Investigation on fracture of fibre/adhesive bondlines

March 1985

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Acknowledgement

The author gratefully acknowledges Prof. dr. ir. J. Schijve for his theoretical contribution and C. Paalvast for drawing the figures.

These investigations were supported by the Netherlands Foundation for Technical Research (S.T.W.).
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Chapter 1: Introduction

In the past, a considerable amount of experimental and theoretical work was performed to obtain strain energy release rate parameters ($G_{IC}, G_{2C}$) for different kinds of adhesives (ref 1,2,3). At the department of Aerospace Engineering of the Delft University of Technology, extensive studies proved that the mechanical properties of metal sheet laminated materials and structures can be significantly improved by introducing unidirectional fibres in the adhesive layers (ref 4). However in these experiments it was also found that for a certain combination of fibres and adhesives, a weak link can be present in the material, which is the fibre/adhesive interface. It was shown that this can apply for several loading modes (shear, tension or a combination of both).

In this report, the behaviour of the fibre reinforced adhesive is examined in both a tension and shear loading mode, using two standardized specimen configurations:

- tension mode: width tapered double cantilever beam (W.T.D.C.B.-) specimen
- shear mode: thick adherend (T.A.-) specimen

In the test series on both types of specimens different parameters have been introduced:

- type of fibre (aramid, glass, carbon)
- fibre orientation
- environmental condition

Further an attempt is made to relate the experimental results with the general fracture toughness behaviour of the material using an energy balance approach. Because a profound discussion of the results is not possible without a fundamental knowledge of the theoretical backgrounds, also a brief review is presented of some important subjects such as:

- strain energy release rate $G$ of the two specimen configurations
- fracture mechanics of anisotropic materials based on an energy balance
- adhesion parameters of importance for organic materials
Chapter 2: Experimental details

As mentioned in the introduction, two different specimen configurations were used:

- width tapered double cantilever beam specimen (mode 1 testing - fig 1), abbreviation: W.T.D.C.B.- specimen

- thick adherend specimen (mode 2 testing - fig 2)
  abbreviation: T.A.- specimen

In this chapter, first the material variables will be discussed followed by a brief survey of both test methods.

2.1: Materials

Both specimen configurations consist of 2 aluminium adherends with an interfacial layer (adhesive or a prepreg):

aluminium : In both test series (W.T.D.C.B. and T.A.) 7075-T6 aluminium alloy was used in the bare condition.
Before curing, the adherends were carefully pretreated which includes:
- degreasing for 30 minutes at 70 °C
- pickling for 20 minutes at 50 °C
Intermediate rinsing of the adherends between the pretreatments was performed using distilled water.

interfacial layer

In the test series 4 different configurations were used:

1) adhesive: AF 163-2K (3M company)
The AF 163-2 is a rubber toughened epoxy metal adhesive. In this test series a type with a carrier was used

2) Aramid prepreg: aramid prepreg NPE 9055 (3M company)
The prepreg layer consists of:
- epoxy metal adhesive (AF 163-2U - 3M company)
- high modulus aramid fibres (TWARON HM - ENKA)
A fibre volume content of 50% is standardized.
The fibre orientation is unidirectional.

* carrier: in most structural adhesives a carrier is used to assure a constant adhesive thickness after curing. The use of a carrier mostly results in somewhat lower but much more reproducible properties than can be obtained for the bulk adhesive
3) Carbon prepreg: The prepreg layer consists of:
   - epoxy metal adhesive: AF 163-2U (3M-company)
   - high strength carbon fibres: T 300HS (Toray)

Fibre orientation and volume fraction are identical to those of the aramid prepreg.

4) Glass prepreg (only used in T.A.-specimen)

The prepreg consists of:
   - epoxy metal adhesive: AF 163-2U (3M-company)
   - glass fibres: S-glass (Owens&Corning)

Fibre orientation and volume fraction are again identical to those of the aramid prepreg.

The curing of the specimens was performed in a computer controlled autoclave. For all the interfacial layers, the curing cycle was identical:
   - temperature: 125 °C
   - pressure: 3 atm. (vacuum)
   - time: 1 hour

2.2: $G_{\infty}$ test (W.T.D.C.B.- specimen)

An increase of the crack length in the W.T.D.C.B.-specimen (fig 1) implies a reduction of the strain energy release rate $G$ if the displacement of the load remains constant.

As a result, crack growth can be easily controlled without the risk of complete failure, which makes the specimen very attractive for crack growth studies at the interface under well controlled conditions.

The specimen is loaded in a 20-tons computer controlled testing machine (AMSLER - fig 3). For the environmental tests (distilled water exposure at 20°C and 50°C), a special environmental cell was designed (fig 4). To obtain identical initial conditions for all tests, the $G_{\infty}$-measurements (or in this case load measurements - see chapter 4) were started after precracking the specimen until a crack length of $a=90$ mm. The following procedure was used for this purpose:

- the precrack is initiated in the rectangular part of the specimen by loading the specimen in the machine; the initial crack length is a function of the applied initial load and the material
properties.

- the crack is forced to grow using a constant displacement rate \( \frac{dv}{dt} = 50\text{mm/h} \) until \( a = 90\text{mm} \) after which the desired displacement rate and/or environmental condition is introduced. The load record \( P-t \), which is the only required test result, is recorded on an X-t recorder (load versus time). A typical example of such a load recording is shown below.

\[ \begin{align*}
\text{P} & \quad \text{initial load} \\
\text{start of Gic measurement} \\
a = 90\text{mm}
\end{align*} \]

During environmental testing (see chapter 3.5) a significant increase of \( G_{ic} \) with increasing crack length was observed. In this case the necessary crack length measurements were performed using an optical microscope.

After specimen failure, fracture surface examination was performed at the ENKA-company using electron microscopy.

Some theoretical remarks concerning the type of specimen are made in chapter 4.

2.3: \( G_{ic} \)-test (T.A.-specimen)

The test is performed in a 6-tons tensile testing machine and, as for the \( G_{ic} \)-test, a constant displacement rate is used \( \frac{dv}{dt} = 30\text{mm/hour} \). The load increases until the critical load level is reached and failure occurs. Also for this test method, the critical load is the only required test result for calculating the strain energy release rate. Some specimens were before testing exposed in a distilled water (60°C) or saltspray (35°C) environment.

In chapter 4, some theoretical remarks are made concerning this test method.
2.4: Specification of different fracture planes

In this report, considerable efforts are made to examine the fracture paths in the different interfacial layers (adhesive or prepreg). Because of the complex built-up of these layers it is necessary to define the different possible fracture paths:

- for the adhesive layer:
  1: cohesive failure in the adhesive (type 1)
  2: interface failure between adherend and adhesive (type 2)
  3: interface failure between carrier and adhesive (type 3)

- for the prepreg layer:
  1: cohesive failure in the matrix (type 1)
  2: cohesive failure in the fibres (type 4)
  3: interface failure between adherend and matrix (type 2)
  4: interface failure between fibre and matrix (type 5)

All fracture paths will be a combination of the above mentioned 5 types.
Chapter 3: Results

In this chapter test results will be presented and observed trends are indicated. The variables of the test series are:

\( G_{1c} \): - influence of the displacement rate  
- influence of adherend thickness  
- influence of different interfacial layers  
- influence of fibre orientation  
- influence of environmental exposure (preliminary results)

\( G_{2c} \): - influence of different interfacial layers  
- influence of fibre orientation  
- influence of environmental exposure

A survey of the different test series and number of specimens per test series is shown in table 1A and 1B.

The test results are presented as \( G_{1c} \) and \( G_{2c} \) values calculated with the following equations:

\[
G_{1c} = P_c \cdot Q \quad \text{(N/m)} \quad (1)
\]

\[
G_{2c} = \frac{P_c}{b \cdot h \cdot E} \quad \text{(N/m)} \quad (2)
\]

where \( P_c \): critical load (N)  
\( Q \): a constant for a certain specimen tapering and adherend stiffness  
\[
Q = 4 \cdot \frac{1}{(3a + h)} \quad (1/Nm) 
\]

\( E \): Young's modulus adherend  
\( a \): crack length  
\( b_a \): specimen width at the crack front boundary (fig 1)  
\( h \): adherend thickness

In chapter 4, the background of both equations is treated in more details and it will be examined whether or not a fracture mechanics approach is acceptable for the specimens used.

3.1: influence of displacement rate on \( G_{1c} \)

The influence of the displacement rate (and also the influence of
the adherend thickness, section 3.2) had to be examined before the more essential variables (influence of different interfacial layers, fibre orientation and environmental exposure) in order to investigate the sensitivity of the test method for parameters not directly related with the interfacial layer (prepreg or adhesive). However for the influence of displacement rate there can be some relation with the properties of the interfacial layer (e.g. visco-elasticity).

