

# **ESTIMATION OF THE RESISTANCE AGAINST STABLE CRACK INITIATION AND UNSTABLE CRACK PROPAGATION USING R-CURVES AND STABILITY ASSESSMENT DIAGRAMS IN DUCTILE POLYMERIC ABS-MATERIALS**

R. Lach and W. Grellmann

Institute of Material Science, Martin-Luther University Halle-Wittenberg,  
D-06099 Halle/S., Germany

## **ABSTRACT**

The toughness behaviour of various ABS-materials under impact-like loading was characterized using different experimental and approximative methods of fracture mechanics to determine critical  $J$ -values, crack resistance curves and  $J$ - $T_I$  stability assessment diagrams. By use of  $J$ - $T_I$  stability assessment diagrams, critical values as resistance against unstable crack initiation can be calculated, also if unstable crack propagation can not be directly observed because of limited specimen boundaries and defined loading conditions respectively. Because stable crack propagation is followed by an unstable one, two 'brittle-to-tough' transitions (BTT) are observed for impact-resistant plastics like ABS. Whereas the 'tough-to-high impact' transition is caused by the matrix, the conventional 'brittle-to-tough' transition is sensitive to the structure and the deformation mechanisms.

## **INTRODUCTION**

Corresponding to the growing application field of polymer materials not only for common but also for technical purposes, besides the improvement of stiffness and strength the optimization of toughness behaviour has become a major goal in polymer research [1]. In this connection, the measurement of crack resistance curves (R-curves) using the stop-block method in multiple-specimen technique and computation of R-curves by means of various procedures [2-4] was proved successfully to evaluate the toughness especially as material resistance against stable crack initiation and propagation. However, that is not highly economical because the experimental determination of R-curves consumes too much time and a great personnel effort is necessary. Therefore, in recent times, some research groups work on the field of development of approximative methods to determine R-curves and to apply these methods on polymer materials [5-7]. Furthermore, because the use of standardized specimens is the basis to compare of results between different groups the approximative definition of instability values is of a great scientific interest even if unstable crack propagation can not be directly observed [8].

In literature, only a small number of fracture mechanics investigations preoccupying with the crack resistance behaviour of ABS-materials can be found [9-13]. Most of these investigations were performed using quasi-static loading conditions [9-13]; impact-like loading conditions were very seldom used [9]. In the light of an adequate quantification of the toughness behaviour for ABS as a typical impact-toughened polymer material that is combined with pronounced elastic-plastic behaviour and predominantly stable crack

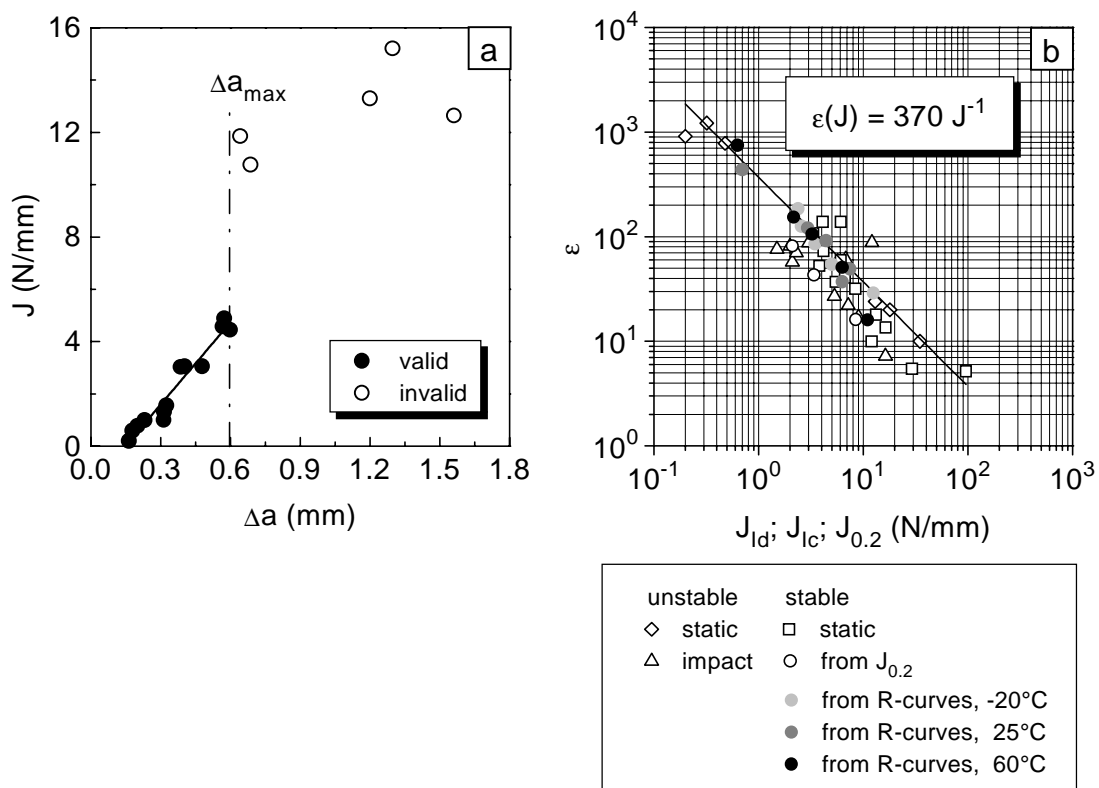
propagation, various concepts of elastic-plastic fracture mechanics those reflect different aspects of the mechanical behaviour should be used. Therefore, in future, a main point of view should be formed by toughness assessment under impact-like loading conditions, for example the determination of so-called ‘dynamic’ crack resistance curves. In some publications, increasing slopes of R-curves and  $J_{0.2}$ -values have been observed in dependence on the test speed [9] and the rubber content [10,12] respectively. The  $J_{0.2}$ -values for two materials showed an opposite manner in the temperature range from  $-50\text{ }^{\circ}\text{C}$  to  $20\text{ }^{\circ}\text{C}$  [9]: Whereas  $J_{0.2}$  increased for a bulk-polymerized ABS with increasing temperatures,  $J_{0.2}$  decreased for an emulsion-polymerized ABS in the given range of temperature, that is caused by different microstructures. From the crack initiation values and tearing moduli versus temperature dependencies ( $-20\text{--}80\text{ }^{\circ}\text{C}$ ) of different emulsion-polymerized ABS-materials, the crack initiation values yielded a minimum value and the tearing moduli a maximum value at  $80\text{ }^{\circ}\text{C}$  [12].

