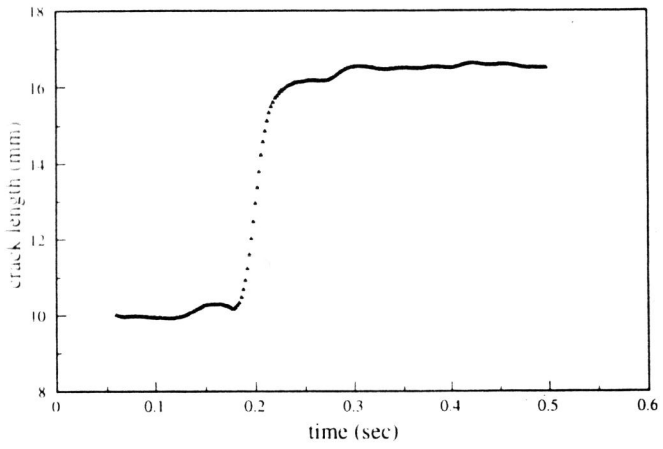

Fig. 3 Measurements of crack length vs. time.

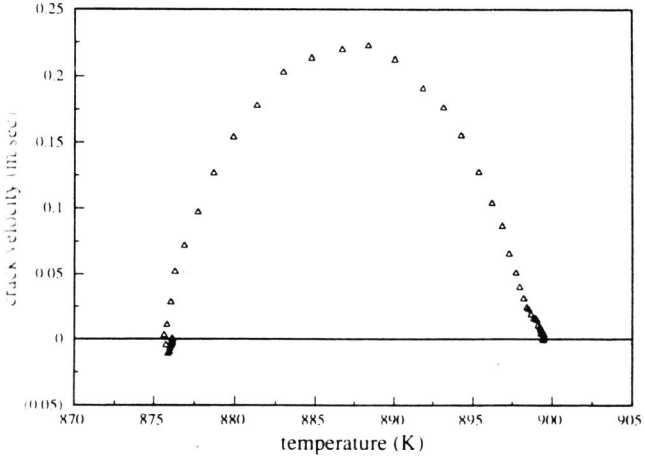

Fig. 4 Results of crack velocity as a function of temperature.

Some additional evidence to support the claim that dislocation mobility controls the BDT in *Si* crystals have been observed on the portion of the fracture surface where fast cleavage fracture occurred at a temperature well above the BDT temperature. There we consistently observed scattered dislocation clusters such as the one shown in Fig. 6. The mechanism to trigger the generation of these dislocations is still unclear. But the shielding effect due to the generation of these dislocations was apparently inadequate to slow down or arrest the fast propagating cleavage crack, although the dislocations could nucleate and their mobility was high enough to form complete loops which extended deep into the material.

CONCLUSIONS AND DISCUSSION

The novel experimental technique developed by Brede et al. (1991) and Hsia et al. (1992) has proven to be an effective way to study the micromechanisms of the BDT in cleavable material systems. The experimental results on *Si* single crystals have indicated that dislocation mobility, rather than dislocation nucleation, is the rate-limiting mechanism in this material system. Similar behaviors have been observed by Dewald et al. (1989) in other material systems such as GaAs and MgO. They observed, using the *in situ* TEM technique, that although dislocations could be generated at or around a cleavage crack tip, it did not prevent the crack from propagating in a cleavage manner. For these material systems in which dislocation mobility controls the BDT, The question which remains to be answered is: what is the critical dislocation configuration at crack tip which would result in the BDT. The etch-pitting technique is no longer good enough because it can not reveal the 3-D dislocation structure at crack tips. X-ray topography can be a candidate technique although its resolution is rather low. The TEM technique will be ideal if samples can be properly cut from the crack arrest location and prepared without destroying the dislocation structure. Clearly, some more theoretical developments are also needed to aid answering these questions.

ACKNOWLEDGEMENT

This research was supported by the Department of Energy through Grant DE-FG02-87ER45294-A003. KJH is also partly supported by the University of Illinois at Urbana-Champaign.

REFERENCES

A. S. Argon (1987), "Brittle to Ductile Transition in Cleavage Fracture", *Acta metall.*, **35**, 185-196.

M. F. Ashby and J. D. Embury (1985), "The Influence of Dislocation Density on the Ductile-Brittle Transition in BCC Metals", *Scripta Metall.*, **19**, 557-562.

M. Brede and P. Haasen (1988), "The Brittle-to-Ductile Transition in Doped Silicon as a Model Substance", *Acta metall.*, **36**, 2003-2018.

M. Brede, K. J. Hsia and A. S. Argon (1991), "Brittle Crack Propagation in Silicon Single Crystals", *J. Appl. Phys.*, **70**, 758-771.

K. S. Cheung, A. S. Argon and S. Yip (1991), "Activation Analysis of Dislocation Nucleation from Crack Tip in α-Iron", *J. Appl. Phys.*, **69**, 2088-2096.

D. K. Dewald, T. C. Lee, I. M. Robertson, and H. K. Birnbaum (1989), "Dislocation Structures Ahead of Advancing Cracks", *Scripta Metall.*, **23**, 1307-1312.

P. B. Hirsch, S. G. Roberts and J. Samuels (1989), "The Brittle-Ductile Transition in Silicon: II. Interpretation", *Proc. R. Soc. London*, **A421**, 25-53.

K. J. Hsia, A. S. Argon and M. Brede (1992), "Experimental Study of the Brittle-to-Ductile Transition in Silicon Single Crystals", to be submitted.

R. E. Peierls (1940), *Proc. Phys. Soc.*, **52**, 23.

J. R. Rice (1992), "Dislocation Nucleation from a Crack Tip: an Analysis Based on the Peierls Concept", *J. Mech. Phys. Solids*, **40**, 239-271.

J. R. Rice and R. Thomson (1974), "Ductile versus Brittle Behavior of Crystals", *Phil. Mag.* **29**, 73-97.

J. Samuels and S. G. Roberts (1989), "The Brittle-Ductile Transition in Silicon: I. Experiments", *Proc. R. Soc. London*, **A421**, 1-23.

Fig. 5a Micrograph of an etched {110} fracture surface at the crack arrest location.

Fig. 5b The same fracture surface as in Fig. 5a with a lower magnification.

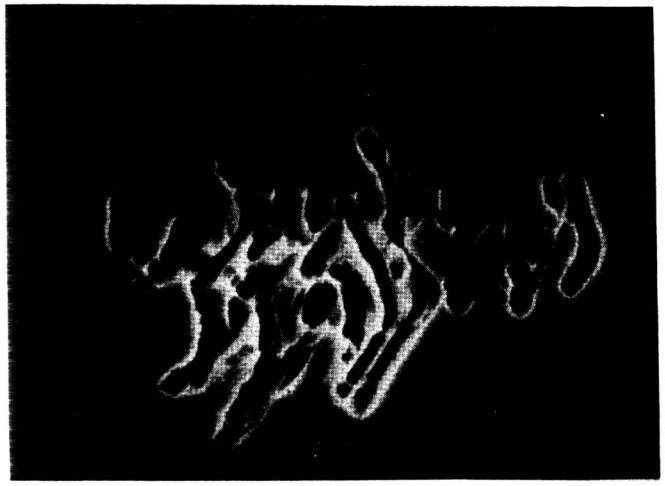

Fig. 6 Micrograph of traces of dislocations during fast fracture at a temperature higher than the BDT temperature.