In the test series, an aramid prepreg interfacial layer (fibres oriented in the length direction of the specimen) is used for all displacement rates. The test results are shown in fig 5. There is a small but consistent $G_{1c}$ increase for an increasing displacement rate, which is in contradiction with the generally observed trend for adhesives (brittleness of adhesive increases with increasing deformation rate). However, as will be shown later on, the adhesive properties have only a limited effect if the adhesive is used in a prepreg configuration, especially in combination with aramid fibres. At this moment, an explanation for the observed trend between $G_{1c}$ and displacement rate cannot be given. Perhaps dynamic effects can cause a different release of stored energy per unit of crack length.

For all displacement rates the fracture surfaces are identical (the fracture surface and fracture path of an aramid prepreg are extensively discussed in section 3.3).

In the other test series, the displacement rates are restricted to two values:

$$\frac{dv}{dt} = 5 \text{ mm/h}$$
$$50 \text{ mm/h}$$

### 3.2: Influence of adherend thickness on $G_{1c}$

As will be shown in chapter 4, $G_{1c}$ can be written as:

$$G_{1c} = P_{c} \cdot Q$$  \hspace{1cm} (1)

where

$$Q = \frac{4}{3} \cdot \frac{3a^{3}}{E_b h} \cdot (3a^{2} + h^{3})$$  \hspace{1cm} (3)

$Q$ is a constant for a certain specimen geometry ($b_{a} = f(a)$) and adherend stiffness ($E, h$).

In ref 7, finite element studies were made to examine, among others, the influence of the adherend thickness on $Q$. The following approximation has been derived from the results:

$$Q = 1.4216 \cdot h^{-2.9433} \text{ (1/Nm)}$$  \hspace{1cm} (4)

where $E = 72500 \text{ N/mm}^2$

It is noteworthy that the exponent of $h$ is very close to the initially noted value ($Q = \frac{1}{h} \cdot f(a)$ for $h \ll 3a$).
Because $G_{lc}$ has to be considered as a material property (or a property of the interface), it should be independent of the specimen stiffness ($E,h$) which means that a variation of $Q$ should result in a corresponding variation of $P_c$ in such a way that the product remains constant, see eq. 1.

To check the independence of $G_{lc}$ for the adherend thickness $h$, two experiments are performed using different adherend thicknesses ($h=10$ and $8\,\text{mm}$). The interfacial layer properties were kept constant (in this case an aramid prepreg is chosen with fibres oriented in the length direction). The following data were obtained:

\[
\begin{align*}
  h = 10\,\text{mm} & \quad Q = 1.62 \times 10^{-3} \, (1/\text{Nm}) & \quad P_c = 836.6 \, \text{N} & \quad G_{lc} = 1134 \, \text{N/m} \\
  h = 8\,\text{mm} & \quad Q = 3.12 \times 10^{-3} \, (1/\text{Nm}) & \quad P_c = 607.0 \, \text{N} & \quad G_{lc} = 1151 \, \text{N/m}
\end{align*}
\]

In spite of the different $Q$ and $P_c$ values for the two thicknesses, the agreement of the experimental $G_{lc}$-values is highly satisfactory. Although eq. 4 seems to be very appropriate, it can only be used in a limited range of thicknesses (plastic deformation of the adherend will occur for small adherend thicknesses).

3.3: Influence of different interfacial layers on $G_{lc}$

In fig 6, $G_{lc}$ results of the adhesive are compared with the results of the aramid and carbon prepreg (for both prepregs, the fibres are oriented in the length direction of the specimen). It is striking that:

- Using an aramid prepreg, based on the same adhesive, strongly reduces the energy release rate; scatter between the four test results is small.
- Using a carbon prepreg, based on the same adhesive, results in higher energy release rates (compared with the aramid prepreg); differences between the four test data are much larger.

A realistic evaluation of the test results is impossible without examining the fracture surfaces of the specimens. Some typical fracture paths are shown in the figs. 7 to 13.

- For the pure adhesive specimens, fracture is occurring at the adhesive/carryer interface (fig 7) while in some cases the carrier is peeled out of the fracture surface. Microscopic examination of the fracture path shows a very yielded surface (fig 8) which is typical for this kind of epoxy adhesives and indicates extensive plastic deformation.
The fracture path of the aramid prepreg is very irregular (fig 9); many of the individual fibres are peeled out of the fracture surface. The fracture path is situated in the prepreg layer although very near to one of the adherends (due to the inhomogeneous distribution of the stresses in the thickness direction of the prepreg - peel stresses maximum near the adherend/prepreg interface). Microscopic examination of the individual fibres (scanning electron microscopy) showed a very smooth fibre surface (no adhesive visible on the fibre surface - fig 10). Considering the amount of individual fibres, peeled out of the surface, interface debonding seems to be the main fracture mechanism (because the fibres are uniformly distributed in the adhesive, there has to be some cohesive failure of the matrix too to enable crack growth).

For the carbon prepreg, a more regular fracture surface is present (fig 11). Microscopic examination of the fracture surface shows fibres fully covered with the adhesive (fig 12) which strongly points to cohesive failure in the matrix as the main fracture mechanism. In some cases, a rather brittle fracture of the fibre is visible (fig 13). These results are in good agreement with the literature (the interface bond strength for carbon fibres and a suitable matrix system is much higher than the cohesive strength of the matrix material).

For loading mode 1, the weakest link of the adhesive is the carrier/adhesive interface which however does not result in premature failure considering the very yielded fracture path. Although the carrier is somewhat decreasing the adhesive properties, it is indispensable for creating a controlled adhesive thickness.

The poor G\textsubscript{c} results obtained with the aramid prepreg are due to the low aramid/adhesive interface strength. This trend was recognized in earlier reports (e.g. ref 13) and also for other test methods (e.g. Bell-peel test, wedge-edge test) lower results were obtained. It is noteworthy that this poor interface strength is mainly a fibre problem (lack of chemical bonding possibilities) and not a problem of the adhesive (reactive unbonded epoxy groups are present at the interface). The adhesion properties of the different fibre types and epoxy system will be briefly discussed in chapter 4. It will be shown in chapter 5 that it is desirable not to aim at a maximum interface adhesion, because it is always accompanied by extreme brittleness, but for an optimum interface adhesion. Unfortunately this optimum can be different for different applications.

The weakest link of the carbon prepreg is the cohesive strength of the matrix which implies a totally different behaviour as compared to the aramid prepreg. Because of the interface strength of the carbon/adhesive combination no individual fibres are visible at the fracture surface which leads to the flat fracture path. The
$G_{ij}$ data of the carbon prepreg are somewhat lower than for the adhesive (although in both cases the fracture path is cohesive in the adhesive) due to:

- stress concentrations caused by the fibres
- fibres hamper full plastic deformation capacity of the adhesive

The large scatter between the carbon prepreg specimens (as compared to scatter for the aramid prepreg and the adhesive specimens) is also directly related to the fracture mechanism and especially to the initial load level necessary to create crack growth in the initial phase (see chapter 2.2). The initial load to create crack growth in the aramid prepreg is much higher than the load for crack propagation which means that at the onset of cracking a large amount of energy becomes available for crack growth. Because of the low interface strength of the aramid prepreg the available energy will create numerous cracks (there are numerous fibre/matrix interfaces and none of them will be dominant). The result is a very irregular crack front (random distribution of the fibres in the prepreg).

For the carbon prepreg there is mostly one dominant crack front, situated cohesively in the matrix. If however, for the carbon prepreg, a second crack front (the eventually of one or more crack fronts can, for the carbon prepreg, be found at the onset of cracking; it was recognized that more than one crack front can occur if the initial load to initiate crack growth is large) or other energy dissipating mechanisms are present, the influence on $G_{ij}$ is considerable. This concept was proved in our test series, the carbon prepreg specimen with the higher $G_{ij}$-value (fig 6) showed a more irregular crack front than the three other specimens.

Already in this stage a first general remark can be made (ignoring differences in fibre stiffness, stress distribution and so on):

energy to create interface debonding at the aramid/adhesive interface

is lower than

energy to create cohesive fracture in the matrix

which is lower than

energy to create interface debonding at the carbon/adhesive interface
3.4: Influence of fibre orientation on $G_{1c}$

For both prepregs (aramid and carbon), specimens with fibres in length and width direction were tested. The $G_{1c}$ data are compared in fig 14.

For the aramid prepreg, the influence of the fibre orientation is negligible and also the fracture surface is very similar. For both fibre orientations, numerous individual fibres are peeling out of the matrix and therefore a very irregular fracture path is obtained.