## EXPERIMENTAL AND THEORETICAL BASIS

### *Determination of Fracture Mechanics Parameters as Material Resistance against Stable Crack Initiation and Propagation under Impact-Like Loading Conditions*

#### *Experimental Determination of ‘Dynamic’ Crack Resistance Curves*

For the characterization of the stable crack initiation and propagation the stop-block method was used where the variation of crack growth  $\Delta a$  is realized by defined limiting of specimen deflection, and crack resistance curves (R-curves) under impact-like loading conditions, so-called ‘dynamic’ R-curves, were recorded [2,14]. The basis of computing  $J$ - and COD-values is formed by measuring load-deflection diagrams [2]. R-curves can be evaluated using different procedures, for instance the standard draft ESIS TC4 [15] where, however, a well-established standardized R-curve routine that has been successfully applied to all polymers does not exist yet. Because of the given data point distribution a linear relationship between the loading parameters ( $J$ ,  $\delta$ ) and the



**Figure 1:** Evaluation of a  $J$ - $\Delta a$  crack resistance curve (ABS 270, 24 wt.-% rubber,  $25\text{ }^{\circ}\text{C}$ ) (a): valid ( $\lambda$ ) and invalid ( $\gamma$ ) data,  $\Delta a_{\max}$  – maximum limit of stable crack growth; Geometry factor  $\varepsilon$  versus  $J$ -values (b): from R-curves for ABS 270 ( $-20\text{ }^{\circ}\text{C}$ ,  $25\text{ }^{\circ}\text{C}$ ,  $60\text{ }^{\circ}\text{C}$ ) and other experimental results [1,2,16,17]

stable crack growth  $\Delta a$  is supposed [14] (Eqn. 1):

$$J; \delta = C_1 \Delta a + C_2 \quad (1)$$

where the fit parameters  $C_1$  and  $C_2$  are dependent on material and loading conditions (test speed, temperature). In order to quantify the resistance against stable crack initiation,  $J_{0.2}$ - and  $\delta_{0.2}$ -values are determined at engineering crack initiation point corresponding to a crack growth  $\Delta a = 0.2$  mm. The tearing moduli  $T_J$  and  $T_\delta$  those are defined after Eqn. 2a,b

$$T_J = \frac{dJ}{d(\Delta a)} \frac{E_d}{\sigma_{yd}^2} \quad \text{and} \quad T_\delta = \frac{d\delta}{d(\Delta a)} \frac{E_d}{\sigma_{yd}} \quad (2a,b)$$

make possible quantification of the material resistance against stable crack propagation ( $E_d$  – dynamic Young's modulus,  $\sigma_{yd}$  – dynamic yield stress).

### *Geometry-Independent Dynamic R-Curves*

For determination of material-specific fracture mechanics parameters, the  $J$ - $\Delta a$  and  $\delta$ - $\Delta a$  data respectively should be geometry-independent, i.e. the minimum specimen geometry limits should be utilized. These minimum specimen geometry limits can be estimated exemplary for the  $J$ -integral concept after Eqn. 3

$$B; a; W - (a + \Delta a) \geq \varepsilon \frac{J}{\sigma_{yd}} \quad (3)$$

where the length of the ligament  $W-(a+\Delta a)$  is the most critical quantity ( $B$ ,  $W$  – specimen thickness and width,  $a$  – initial crack length). The geometry factor  $\varepsilon$  ranging from 7.2 to 1220 is material-dependent [2,16,17]. Because of the finite specimen dimension (especially the limited ligament length), during crack propagation, a transition from predominantly plane strain to predominantly plane stress state is observed. Corresponding to that, a strong increase of  $J$ - and  $\delta$ -values is found because the external energy can not be dissipated into a greater volume due to the interaction of the plastic zone with the specimen boundary but a higher plastic deformation capacity in correspondence to a greater energy density in a limited volume occurs. Furthermore, a propagation of the crack into the pressure zone of the specimen is possible, that also causes an increasing crack resistance. The main conclusion is that only up to the begin of the strong-increasing  $J$  (Figure 1a) and  $\delta$  respectively, i.e. for  $\Delta a \leq \Delta a_{\max}$ , valid R-curve data are measured. Using the experimental derived maximum crack growth  $\Delta a_{\max}$  and  $J(\Delta a_{\max})$  the geometry independence of fracture mechanics values for  $\Delta a \leq \Delta a_{\max}$  is confirmed by application of the polymer-specific  $\varepsilon$ - $J$  relation (Figure 1b) [2,16,17].

### *Fracture Safety Criteria of Elastic-Plastic Fracture Mechanics*

For greater stable parts of crack propagation (ductile behaviour) following criteria of fracture safety can be formulated according to Blumenauer and Pusch [18] by use of  $J$ - $\Delta a$  crack resistance curves ( $J$ -R curves):

#### *Material Resistance against Stable Crack Initiation*

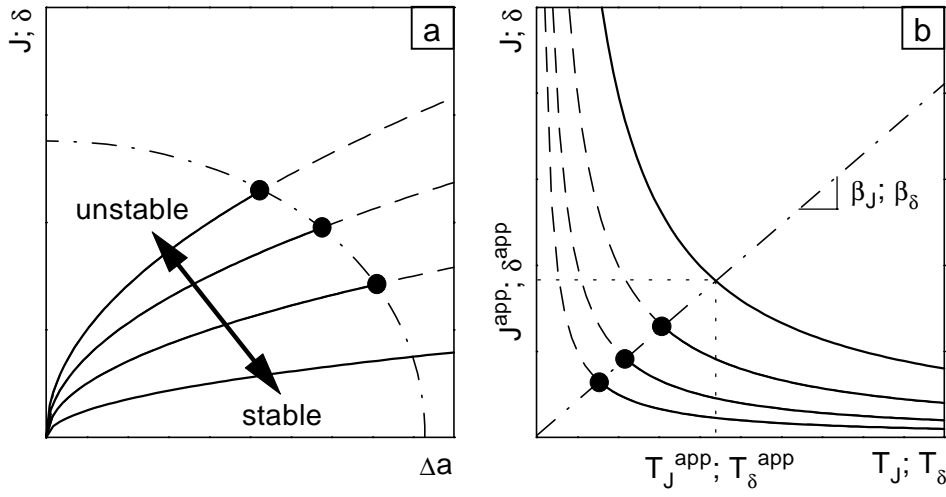
$$J^{\text{component}} \leq J_i \quad \text{or} \quad J^{\text{component}} \leq J_{0.2} \quad (4)$$

The loading parameter  $J^{\text{component}}$  is either numerically computed or by using approximation methods.

#### *Material Resistance against Stable Crack Propagation*

$$\frac{dJ^{\text{component}}}{d(\Delta a)} < \frac{dJ^{\text{material}}}{d(\Delta a)} \quad (5)$$

The measurement of  $J$ - $\Delta a$  curves makes quantification of the material parameters on the right side of unequations 4 and 5 possible. Regarding unequation 4 crack initiation values can be derived either at the physical initiation point or in form of engineering crack initiation values  $J_{0.2}$ .