For the carbon prepreg, the influence of the fibre orientation is significant. The large difference can be explained by the essentially different fracture path and the energy which is necessary to create that fracture path. A crack growing perpendicular to the fibre direction will at one moment or another one encounter a fibre on its preferred fracture path. For carbon fibres with a high radial strength and a high interface strength this will imply that the crack is forced to leave its preferred fracture path (fig 15). At this new path again the presence of a new fibre will force the crack to leave it and the result will be a kind of step-wise fracture path (fig 16). Such a path requires significantly more fracture energy than a flat fracture path which is found if the fibres are oriented in the length direction of the specimen. As mentioned for the aramid prepreg, this $G_{1c}$-increase will only be present if the crack is forced to leave its preferred path which is not the case for the aramid prepreg. Here the interface strength is the weakest link and the crack will "jump" from one interface to another one (it is well possible that interface debonding does occur before the crack reaches the fibre). As a result, for prepregs with a poor interface strength the fibre orientation is much less important than for the prepregs with a high interface strength (this will be important if, in chapter 4, general fracture toughness is discussed).

3.5: Influence of environmental exposure on $G_{1c}$ (preliminary results)

Two different types of environmental tests were executed on the W.T.D.C.B. -specimens:

1: environmental exposure in distilled water at 90°C, followed by $G_{1c}$ testing in laboratory air.

   exposure periods: 55 days
   160 days

2: $G_{1c}$-test under environmental exposure in an environmental cell (fig. 4), but without pre-exposure.
environment: distilled water

temperature: 20°C
50°C

For both types of environmental tests, specimens with a carbon and aramid prepreg were examined (for both prepregs the fibres are oriented in the length direction of the specimen); for type 2, also the adhesive system was investigated. Although distilled water was used in both types of testing, the environmental exposure is totally different.

test of type 1: - during the pre-exposure, moisture can enter through the edges of the specimen only
  - water absorption is controlled by diffusion properties of the different prepreg layers
  - the moisture affected width of the specimen will be limited, because in the proposed exposure periods saturation of the specimen is not expected. Due to the tapered geometry, the influence of the affected width is decreasing for larger cracks (fig 18).
  - in the pre-exposure periods, corrosion processes on the adherends can be expected, especially for the carbon prepreg (galvanic corrosion!)

test of type 2: - moisture is present at the crack tip over the entire width of the specimen
  - effect of moisture diffusion through the edges is negligible due to the short testing period (about 5 hrs)
  - the crack growth rate is much higher than the diffusion rate of the moisture
  - in the short testing period (± 5hrs) no significant corrosion processes have to be expected.

The results of both types of testing will be discussed separately.

3.5.1: environmental exposure followed by $G_{1c}$-testing

In fig 17, the $G_{1c}$-results obtained after exposure periods of 55 and
160 days are shown and compared with results for unexposed specimens. It is noteworthy that the environmentally exposed specimens do not show a constant $G_{1c}$ value, but $G_{1c}$ is a function of the crack length. $G_{1c}$ is increasing with increasing crack length. This is a result of the tapered specimen geometry. Moisture has a deteriorating effect on both the carbon and aramid prepreg. Moisture can only enter along the edges of the specimen with a rate controlled by the temperature. Because of the tapered geometry, the deteriorated part of entire width of the fractured area is decreasing with increasing crack length (and increasing width of the specimen: $h_a = a/3$) which should result in an increasing $G_{1c}$-value (per unit crack length, the energy necessary for crack growth becomes larger because the unaffected part of the total surface is increasing - fig 18).

For the aramid prepreg, there is after 55 days of exposure a small increase of $G_{1c}$ after 160 days of exposure a strong decrease was found. The fractured surface of the aramid prepreg after environmental exposure is shown in fig 19. The presence of single fibres is more pronounced than for the unexposed specimens; no corrosion is visible at the adherend surface (aramid is an organic material which is electrically non-conductive). Microscopic examination of the single fibres shows a perfectly smooth fibre surface. It seems that moisture again decreases the already poor strength of the aramid/adhesive interface.

The decrease of $G_{1c}$ due to environmental exposure is dramatic for the carbon prepreg. For this system, the influence of the environment is a result of two contributing factors:

- severe galvanic corrosion of the aluminium alloy adherend
- moisture causes a change in the fracture mode from cohesive failure in the matrix to partially interface failure between carbon fibre and adhesive.

The fracture surface of the carbon prepreg is shown in fig 20. The influence of the corrosion process is somewhat decreasing with increasing crack length (due to the tapered geometry). It is well-known that aluminium and carbon form a galvanic cell which will, because of the large potential difference, lead to rapid corrosion of the aluminium part. Most probably as a result of the galvanic corrosion process, the fracture path is much closer to the adherend and more single fibres are present at the fracture surface. Microscopic examination of these fibres shows a more interface dominated failure (fibre/adhesive interface) which was fully absent for the non-exposed specimens. The corroded area at the edges of the specimen facilitates the moisture to enter the specimen after which the moisture creates the galvanic cell. This deteriorating process seems to propagate at a very high rate and results in very poor $G_{1c}$-data.
3.5.2: Combined $G_{IC}$-test and environmental exposure

The results of a $G_{IC}$-test, performed in a distilled water environment (2 temperature levels) are shown in fig 21 and compared with results of unexposed specimens. As for the first type of environmental testing, $G_{IC}$ is mostly increasing with increasing crack length (except for one case - carbon prepreg - where a decrease of $G_{IC}$ with increasing crack length is observed) but the explanation used for the first type of testing cannot be applied in this case (no edge-controlled deterioration process). For the adhesive, the $G_{IC}$-increase with crack length is large, especially at the highest test temperature. The fracture surface of the adhesive (fig 22) points to a few possible reasons for the $G_{IC}$-increase during crack growth:

- Increased plastic deformation due to the distilled water exposure. However this feature should lead to a constant increase of $G_{IC}$, and not to a linear increase with crack length (tapered specimen geometry; area available for plastic deformation is proportional to the specimen width)

Only if time-dependent properties are present (e.g. per unit width increasing plasticity with increasing crack length or decreasing crack growth rate with increasing crack length) plasticity effects can cause an increasing $G_{IC}$-value. However this time-dependency was not encountered. In fig. 21B it is shown that the crack growth rate was even increasing with increasing crack length.

- Increasing irregularity of the fracture surface with increasing crack length. With increasing crack length, the number of irregularities in the fracture surface (zones B and C in fig 22B) are increasing but not more than proportional to the specimen width. Also such irregular crack fronts (fracture at adherend/adhesive interface on localised spots) should create a sudden increase of $P_c$ (and $G_{IC}$) but by no means a kind of continuous increase as it is encountered in the experiments.
Up to now, a covering explanation for the continuously increasing $G_{IC}$-value with increasing crack length has not been found. Since the presented results are only preliminary, further experiments should solve this problem.

The fracture surface of the adhesive after environmental testing is shown in fig 22. Three typical areas, corresponding to two different fracture modes can be observed.

**area A:** cohesive fracture in the adhesive near carrier interface - fig 23
For the 20°C exposure, the surface is identical with the unexposed fracture surface (yielded character); for the 50°C exposure, a discolouring of the adhesive is visible which can point to an activated chemical process. The yielding for this test temperature is even more pronounced than for the 20°C, distilled water exposure.

**area B (adhesive side - fig 24) C (aluminium side - fig 25)**  
interfacial fracture at the adhesive/aluminium interface

This fracture mode, absent for the unexposed specimens, is characterized by a very brittle fracture surface and will ask significantly less energy for crack growth than cohesive fracture in the adhesive. If this fracture mode occurs only at small localized spots, the influence on the total required energy for crack growth will be small. Moreover, a staggered fracture path is introduced which can even improve $G_{IC}$ as observed at 20°C. However, for a 50°C, distilled water exposure, this fracture mode occurred much more abundantly which results in lower $G_{IC}$-values. An improved pretreatment of the adherends (specimens were only degreased and pickled) will most probably prevent this fracture mode.

For the prepregs, the presence of moisture at the crack tip results in a somewhat lower $G_{IC}$-level and a temperature effect is present (increased deterioration with increasing temperature). The increase in $G_{IC}$ with increasing crack length is less pronounced than for the adhesive which is understandable considering the hampered plastic deformation capability of the matrix (presence of the fibres in matrix) and the absence of partial adherend/matrix failure (irregular fracture surface). In fig 26, a microscopic view of an aramid fibre surface is shown after testing. The fibre surface is completely smooth which points to a very low interfacial strength. A carbon fibre surface after 50°C, distilled water exposure is shown in fig 27. It is clear that for this exposure condition, also the carbon prepreg has an interface dominated fracture (carbon fibre/matrix interface). This change in fracture mode results in much lower $G_{IC}$-values, as shown by the results in fig 21.
3.6: Influence of different interfacial layers and fibre orientation on $G_{2c}$

The thick adherend specimen, described in chapter 2, is used to examine the energy release rate in the shear fracture mode. The theoretical background of the specimen and the test method is presented in chapter 4. As will be discussed in chapter 4, a fracture mechanics approach for the shear fracture mode is at least debatable. Therefore, besides $G_{2c}$, also the average shear stress at fracture is noted in the figures and tables.

In this test series a glass prepreg is used besides the adhesive, aramid and carbon prepreg. The results of the experiments are shown in table 2 and fig 28.