**Figure 2:** Scheme of transformation of  $J-\Delta a$  crack resistance curves (a) with ( $\lambda$ ) or without points of instability into  $J-T_J$  stability assessment diagrams (b) to determine approximative fracture mechanics values,  $J^{app}$  and  $\delta^{app}$ , as resistance against unstable crack initiation

### $J-T_J$ stability assessment diagram

Furthermore, as another method for polymer materials,  $J-T_J$  stability assessment diagrams [18;20] can be used to evaluate the failure safety, i.e. the safety against unstable crack propagation, if limited crack growth is observed. By use of the tearing modulus (Eqn. 2a), an instability criterion to avoid a crack instability corresponding to increasing crack growth can be formulated [19]:

$$T_J^{component} \leq T_J^{material} \quad (6)$$

where the loading parameter  $T_J^{component}$  can be estimated from maximum plastic load. Thus, unstable crack propagation can be observed, if the tearing modulus of the component is greater than that of the material.

Generally, by using load-deflection diagrams of specimens those show predominantly unstable crack propagation, critical  $J$ -values  $J_{Id}$  as resistance against unstable crack initiation can be computed by means of various approximation methods based on the energetical interpretation of the  $J$ -integral [1,18]. In all other cases, if no unstable crack propagation is observed because of the finite specimen geometry, i.e. for predominantly stable crack propagation, nevertheless it is possible to estimate approximative  $J$ -values  $J^{app}$  as resistance against unstable crack initiation on the basis of the instability criterion (Eqn. 6). In order to evaluate components,  $J-\Delta a$  curves (Figure 2a) are transformed into  $J-T_J$  plots, the so-called  $J-T_J$  stability assessment diagrams (Figure 2b).

TABLE 1  
EMPIRICAL PARAMETERS  $\beta_J$  AND  $\beta_\delta$  FOR ABS AND OTHER PLASTICS

Materials	$\beta_J$ (N/mm)	$\beta_\delta$ ( $10^{-3}$ mm)
ABS [8]	2.0 and 26	
ABS (this work)	10	75
all plastics [8]	0.06-31	2.7-75

According to an idea of Paris and Johnson [20] the  $J-T_J$  curves are intersected with the load line

$$J_{Id} = \beta_J T_{Jd} \quad (7)$$

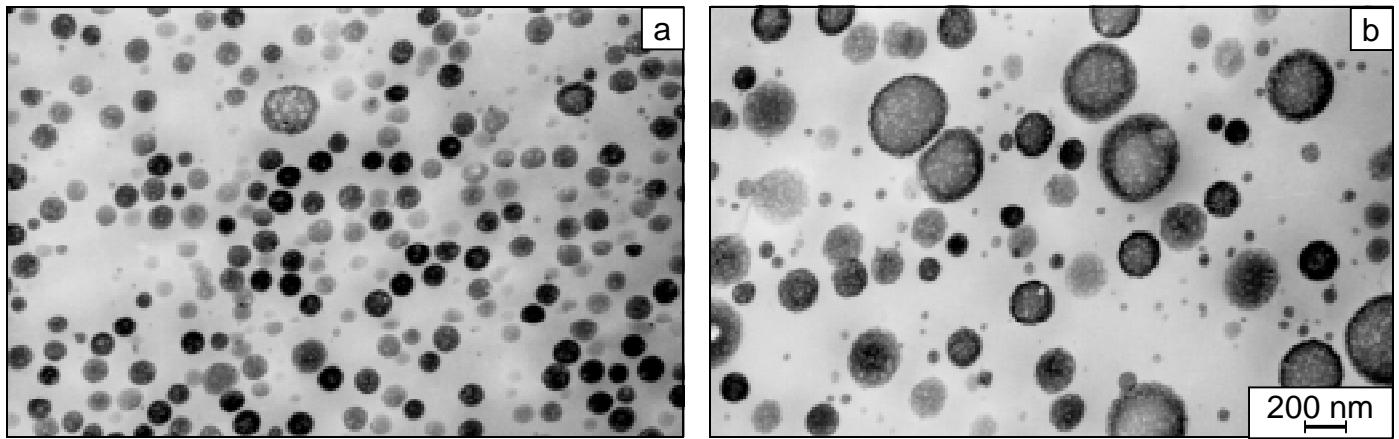
at the instability point introducing a material-specific parameter  $\beta_J$ ;  $T_{Jd}$  is named tearing instability. Because an empirical value  $J_{50}$  (with  $\beta_J = 50$  psi-in = 8.8 N/mm) [20] defined as an instability value  $J^{app}$  is not useful for polymer materials [8],  $J^{app}$ -values for ABS are estimated from

$$J^{\text{app}} = \beta_J C_1 \frac{E_d}{\sigma_{yd}} \quad (8)$$

Analogous to  $J$ - $T_J$  diagrams,  $\delta^{\text{app}}$  can be derived from  $\delta$ - $T_\delta$  stability assessment diagrams by introduction of a parameter  $\beta_\delta$ . If experimental  $J_{\text{ld}}$ - or  $\delta_{\text{ld}}$ -values exist, a calculation of  $\beta_J$  or  $\beta_\delta$  is possible for given materials or material groups (see Eqn. 7 and Figure 2b). In Table 1  $\beta_J$ - and  $\beta_\delta$ -values for plastics are summarized.

## MATERIALS

Three emulsion-polymerized ABS-material groups those differ from each other concerning the averaged particle diameter and the particle size distribution were investigated (Figure 3a,b and Table 2). For all materials, small rubber particles are finely dispersed in a SAN matrix ( $M_w = 85000$  g/mol,  $M_w/M_n = 4$ , oligomer content  $\approx 1$  %, styrene/acrylonitrile-ratio = 72.5/27.5). The rubber content is varied from 0 wt.-% (pure SAN) to 36 wt.-%. Whereas the rubber particle size distribution of ABS 110 that was measured using the ultra-centrifugation method shows one pronounced peak, the size distributions of ABS 270 as well as of ABS 330 are characterized by three separated peaks.



**Figure 3:** Transmission electron microscopic micrographs of ultra-thin sections stained with  $\text{OsO}_4$  for ABS 110 (a) and ABS 330 (b) with 16 wt.-% rubber

TABLE 2

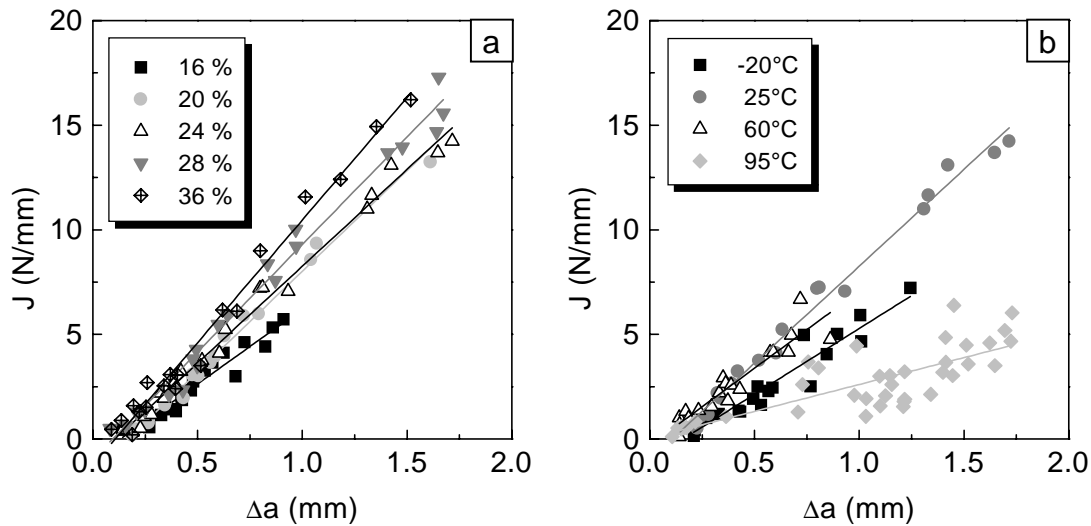
PEAK PARTICLE DIAMETERS, AVERAGED PARTICLE SIZES  $d$  AND RELATIVE PARTICLE NUMBER CORRESPONDING TO THE PEAKS

Materials	Particle diameter		
	$d$ (nm)	Peak at (nm)	Relative number (%)
ABS 110	110	110 / 270 / 415	98 / 1 / 1
ABS 270	270	130 / 280 / 480	23 / 64 / 13
ABS 330	330	95 / 210 / 475	21 / 27 / 52

## TOUGHNESS BEHAVIOUR IN DEPENDENCE ON RUBBER CONTENT AND TEMPERATURE

The determination of  $J$ - $\Delta a$  and  $\delta$ - $\Delta a$  crack resistance curves ( $J$ -R and  $\delta$ -R curves) results in quantification of engineering toughness values as material resistance against stable crack initiation and propagation. Because

ABS 110 materials show only macroscopically linear-elastic behaviour corresponding to predomi-