The adhesive shows the highest $G_{2c}$-value. The fracture surface of the adhesive is shown in fig 29. Fracture occurs mainly at the carrier/adhesive interface although on some localised places adhesive/adherend interface fracture is present. The latter fracture type is, as for the $G_{1c}$-test, extremely brittle (fig 30); fracture at the carrier/adhesive interface has a typical yielded character.

The prepregs have a lower $G_{2c}$-value and, in agreement with the $G_{1c}$-testing, the aramid prepreg shows significantly poorer results than the carbon prepreg. The difference is less pronounced if the average shear stress ($\bar{\tau}$) is considered. Using this parameter, the carbon prepreg is even equivalent to the adhesive. One should however notice that in the test series two different adherend thicknesses have been used which cannot be represented in the average shear stress equation. In chapter 4, the advantages and disadvantages of both parameters ($G_{2c}$ or $\bar{\tau}$) are discussed.

The prepregs also show a considerable fibre orientation effect. In all cases, fibre orientation in the width direction of the specimen results in poorer $G_{2c}$ and $\bar{\tau}$ data (the opposite trend was obtained for the $G_{1c}$-test). For the T.A.-specimen, the lower results for specimens with fibres in the width direction can be explained by the more pronounced stress concentration effect of the fibres and the absence of stable crack growth for this specimen.

The fracture surface of the aramid prepreg (both for fibres in length and width direction) for the shear fracture mode is very similar to the one obtained in the $G_{1c}$-test (fig 31). The fracture surface is very irregular and a microscopic examination of the fibre shows a very smooth fibre surface which points to an interface dominated fracture.

The fracture surface of the carbon prepreg is very smooth (fig 32); no individual fibres are visible at the fractured surface.
For the glass prepreg, there is a significant difference in fracture surface between the two fibre orientations. Specimens with fibres in the length direction show a rather smooth fracture surface with only some individual fibres peeled out of the surface (fig 33A); specimens with fibres in the width direction have a much more deteriorated surface (fig 33B), comparable with an aramid prepreg surface. As shown in table 2, this difference in fracture behaviour is also represented in the $G_{2c}$-data.

3.7: Influence of environmental exposure on $G_{2c}$-

The influence of environmental exposure on an aramid prepreg layer (fibres oriented in width direction) is shown in fig 34. The influence of a one month saltspray exposure period is limited; a one month distilled water exposure (60°C) results in a small decrease of the critical shear stress. These first results confirm literature data (the deteriorative effect of distilled water exceeds the one of saltspray exposure if adhesives or prepregs are concerned; the opposite relation is present if metals are examined).

The fracture surface of the specimens after environmental exposure still shows a fibre/matrix interface dominated failure.
4: Theoretical aspects

In this chapter, the theoretical background is presented for both the \( G_{1c} \) and \( G_{2c} \) test method. It is examined whether or not the proposed specimen configurations (W.T.D.C.B. and T.A.-specimen) are useful for a fracture mechanics approach. Further it was demonstrated in chapter 3 that adhesion is an important parameter for both \( G_{1c} \) and \( G_{2c} \) and that interface strength of the different fibre types (glass, aramid, carbon) with epoxy systems largely differ. Therefore a brief review is made of the possible interfacial bonds between the different fibres and an epoxy matrix. In chapter 5, an attempt will be made to relate the experimental results with the general fracture toughness behaviour of the prepregs and more generally of composite systems. In that respect and starting from the energy balance approach of isotropic materials, an extension of this approach is made towards anisotropic materials.

4.1: \( G_{1c} \)-test method (W.T.D.C.B.-specimen) - theoretical background

In literature most attention is paid to mode 1 fracture (tension mode) because of the sensitivity of most adhesives for this loading mode. In the present test series this loading mode is examined by testing a double cantilever beam specimen (fig 1) consisting of two aluminium adherends bonded together by an adhesive or a prepreg layer.

For this specimen, the following relation can be found:

\[
G_{1c} = \frac{\delta F - \delta U_a}{\delta A}
\]  \( (5) \)

where: \( G_{1c} \): energy release rate  
\( F \): external work  
\( U_a \): elastic energy  
\( A \): fractured area

For a specimen with a variable width (e.g. the width tapered double cantilever beam specimen of fig 1) one gets:

\[
G_{1c} = \frac{1}{b} \frac{\delta F - \delta U_a}{\delta a}
\]  \( (6) \)

where \( b_a \): width of the specimen at crack length \( a \), see fig 1
For a constant load condition (external load remains constant during crack growth which implies that external work will be done):

\[ \frac{\delta F}{\delta a} = P \cdot \frac{\delta v}{\delta a} \]  (area ABCD, see sketch)

\[ \frac{\delta u}{\delta a} = \frac{1}{2} P \cdot (v + \frac{\delta v}{\delta a}) - \frac{1}{2} P v \]

area (OBD) = area (OAC)

\[ = \frac{1}{2} P \cdot \frac{\delta v}{\delta a} \]

Substitution in eq. (6) results in:

\[ G_{1c} = \frac{1}{2b_a} \cdot P \cdot \frac{\delta v}{\delta a} \]  (7)

The compliance \( C \), can be defined as:

\[ C = \frac{v}{P} \]

If the compliance is introduced in eq. (7), a commonly used equation is obtained:

\[ G_{1c} = \frac{1}{2b_a} \cdot P^2 \cdot \frac{\delta C}{\delta a} \]  (8)

It is shown in literature (e.g. ref. (5)) that a fixed grip condition (displacement \( v \) is constant, no supply of external work, external load drops with increasing crack length) and all the intermediate conditions result in the same equation.

Several authors (e.g. ref. (6)) calculated \( \frac{\delta C}{\delta a} \) of a width tapered double cantilever beam specimen which in most cases resulted in:

\[ \frac{\delta C}{\delta a} = \frac{8}{E_b a} \cdot \left( \frac{3a^2}{h^3} + \frac{1}{h} \right) \]  (9)

\( h \): adherend thickness

\( E \): Young's modulus of adherend

where the first part between brackets corresponds to the contribution of bending deformation and the second part to shear deformation.
It can be easily demonstrated that the contribution due to bending in eq. (9) results from a straightforward use of the slender beam theory with a clamped condition at \( x = a \) (fig 35):

\[
M = P x = EI \frac{d^2w}{dx^2} \\
= E b x h^3 \cdot \frac{d^2w}{dx^2} \\
= E q x h^3 \cdot \frac{d^2w}{dx^2}
\]

with \( q = \frac{b x}{x} = \text{constant} \)

\[
\frac{d^2w}{dx^2} = \frac{12P}{E q h^3}
\]

Integration and application of the boundary conditions

\( x = a \rightarrow w = 0 \) and \( \frac{dw}{dx} = 0 \)

lead to:

\[
w = \frac{12P}{E q h^3} \cdot \left( \frac{x^3}{2} - ax + \frac{a^3}{2} \right)
\]

For the displacement of \( P \) at \( x = 0 \), the result is:

\[
(v)_{x=0} = 2 (w)_{x=0} = \frac{12Pa^3}{E q h^3}
\]

\[
C = \frac{v}{F} = \frac{12a^3}{E q h^3}
\]

\[
\frac{dC}{da} = \frac{24a}{E q h^3}
\]

and with \( q = \frac{b x}{a} \)

\[
\frac{dC}{da} = \frac{24a^2}{E b a^3} = \frac{9}{E b a} \left( 3a^2 / h^3 \right)
\]

which is equal to the first term of eq. (9).
The contribution of the shear deformation can also easily be found:

\[ dw = \gamma da \]

\[ = \frac{T}{G} da \]

\[ = \frac{\alpha P}{bhG} da \]

\[ \frac{dw}{da} = \frac{\alpha P}{bhG} \]

\( \alpha \) is a function of the cross-section of the beam; for a rectangular section \( \alpha = 1.5 \) (ref. 29)

\[ \frac{dv}{da} = 2 \frac{dw}{da} = \frac{8P}{bhE} \]

where \( G = E/(2(1+\nu)) \) and \( \nu = 1/3 \)

\[ \frac{dc}{da} = \frac{1}{P} \cdot \frac{dv}{da} = \frac{8}{bhE} \]

which is equal to the second term of eq. (9).

A totally clamped condition is not realistic for the W.T.D.C.B.-specimen in view of the layer between the two adherends. As a correction for rotation of the beam at the crack tip, some authors add an apparent extra crack length increment \( a_\alpha \).

An often used approximation for \( a_\alpha \) is:

\[ a_\alpha \approx 0.6h \]

Equation (9) then becomes:

\[ G_{1c} = \frac{4P^2}{Eh^3} \cdot \left( 3(a + 0.6h)^2 + h^2 \right) \]  

(10)

\[ = P^2 \cdot \mathcal{Q} \]

As shown by eq. (10), \( \mathcal{Q} \) is a function of \( a \). It should be realized that \( b \) in this equation is also a function of \( a \), viz.: \[ b = qa \]

Substitution in eq. (10) and assuming \( a_\alpha < a \) leads to:
\[ Q = \frac{4}{Ea^2h^3} \cdot (3 + (h/a)^2) \] (11)

For \( a/h > 10 \) (applicable to the present test series), the latter equation implies that \( Q \) is practically constant. Consequently a constant \( G_{1c} \) during a WTDCB test implies that \( P \) will remain constant.