**Figure 4:**  $J$ - $\Delta a$  crack resistance curves of ABS 330 versus rubber content at room temperature (a) and in dependence on temperature for 24 wt.-% rubber (b)

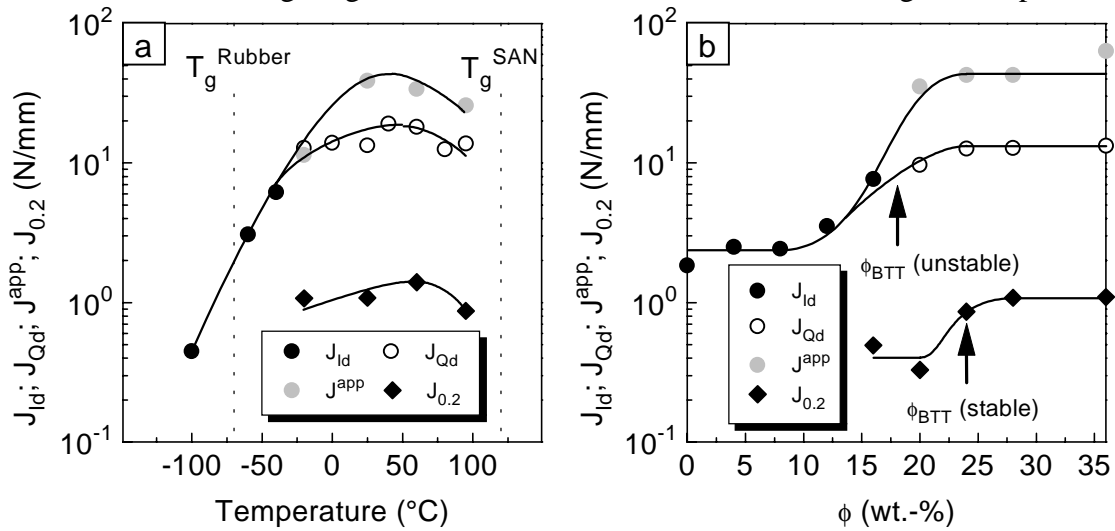
nantly unstable crack propagation, no crack arrest and, thus, no R-curves could be produced. Analogous to that, an R-curve determination of all ABS 270 and ABS 330 materials was also impracticable at low temperatures and/or low rubber contents because these materials show predominantly unstable crack propagation at temperatures  $< -40$  °C and rubber contents  $< 16$  wt.-%. Although a single-parameter description of the toughness behaviour is impossible [1], also for the ABS-material presented in this study [21], the followed passages are confined to one loading parameter, the  $J$ -integral.

Figures 4a,b show  $J$ - $R$  curves of ABS 330 as a function of rubber content (25 °C) and temperature (24 wt.-% rubber) (in view of R-curves of ABS compare also [14,22]). From Figure 4a it is clearly visible that the slopes  $dJ/da$  of the R-curves and, thus, the tearing moduli  $T_J$  increase with increasing rubber contents [23]. In contrast to this observation, the resistance against stable crack propagation  $dJ/da$  and  $T_J$  respectively pass a maximum value in the range of 25–60 °C (Figure 4b) [23]. In this connection, the pronounced toughness decrease at 95 °C that corresponds to a clearly visible increasing data dispersion should be pointed out. The rubber content and temperature dependencies of the  $J$ -values  $J_{0,2}$  as resistance against stable crack initiation are qualitatively comparable to those of  $dJ/da$  [14,22], but  $J_{0,2}$  reflects the matrix toughness behaviour to a high degree and  $T_J$  gives morphology-sensitive information corresponding to the particle-matrix structure [1]. However,  $J_{0,2}$  does not correspond to the physical initiation process because of its empirical definition. An explanation of the increasing  $J_{0,2}$ -values in dependence on rubber content is the interaction between matrix-SAN and SAN surrounded the rubber particles in form of about 10 nm thick graft shells. The resulting changes in molecular matrix structure those increase with rising rubber content have a great influence on  $J_{0,2}$  [22].