4.2: \( G_{2c} \)-test method (T.A.-specimen) - theoretical background

Experimental methods to obtain a \( G_{2c} \)-value for interfacial layers (prepreg or adhesive) are far less common than for \( G_{1c} \). Ripling and Mostovoy (ref. 8,9) suggested a similar approach for \( G_{2c} \) as for \( G_{1c} \) using a kind of thick adherend specimen (fig 36). In this case the relation corresponding to eq. (8) is:

\[ G_{2c} = \frac{P^2}{2b} \cdot \frac{dC}{da} \] (12)

It is assumed by the authors that the shear stresses have a maximum near one end of the joint (fig 36) and that crack initiation should occur at that point. Further it is assumed that \( \frac{dc}{da} \) for this type of specimen can be derived from the elastic properties of the adherends:

\[ C = \frac{2a}{bhE} \] (13)

\[ \frac{dc}{da} = \frac{2}{bhE} = \text{constant} \]

Substitution in eq. (12) gives:

\[ G_{2c} = \frac{P^2}{b^2hE} \] (14)

Both authors encountered numerous problems in achieving a kind of controlled crack growth in a shear loading mode:

1) starting from an initiated crack (initiation executed in tension mode!), it was hardly possible to measure crack growth. At the critical load level, rapid fracture did occur and resulted immediately in complete failure.

2) in spite of carefully arranged loading conditions (loading in plane of interfacial layer), fracture surface examinations showed that crack propagation from the mode 1 precrack did not occur in shear but was still controlled by mode 1 stresses.
Hartman (ref. 10) used a height tapered double cantilever beam specimen to examine $G_{2C}$. Using the same assumptions as Ripling and Mostovoy, his results were not encouraging concerning the use of fracture mechanics for this specimen; especially the statement that $dC/da$ is independent of the crack length was not confirmed in his experiments.

Considering the unfavourable results obtained in literature, in our test series a simplified specimen (compared to fig 36) is used namely the thick adherend specimen which of course implies that for this specimen a fracture mechanics approach is even more debatable. Compared with the specimen of Ripling and Mostovoy (fig 36), two main differences are notable:

1: absence of an initial crack: from ref 8,9, it was clear that an initial crack does not result in crack propagation in a shear mode; because of the limited overlap length (see point 2) and the adherend stiffness, it can be practically excluded to produce an initial crack by mode 1 loading. For these two reasons, no attempt was made to create an initial crack.

2: limited overlap length: compared with fig 36, the overlap length of a T.A.-specimen (fig 2) is very small (10 mm). The crack growth possibility is therefore very limited for the T.A.-specimen. The difference in overlap length has also a significant influence on the shear stress distribution in the overlap; a decrease in overlap length results in a more uniform shear stress distribution although still shear stress peaks will be present at the overlap ends (fig 37). For the T.A.-specimen, the released energy during crack growth will be high along the total overlap length which also makes a kind of stable crack growth unexpected.

In conclusion, it is not expected (and confirmed in the test series) that the T.A.-specimen is useful for a fracture mechanics approach because of the absence of a growing crack in a shear mode. Although the use of an average shear stress as a parameter indicating the sensitivity for delamination growth instead of $G_{2C}$ is also debatable (shear stress peaks at overlap ends!) it is most probably a more appropriate parameter. However, in the equation used for this parameter

$$\bar{\tau} = \frac{P}{cr number\_of_page}$$

where $l$: overlap length

$b$: width of specimen

adherend properties are not included (e.g. $E,h$), although it is
expected that these properties will have a significant effect on the shear stress distribution and therefore on the measured average stress. Comparisons between the different interfacial layers should be performed with identical adherend properties. In our test series the adherend thickness was 6.38 mm for all specimens except for the adhesive bonded specimens without fibres (h=9.06 mm)

4.3 Adhesion between different fibre types (glass, carbon, aramid) and an epoxy system

The importance of good adhesion between fibres and the matrix system was clearly shown in chapter 3. It will be discussed more generally in chapter 5. In the present section adhesion will be surveyed in terms of physical and chemical bonds. It is noteworthy that on a chemical and physical level, there is no difference between interface and cohesive bonds. Both types of bonds consist of a certain combination of covalent and/or electrostatic bonds. In both conditions (interface or cohesion), the strength of the bonds will be a function of:

- the type of bond
- the number of bonds

The survey given below will be rather brief and it certainly will not cover all aspects. Before discussing the interface properties of the fibre/matrix systems, a survey of the different possible bonds is given. Such a survey is necessary to determine which bonds should be aimed at to create a suitable interface bond.

4.3.1 Type of bonds

In general, two types of bonds should be considered (ref. 23):

- electrostatic bonds: these bonds are related to the interaction of atoms of an electronic structure which is essentially independent of the proximity of other atoms around it

- covalent bonds: for covalent bonds, the electrons between the atoms are shared and the electronic structure of the atoms depends on the proximity of interacting atoms (the larger the number of electrons shared between the atoms, the stronger the bond and the smaller the interatomic distance)
Hydrogen bonds (e.g. ref. 24, 25, 26), often encountered when interfaces are considered, have a partially electrostatic character (dipoles formed by chemical bonds of hydrogen to more electronegative polyvalent atoms such as N, F, O) and a partially covalent character. In general one may state:

\[
\text{electrostatic bond strength} \quad < \quad \text{hydrogen bond strength} \quad < \quad \text{covalent bond strength}
\]

Both the electrostatic and covalent bonds can be subdivided in different groups. Here, only the more relevant ones will be briefly treated. The bonds are presented in order of decreasing bond strength.

a) electrostatic bonds

- ionic bond: a positive and a negative ion attract each other, each ion acting as a nucleus surrounded by a rigid spherical distribution of electrons

\[
\text{Interaction energy: } U = \frac{q^+ q^-}{4\pi \varepsilon_0 r}
\]

where \( q^+ \), \( q^- \) : charge of the ions
\( r \): distance between the ions
\( \varepsilon_0 \): dielectric constant

- dipole-dipole bonds: the dipole-dipole force can be described by the electric dipole moment \( \mu \)

\[
\mu = q l
\]

the origin of the dipole-dipole forces is shown in the figure below

![Diagram showing dipole-dipole forces](image)
Interaction energy:

\[ U = \frac{\mu_1 \mu_2}{r^2} \cdot (2 \cos \theta_1 \cos \theta_2 - \sin \theta_1 \sin \theta_2 \cos (\phi_1 - \phi_2)) \]

(fixed dipole orientation)

\[ U = -2 \frac{\mu_1 \mu_2}{r^3} \]  
(fixed dipole orientation; head to tail configuration)

\[ U = -\frac{2 \mu_1^2 \mu_2^2}{3kT r^6} \]  
(free rotation dipoles)

where \( \mu_i \): electric dipole moments

\( r \): distance between two dipoles

\( kT \): average thermal energy

- dipole induced dipole bonds: in an electric field, an originally neutral atom or symmetric molecule can be polarized because of an attraction in opposite direction of the electron cloud and nucleus. The electric field necessary to induce the dipole can be produced by a neighboring dipole.

Interaction energy:

\[ U = -\frac{\mu_1^2 \alpha_2}{r^6} \]  
(dipolar + nonpolar molecule; ref 27)

\[ U = -\frac{\mu_1^2 \alpha_2 + \mu_2^2 \alpha_1}{r^6} \]  
(2 permanent dipoles)

where \( \mu_i \): dipole moments

\( \alpha_i \): polarizabilities

- London dispersion bonds: any instant in time, one can find a particular configuration of nuclei and electrons that presents an instantaneous dipole moment. This produces an electric field which interacts with the polarizability of an adjacent molecule.
and results in an instantaneously induced dipole and mutual interaction to provide the dispersion force. The London dispersion force is then called the instantaneous force of attraction averaged over all instantaneous configurations of the electrons in the molecule. It is noteworthy that the London bond is the only bond possible between two nonpolar atoms (ref 28).

Interaction energy:

\[ U = \frac{3}{4} \frac{\alpha_1 \alpha_2}{r^6} \cdot \frac{2C_1 C_2}{C_1^+ C_2^-} \]

where \( \alpha \): polarizability
\( r \): distance between two dipoles
\( C_1^+ \): ionization energy

b) covalent bonds

The different covalent bonds (triple, double, single) describe the number of involved electrons (resp. six, four, two). Covalent bonds are formed by the shearing of unpaired outer orbital electrons between atoms (Pairs of electrons with opposing spins which occupy the same orbital are not capable of bonding).