Figures 5a,b give a survey of the  $J$ -values in dependence on rubber content and temperature. On the one hand, the approximative  $J$ -values  $J^{app}$  as resistance against unstable crack initiation those were calculated using  $\beta_J = 10$  N/mm are compared to the conventional critical  $J_{Id}$  and the  $J$ -values  $J_{Qd}$  measured at maximum load. Additionally to these quantities, on the other hand,  $J_{0,2}$ -data are also plotted in Figures 5a,b. A main methodical result is that values of fracture resistance can be quantified in respect of unstable crack initiation by use of  $J$ - $T_J$  stability assessment diagrams, also in cases if unstable crack propagation can not be directly observed because of the limited specimen geometry and defined loading conditions respectively [8]. Especially, this method is substantial to evaluated the fracture mechanical behaviour of new polymer materials by means of miniature specimens because these materials are often synthesized in the first stage of development in small samples. Furthermore, the  $J_{Qd}$ -values are impracticable to assess the crack resistance because they significantly undervalue the real level of toughness.

All  $J$ -values increase generally up to a maximum value at 25–60 °C and decrease after passing this maximum point (Figure 5a) [14,21,22,24]. Whereas the causes of increasing toughness versus temperature

(especially, at temperatures higher than the glass temperature  $T_g^{\text{Rubber}}$  of rubber) have been well established for ABS, the causes of decreasing toughness those can be observed closed to the glass temperature  $T_g^{\text{SAN}}$  of

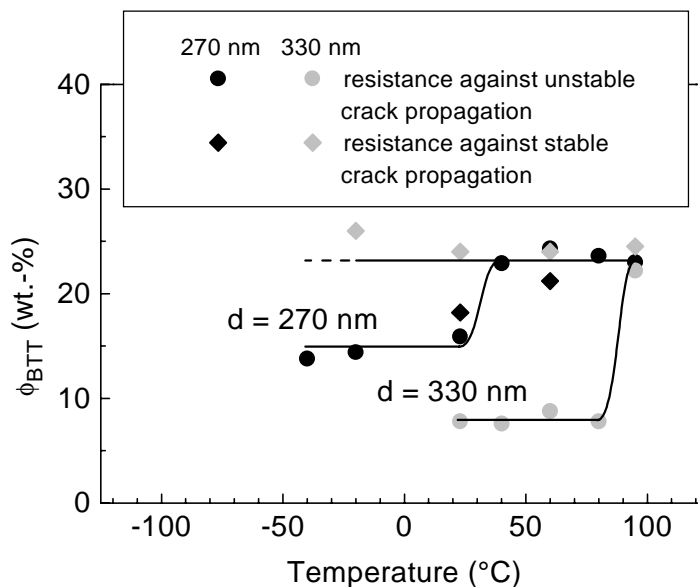


**Figure 5:**  $J$ -values for ABS 330 as resistance against unstable ( $J_{Id}$  and  $J^{app}$ ) and stable crack initiation ( $J_{0.2}$ ) and  $J$ -values measured at maximum load ( $J_{Qd}$ ) respectively in dependence on temperature for 28 wt.-% rubber (a) and on rubber content  $\phi$  at room temperature (b);  $\phi_{BTT}$  – ‘brittle-to-tough’ transition concentrations corresponding to crack resistance against stable or unstable crack initiation;  $T_g^{\text{Rubber}}$  and  $T_g^{\text{SAN}}$  – glass temperatures of disperse rubber phase and of matrix (SAN) at loading frequencies comparable to that in impact tests (~1 kHz)

SAN-matrix are fundamentally unsolved. This phenomenon that was detected most clearly at a rubber content of 16 wt.-%, is connected with a reduction in the size of the stable crack growth area and the plastic zone size [21,24] and does not result from physical ageing [21]. Besides the decrease in the Young’s modulus at higher temperatures, another causes are matrix-embrittlement effects by initiation and growth of so-called intrinsic crazes [21].

The application limits of plastics can be described, for example, using ‘brittle-to-tough’ transition (BTT) concentrations  $\phi_{BTT}$  those are formally calculated by averaging the lower and upper shelf of toughness (Figure 5b). Some impact-resistant polymer materials, like ABS [21] and heterogene PP-materials [25], show two BTT’s:

- BTT(1) of  $J_{Id}$  and  $J^{app}$  as resistance against unstable crack initiation (conventional transition ‘brittle/tough’),
- BTT(2) of  $J_{0.2}$  as resistance against stable crack initiation (transition ‘tough / high impact’).



**Figure 6:** Temperature dependencies of ‘brittle-to-tough’ transition concentrations  $\phi_{\text{BTT}}$  from resistance against stable or unstable crack initiation for ABS 270 and ABS 330

Because the stable crack propagation is followed by an unstable one concentrations of BTT(2) are equal or smaller than that of BTT(1) (Figure 6). In accordance to investigations of heterophase PP-materials [25], for ABS [21], the ‘tough-to-high impact’ transition is caused by the matrix and, thus, it is independent on temperature and the particle size [21] or the relative particle distance [25] respectively. In contrast to this observation, the BTT-concentrations [25] and the relative critical interparticle distances [25] measured at the transition point BTT(1) converge to the values of ‘tough-to-high impact’ transition {BTT(2)} for higher or lower temperatures respectively. Summarizing it is to be pointed out that only the BTT(1) is sensitive to the structure and the deformation mechanisms.