Two types of molecular orbitals result from covalent interaction of simple s, p orbitals or their sp- hybrids:

- sigma bonds: are obtained by molecular bonding interactions of s-orbitals with p-orbitals or sp-hybrid orbitals. In this case the bonding electrons are centered between the two nuclei.

- pi-bonds: are the result of molecular interactions of p-type orbitals. The interaction may occur through lateral overlapping of the individual lobes oriented into an equivalent plane of rotation.

A single covalent bond (e.g. C–C) is formed by a sigma bond

A double covalent bond (e.g. C=C) is formed by a sigma and a pi bond

A triple covalent bond (e.g. C≡C) is formed by a sigma and two pi bonds

Resonance effects (e.g. benzene C₆H₆) can cause intermediate properties between single and double covalent bonds.
4.3.2: Fibre/matrix adhesion

Good adhesion obviously requires molecular groups in both adjacent structures which can react with each other. Because both fibres and matrix consist of macromolecules built up from long chains with reactive end groups, these end groups can react with each other during the curing process. These chemical bonds will, however, not result in a strong chemical adhesion (number of bonds is very limited). Mostly by etching (carbon fibres), the number of active groups on the fibre surface can be increased. If these groups are unable to react chemically with the matrix groups, coupling agents can be used with adequate reactive groups to both sides of the interface. As mentioned before, the stronger the interaction energy of the chemical bond, the better the adhesion (if a sufficient number of bonds is involved). It is therefore not surprising that, considering the order of possible bonds in chapter 4.3.1, for a good adhesion, covalent, hydrogen or dipole bonds are aimed at.

Except for chemical bonds, mechanical joining can give a favourable contribution to the adhesion if a certain roughness of the fibre surface is present and if the wetting of the matrix material on the fibre is suitable during curing. It is noteworthy that the mechanical locking can even be present after interface debonding.

The three examined fibre/matrix combinations will now be shortly discussed.

A: carbon/epoxy adhesion

The epoxy group can be presented as a very reactive three membered ring consisting of two carbon and one oxygen atom:

\[
\begin{array}{c}
\text{\text{O}} \\
\text{\text{C}} \\
\text{\text{C}}
\end{array}
\]

The epoxy polymer has the following structure for its repeating unit:

\[
\left[ \begin{array}{ccc}
\text{CH}_3 & \text{C} & \text{OH} \\
\text{OH} & \text{OCH}_2 & \text{CHCH}_2\text{O} \\
\text{CH}_3 & \text{CH}_3 & \text{CH}_2\text{Cl}
\end{array} \right]
\]

The present unreacted secondary hydroxyl groups (OH\(^{-}\)) serve as reactive centers for cross-linking (or adhesion). The polymer is formed from epichlorohydrine and bisphenol - A

\[
\begin{array}{c}
\text{CH}_3 \\
\text{OH} \\
\text{CH}_3
\end{array}
\]

\[
\begin{array}{c}
\text{HO} \\
\text{CH}_3 \\
\text{CH}_2\text{C}1
\end{array}
\]

bisphenol-A  epichlorohydrine
The basis material for the production of carbon fibres is mostly PAN (polyacrylonitrile):

After some heating stages, the two-dimensional network of the carbon fibre is obtained. The process is mostly followed by an oxidation process of the fibre surface to improve the adhesion properties.

The surface reactivity of carbon fibres is low because of the parallel orientation of the graphite planes with the fibre surface. The C-atoms in the six-ring structure are strongly coupled by covalent bonds; only the boundary C-atoms can be reactive and functional groups can be added to these boundary atoms.

In general, three methods do exist to improve the carbon fibre/matrix adhesion:

1: whiskerisation of the fibre surface: it is possible to create mono-crystalline whiskers on the fibre surface, growing perpendicular to the surface. This method improves the mechanical joining and results in an improved interfacial shear strength.

2: oxidation of the fibre surface: functional groups of the matrix system can be coupled with the O-atoms

3: use of a coupling agent: a polymer can be added to the fibre surface with both coupling possibilities towards the fibre and the matrix. Mostly a dipole bond is created between the polymer and the fibre. To enable this bond, the carbon structure is supplied with a suitable reactive group (e.g. carboxylic group)

\[ -COOH \]

Because of the addition of functional groups to the fibre structure it is well possible, very strong interfacial bonds can be created with the reactive functional groups of the epoxy matrix.
B) Glass/epoxy adhesion

The chemical structure of glass is based on that of silica (SiO₂) with addition of oxides, calcium, boron, sodium, iron and other elements. The structure of glass shows no long range ordering at a molecular level but consists of a three dimensional array of atoms.

Glass fibres have the following reactive groups at the fibre surface:

- Si-end atoms which have OH-groups
- alkali metals which have OH-groups

Although interfacial bonding of the end atoms with the OH-groups of the epoxy is well possible, mostly coupling agents are used to further improve the adhesion. Silane coupling is often used and consists of:

\[ Y - R - Si - X_n \]

- The X-group is hydrolysed and in water containing solvents one gets:

\[ Y - R - Si - X_n \xrightarrow{nH_2O} Y - R - Si - (OH)_n + nHX \]

The OH-group is bonded with glass surface.

- The Y-group can be chosen in relation to the matrix structure and can form strong covalent bonds with the epoxy matrix.

The silane coupling agent has also a water repulsive function (in the presence of water, the OH-groups could form chemical bonds with H₂O).

As for the carbon fibres, the presence of reactive functional groups (added or not) at the fibre surface results in a very strong interfacial bond strength.

C) Aramid/epoxy adhesion

The aromatic polyamides (aramid) are manufactured in a wet spinning process and consist of sequential segments of phenylene and amide (located at the para - carbons):

- \[
\text{phenylene} \quad \text{amide}
\]
This sequential arrangement results in rigid chains like poly-p-phenylene-terephthalamide (PpPTA) (=chain structure actually used in the production of both KEVLAR and TWARON fibres).

The formation of the chain is governed by competitive intramolecular interactions between the conjugated groups in the chain. Regularly positioned amide segments allow for medium strong intermolecular hydrogen (O---H) bonds; adjacent hydrogen-bonded planes interact predominantly by means of van der Waals forces (dipole-induced dipole, dipole-dipole and London dispersion forces) with some pi-bond overlap of the phenylene segments (ref. 30).

The one-dimensional structure of the chain and the already weak bonds between adjacent chains and planes points the the absence of reactive functional groups in the chain structure. The addition of functional groups to the fibre surface or the use of coupling agents is extremely difficult. One method to improve interfacial adhesion is introduced by Allred and consists of getting amine groups (R-NH₂) on the fibre surface by a treatment with amine containing low temperature plasmas. The amine groups provide reactive sites for covalent bonds with the epoxy system (ref. 14). Although this method proved to be successful, the accompanying costs are so excessive that it is not yet used on an industrial scale.

4.4: Extension of the energy balance approach for anisotropic materials

4.4.1: energy balance for isotropic homogeneous materials

Griffith (ref. 15) stated, that, in the absence of external work, a crack will propagate if the incremental loss of strain energy with increasing fracture area, just equals the work required to create a new fracture surface:

\[
\frac{\delta U_a}{\delta a} \geq \frac{\delta U_Y}{\delta a}
\]

(15)

where \( U_a \): elastic energy
\( a \)

\( U_Y \): surface energy
\( a \): crack length
For an infinite plate under a tensile stress $\sigma$, Griffith derived:

$$U_a = \frac{m\sigma'a^2}{E}$$

Further: $U_\gamma = 2(2\alpha a)$

The left part of equation (15) is the elastic energy which is released during crack growth, the right part is the energy which is necessary to create further crack growth. Unstable crack growth will occur if the released energy available for crack growth is larger than the necessary energy to create further crack growth. It is important to recognize that $\delta U_a / \delta a$ is depending on the stress-strain (or energy) distribution in the system whereas $\delta U_\gamma / \delta a$ for isotropic, homogeneous materials, is a constant without any relation to the energy distribution in the system. This difference is important if the crack growth direction is of interest. In an isotropic material, the energy to disrupt bonds is constant for all crack extension directions. Still, a crack will grow in one preferred direction. The direction of crack growth is therefore controlled by $\delta U_a / \delta a$. In general, the crack will grow in that direction where the strain energy release rate for the system is maximum (It should be mentioned here that the maximum strain energy release rate criterion is one of the available criteria to predict the direction of crack growth. In ref. (16) and (17), a review of different criteria is presented).

Irwin and Orowan (ref. 18, 19) extended Griffith's theorem to the presence of external work and plasticity at the crack tip. The necessary energy for crack growth now consists of the earlier defined energy to disrupt bonds ($U_\gamma$) and a part which stands for the energy dissipation due to plastic deformation.

$$\frac{\delta(F - U_a)}{\delta a} \geq \frac{\delta(U_\gamma + U_\psi)}{\delta a}$$

(16)

where $F$: external work

$U_\psi$: dissipated energy due to plastic deformation

In the right part, $\delta U_\psi / \delta a$ is not only a function of the elasto-plastic response of the material, but it also depends on the distribution of the plastic deformation around the crack tip and can therefore not be regarded as a material property.

In the presence of external work, the available energy is partially used to increase the elastic strain energy of the system. One
can prove that the increase in elastic strain energy in the presence of external work just equals the decrease in elastic strain energy in the absence of external work. As a result, the energy available for crack extension is the same. For an actual loading condition of a structural part it is not necessary to have either fixed grip or fixed load conditions: Both the load and the displacement may vary. Still, the available energy for crack growth is equal to the available energy for both the fixed grip and fixed load condition.

Returning to eq. (15), one can define:

\[
G = \frac{\delta (F - U_a)}{\delta a}
\]

\[
R = \frac{\delta (U_\gamma + U_\psi)}{\delta a}
\]

\(G\) is called the crack driving force (it stands for the energy available for crack growth); \(R\) is called the crack resistance (energy necessary to create crack growth). Equation (16) reduces to:

\[
G \geq R
\]  

(17)

4.4.2: Extension of the energy balance towards anisotropic materials

(fibre reinforced materials)

The presence of fibres in an adhesive layer (e.g. prepreg as used in our experiments) or in matrix systems (composites) complicates the usefulness of an energy balance approach because of:

- the occurrence of interfacial failure
- very inhomogeneous distribution of the elastic strain energy density
- existence of several energy dissipating fracture mechanisms
- complicated evaluation of the plastic energy dissipation
- possible presence of energy "barriers" for the activation of fracture mechanisms

The above mentioned points will be shortly discussed in this chapter.

- appearance of interfacial failure: in the energy balance for isotropic materials, the right part of the equation (15) was a material
constant ($\gamma_f$: surface tension). In the case of interfacial failure, $\gamma_f$ is not only a function of the two adjacent materials and their chemical and physical reaction possibilities (see chapter 4.3), but it is also strongly depending on the surface treatments of these materials. If interfacial fracture is present, $\gamma_f$ cannot be regarded as a material property but as a zone-property. The zone represents a small area of both adjacent materials near the interface (it has to be considered that the physical, chemical and mechanical properties of the material near the interface can differ largely from the bulk material: e.g. increased cross-linking of an adhesive near an interface).

- Inhomogeneous distribution of the elastic strain energy density: Because of the presence of highly different materials in the same system, the energy density distribution in the material will be very inhomogeneous. For example, the energy density in the fibres will be much larger than in the resin system (e.g. fig. 38 where a typical fibre (aramid) is compared with a typical adhesive (epoxy) under tensile loading). The inhomogeneity will be amplified by the presence of a stress concentration at a crack tip where a considerably higher energy density exists.

- Existence of several independent energy dissipating fracture mechanisms: For homogeneous elastic materials, the energy balance can easily be defined. In general only one principal crack is present and there is one energy level required for crack growth ($\gamma_f$ is a constant for the system). Fibre reinforced materials can show a variety of fracture mechanisms with different required energy levels to cause crack growth and, because of the complex distribution of the energy density, different elastic energy release rates. Every mechanism will have its own energy balance and a prediction of which mechanism (or mechanisms) will occur is often very difficult. Aronsson (ref. 11) defined the following fracture modes which may occur in fibre-reinforced materials (fig. 39):

- fibre/matrix interfacial failure
- fibre pull-out
- matrix micro-cracking
- ductile fibre failure
- fibre fracture at a weak flaw matrix crazing
- longitudinal matrix failure

Based on the work of Beaumont (ref. 12) and Aronsson (ref. 11) some possible energy dissipating mechanisms will be briefly discussed. Both authors mention equations for the different energy dissipating mechanisms. As some of these equations seem to be rather
speculative, in this survey only the major influencing parameters are noted for every mechanism. It should be recognized that all these mechanisms need not necessarily occur. For example, crack bridging by fibres will occur only if the failure strain of the fibre is considerably larger than the failure strain of the resin. For the latest generation of adhesives in combination with carbon fibres this is by no means the case.

energy absorbing mechanisms (according ref. 11 and 12)

- frictional shear force between fibre and resin: This energy absorbing mechanism can only occur if the failure strain of the matrix is considerably lower than the failure strain of the fibre. Energy dissipation is a result of differential displacements of matrix and fibre. The mechanism is only present after interfacial debonding at the fibre/matrix interface (fibre/matrix interaction results from mechanical keying).

Both authors from ref. 11 and 12 present a similar equation for the friction work done per fibre:

\[ W_F = d, l^2, \tau \]

where
- \( W_F \): post-bond friction work (per fibre)
- \( d \): diameter of fibre
- \( l \): debonded length of fibre
- \( \tau \): interfacial friction stress

- fibre debonding energy: This energy dissipating mechanism is also assumed to occur only if the strain to failure of the fibre is considerably larger than the strain to failure of the matrix. Fibre/matrix debonding will occur after the matrix crack has passed the fibre or in front of the matrix crack.
In all models (e.g. Beaumont -ref. 12; Outwater/Murphy -ref. 21), the following parameters are present:

\[ W_{FD} \div E_f^{-1}, d, l \]

where \( W_{FD} \): debonding work done per fibre
\( l \): fibre length
\( d \): fibre diameter
\( E_f \): Young's modulus fibre

One should expect for interface debonding that the surface tension (\( \gamma_s \)) of the interface is an important parameter (e.g. \( \gamma_s \) for aramid and carbon are largely different). However this parameter is not included in the models which means they should be independent of the used fibre/matrix combination. Considering the largely different results obtained with carbon and aramid fibres (chapter 3), the absence of an interface parameter is most probably not very accurate.

- fibre pull-out: This energy dissipating mechanism will occur if a brittle fibre is surrounded by a relatively tough matrix system. The dissipated friction energy will be a function of:
  - distance between the fibre failure and the plane of the crack
  - the critical length of the fibre

\[ W_{FP} \div d, \tau, l_f \]

where \( W_{FP} \): pull-out work done per fibre
\( d \): fibre diameter
\( \tau \): shear stress at fibre/matrix interface
\( l_f \): maximum fibre pull-out length (function of \( l_c \): critical fibre length)

- complex evaluation of the plastic energy dissipation: For homogeneous materials like aluminium, plastic energy deformation is the major energy dissipating mechanism which is much more important than the necessary energy to disrupt bonds. Because the stress and strain distribution around a crack tip is well known for isotropic homogeneous materials, a good prediction of the plastically deformed region (and the required energy) is possible.

For fibre reinforced materials, plastic energy dissipation is limited to one component (adhesive, because the fibres are linear-elastic up to failure). The plastic energy dissipation can be
the result of tensile or shear deformation. In general load transfer by the matrix material of a composite occurs by shear stress. Therefore the plastic energy dissipation will mostly result from a shear deformation. In composites, the plastic energy dissipation is a function of:

- deformation properties of the matrix material
- presence of fibres in the local area where plasticity occurs. It is well-known that the presence of fibres in an adhesive limits the plastic energy dissipating capacity of the adhesive.
- stress and strain distribution at the crack tip (also a function of the system and not of one component of the system, even if the crack is situated in that component).

Another problem in examining the plastic energy dissipation of composites is the fact that present fracture mechanisms can cause failure before the energy density is reached at which plastic deformation does occur (e.g. if a fibre/matrix combination is introduced with a very poor interfacial strength, interfacial debonding will occur before the adhesive is able to deform plastically).

- possible presence of energy "thresholds" for the activation of fracture mechanisms: In some cases a crack will grow on a plane (or: a fracture mechanism does occur) which is energetically not a favourable one. It was not expected to dominate if an energy criterion was used. The absence of an energetically more favourable fracture mechanism is caused by the appearance of an energy "threshold" which has to be passed before the mechanism can occur. If that barrier is not exceeded, the mechanism will not be present even if it is energetically favourable (e.g. fibre failure is often energetically favourable considered the large possible amount of released energy; still a high initial threshold energy level can result in the absence of fibre failure).
Chapter 5: Discussion

In the experiments, two essentially different loading conditions were applied, i.e. mode 1 loading in the W.T.D.C.B.-test and mode 2 loading in the T.A.-test. The prime purpose of these tests was to compare the crack growth resistance ($G_{1c}$ and $G_{2c}$) of different prepregs (glass, carbon and aramide prepreg) with a pure adhesive and to examine how the crack growth resistance was affected by environmental conditions.