## ACKNOWLEDGEMENT

The Bayer AG, Leverkusen, especially Dr. P. Krüger, is gratefully acknowledged for providing the ABS-materials. The authors also thank for the financial support of the Deutsche Forschungsgemeinschaft (DFG).

## REFERENCES

1. Grellmann, W. and Seidler, S. (Eds.) (2000) *Deformation and Fracture Behaviour of Polymers*. Springer, Berlin Heidelberg, in press.
2. Grellmann, W., Seidler S. and Hesse, W. (2000) In: *Deformation and Fracture Behaviour of Polymers*, Grellmann, W. and Seidler, S. (Eds.). Springer, Berlin Heidelberg, in press.
3. Grellmann, W. and Seidler, S. (1999) In: *Material Mechanics – Fracture Mechanics – Micro Mechanics*, pp. 336-341, Winkler, T. and Schubert, A. (Eds.). DDP Goldenberg, Dresden.
4. Seidler, S., Grellmann, W. (1995) *Polym. Test.* **14**, 453.
5. Steiner, R. and Grellmann, W. (2000) In: *Deformation and Fracture Behaviour of Polymers*, Grellmann, W. and Seidler, S. (Eds.). Springer, Berlin Heidelberg, in press.
6. Che, M., Grellmann, W., Seidler, S. and Landes, D. (1997) *Fatig. Fract. Engng. Mater. Struct.* **20**, 119.
7. Bernal, C.R., Montemartini, P.E. and Frontini, P.M. (1996) *J. Polym. Sci. / Part B: Polym. Phys.* **34**, 1869.
8. Lach, R. and Grellmann, W. (2000) In: *Deformation and Fracture Behaviour of Polymers*, Grellmann, W. and Seidler, S. (Eds.). Springer, Berlin Heidelberg, in press.
9. Castellani, L., Frassine, R., Pavan, A. and Rink., M. (1996) *Polymer* **37**, 1329.
10. Giaconi, G.F., Casttelani, L., Maestrini, C. and Ricco, T. (1998) *Polymer* **39**, 6315.
11. Bernal, C.R., Frontini, P.M., Sforza, M. and Bibbo, M.A. (1995) *J. Appl. Polym. Sci.* **58**, 1.
12. Bernal, C.R., Cassanelli, A.N. and Frontini, P.M. (1995) *Polym. Test.* **14**, 85.
13. Lu, M.-L., Lee, C.-B. and Chang, F.-C. (1995) *Polym. Engng. Sci.* **35**, 1433.
14. Han, Y., Lach, R. and Grellmann, W. (2000) *J. Appl. Polym. Sci.* **75**, 1605.
15. Standard Draft ESIS TC4 (1992) *A Testing Protocol for Conducting J-Crack Growth Resistance Curve Tests on Plastics*.
16. Seidler, S. and Grellmann, W. (1999) *Intern. J. Fract., Letters Fract. Micromech.* **96**, L17.
17. Grellmann, W. and Seidler, S. (1994) *Intern. J. Fract.* **68**, R19.
18. Blumenauer, H. and Pusch, G. (1993) *Technische Bruchmechanik*. Deutscher Verlag für Grundstoffindustrie, Leipzig.
19. Tada, H., Paris, P.C. and Irwin, G.R. (1985) *The Stress Analysis of Crack Handbook*. Paris Production Inc.
20. Paris, P.C. and Johnson, R.E. (1983) *ASTM STP* **803**, 5.
21. Han, Y., Lach, R. and Grellmann, W. (2000) *J. Appl. Polym. Sci.*, in press.
22. Lach, R., Grellmann, W. and Krüger, P. (2000) In: *Deformation and Fracture Behaviour of Polymers*, Grellmann, W. and Seidler, S. (Eds.). Springer, Berlin Heidelberg, in press.
23. Han, Y., Lach, R. and Grellmann, W. (1999) *Appl. Macromol. Chem. Phys.* **270**, 5.
24. Han, Y., Lach, R. and Grellmann, W. (1999) *Appl. Macromol. Chem. Phys.* **270**, 13.
25. Grellmann, W., Seidler, S., Jung, K. and Kotter, I. (2000) *J. Appl. Polym. Sci.*, in press.