For the tension mode (W.T.D.C.B.-test), the following sequence was obtained:

$$G_{1c_{am}} < G_{1c_{m}} < G_{1c_{cm}}$$ (no environmental exposure)

where the subscripts
- $am$: aramide/matrix dominated fracture
- $m$: matrix dominated fracture
- $cm$: carbon/matrix dominated fracture

In chapter 4, it is argued that, because of the absence of stable crack growth for the T.A.-specimen, a fracture mechanics approach is debatable for the shear mode. Although there is no clear theoretical background one can, based on the experimental results, assume the following sequence for a shear loading mode:

$$G_{2c_{am}} < G_{2c_{m}} < G_{2c_{cm}}$$ (and $G_{2c_{gm}}$) (no environmental exposure)

where the subscript $gm$: glass/matrix interface dominated fracture

Attention will now be paid to the significance of the crack growth resistance for cracks, delaminations or damage occurring in a laminar material. Because the present investigation is part of a study on the development of ARALL (Aramid Aluminium Laminate - ref. 31), multi-layered composites (several fibre orientations in different layers) will not be considered. The discussion will be restricted to three simple cases (figs. 40, 41 and 42): sheets with a central through-crack (case A and B; figs 40 and 41) and a typical ARALL configuration (case C; fig. 42).

For the cases A and C it is assumed that initial damage (caused by impact or fatigue) did not result in fibre failure; only for case B it is assumed that the fibres failed at the crack.

It is obvious that the cases A and B do not have a direct practical relevance since in almost all structural parts, a composite material is built-up from prepreg layers with different fibre orientations. However, an analysis of these cases is still worthwhile in view of the meaning of the energy balance principle discussed before. Case C
is, for the ARALL material, directly relevant to practical problems since it has been shown by ultrasonic scanning that such delaminated areas occur around growing fatigue cracks (fibres remain intact). In this case, two failure modes should be considered:

- extension of the crack in the aluminium alloy sheet
- extension of delaminated area in plane of the specimen

In this qualitative comparison between the fracture toughness behaviour of different types of fibres, the following assumptions are made for the specimen built-up:

- all fibres are oriented in one direction
- specimen is built-up from U.D.-prepregs where for all fibres the same matrix system is used
- for all fibres, fibre volume content is considered to be constant (± 50%)
- through-crack (case A and B) is introduced without causing delamination
- four types of fibres are compared; the stress-strain curves of the fibres are shown in fig. 43.

5.1: Case A: through-crack

fibre orientation perpendicular to the loading direction

Because of the fibre orientation perpendicular to the loading direction, crack (delamination) growth will be dominated by the matrix or the fibre/matrix interface. Especially the influence of the carbon and glass fibres will be limited (crack resistance of the matrix is significantly lower than the crack resistance at the fibre/matrix interface). On a micro-mechanical level, the fibre distribution in the vicinity of the matrix crack will influence both the energy storage in the matrix and possible plastic deformation. Although the plastic energy dissipation capacity of most matrix systems (e.g. epoxies, to a lower degree thermoplastics) is small compared to e.g. aluminium, it can be, for composite materials, a significant part of the necessary energy for crack (delamination) growth. As mentioned before, possible plastic deformation of the matrix is a function of:

- properties of the matrix material
- fibre distribution and fibre volume fraction: the presence of fibres in the matrix will limit the deformation capacity of the matrix
- fibre properties: fibres with a small strain to failure can cause catastrophic failure of the composite before the full energy dissipating capacity of the matrix is reached (e.g. carbon fibres with a thermoplastic matrix)
- presence of a fracture mechanism with a low crack resistance can cause premature failure before the full plastic energy dissipating capacity of the matrix is reached.

Especially the last point can be unfavourable for an aramid composite (low $R$ at the fibre/matrix interface; the strongly reduced deformation capacity of the matrix because of premature fracture at the aramid/matrix interface is experimentally confirmed by R. Marissen -ref. 32). Already in this stage one can notice that the aim for tougher matrix systems (e.g. the use of thermoplastics instead of thermosetting matrices because of their high strain to failure) will not result in high-toughness composites if either the fibre/matrix interface adhesion is poor (aramid fibres) or the strain to failure of the fibres is small (e.g. carbon fibres, see case B). Especially for aramid fibre composites, the strain at which yielding of the matrix occurs can be more important than the strain to failure of the matrix. In the figure below, an "ideal" matrix is compared to an "actual" matrix system. The arrows shown possible matrix improvements.

The in this figure described "ideal" matrix would of course also be beneficial for glass and carbon fibres because the plastic energy capacity is highly increased.

Returning to case A, for both the carbon and glass specimens, a sharp crack front can be expected. High tensile stresses will occur at the crack tip and since cohesive crack extension in the matrix demands less energy than interface dominated failure, crack extension in the matrix will occur. For aramid fibres, with their low crack resistance $R$ at the fibre/matrix interface, the situation is more complicated and crack extension will be largely influenced by the distribution of the fibres in the vicinity of the crack. A prediction of the dominating crack growth mechanism is therefore, in the case of aramid fibres, rather doubtful.
5.2: Case B: through-crack
fibre orientation parallel to the loading direction

Because of the importance of the fibre properties in this case, a survey of the stress-strain curves of the different fibres is shown in fig 43. The following sequences can be found:

1) stiffness
   carbon HS > aramid HM > S-glass > E-glass

2) strain to failure
   S-glass > E-glass > aramid HM > carbon HS

3) maximum energy storage
   S-glass > aramid HM = E-glass > carbon HS

It is obvious that the maximum energy storage is a function of both the stiffness and the strain to failure:

$$U_{\text{max}} = \frac{Ec^2}{2} f$$

In this case, in succession or simultaneously, a variety of mechanisms can occur and interact with each other:

- matrix cracking (Y-direction)
- fibre failure
- delamination (dominantly in the Y-Z plane)

Interaction is possible because of:

- energy released during crack growth in one mechanism influences the energy balance of other possible mechanisms since it affects the stress distribution in a local area around the growing crack and therefore the crack driving force $G$ of other possible mechanisms.

- mechanism interaction is not only depending on the effect of the released energy of one mechanism on the local stress distribution but depends also on the threshold energy levels for crack initiation of the other possible mechanisms. The initiation phenomenon is clearly present if single fibre failure is examined. Initiation demands a lot of energy while propagation is mostly unstable.

Besides these mechanisms, secondary energy dissipating mechanisms can occur (e.g. fibre pull-out in the case of fibre failure; friction forces between matrix and fibre in the case of matrix cracking; see chapter 4).
If the stress distribution in the matrix in the vicinity of the crack is examined one may expect:

- high tensile stresses at the crack tip (high stress concentration because no fibre bridging in the cracked area!)
- high shear stresses at the crack edges because of load transfer between broken and unbroken fibres.

The stress distribution in the fibres will be rather inhomogeneous with peak loads in fibres near the crack tip.

If the matrix cracking along the Y-axis is considered as the main fracture mechanism (considering the stress distribution in the matrix), the possible occurrence of other mechanisms is depending on:

- the energy balance of different possible mechanisms: in general that mechanism will be active for which the released energy per unit of crack extension minus the energy to create a unit of crack extension \( G - R \) is maximal.
- the load level at which \( G \) equals \( R \): this can be illustrated in the figure below where two largely different mechanisms are compared:

![Energy balance diagram](image)

It is expected that mechanism 1 will dominate.

- the presence of possible energy barriers which have to be exceeded to initiate a certain mechanism
- the presence of energy dissipating secondary mechanisms (e.g. fibre pull-out)
- the presence of plastic energy deformation of the matrix

Since high tensile stresses are present at the tip of the initial through-crack (stress concentration!) one should expect crack extension in the matrix (in Y-direction). If however, further matrix cracking occurs without fibre failure, the stiff unbroken fibres will limit the crack opening and therefore diminish the stress concentration at the crack tip. Further, besides plastic deformation of the matrix, friction forces at the matrix/fibre interfaces will consume part of the available released energy. Especially when the crack growth resistance of a second mechanism is poor (aramid/matrix interface!) an initiation and propagation of a second mechanism is well possible. Therefore matrix cracking can be followed by delamination mechanisms in X-direction. This activation of a second mechanism is expected to occur first for aramid fibres (low R and comparable G at interface with other type of fibres) and also the delamina-
ted area will be larger in the case of aramid fibres.
If the load (displacement) is further increased, the fibres in the wake of the crack (assumed is some growth of the initial through-crack in the Y-direction) will fail. At the moment of fibre failure, a large amount of energy is released which can result in further matrix cracking or interface debonding (delamination) or in the worst case, cause new fibre failure. The latter mechanism will result in direct failure of the whole specimen. It is obvious that the presence of easily activated second mechanisms will be advantageous if fibre failure occurs since it will divert energy from the main crack and decrease the possibility of a "zipper" fibre failure mechanism. Regarding fig. 43, it is expected that carbon fibres fail first because of their low strain to failure. Since the release of stored energy is small, compared to the other fibres, secondary damage (e.g. delamination or matrix cracking) will be limited which however does not have to be advantageous. Both aramid and glass fibres can store more energy and will therefore show higher fracture toughness data. The possible occurrence of an easily activated mechanism (fibre/matrix debonding) is a further advantage of aramid fibres.

5.3: case C: ARALL configuration (fig 42)

Compared to the pure composites of case A and B, the fatigue crack in the aluminium sheets is for case C a complicating factor. In contradiction with most matrix systems (thermosetting materials), extensive plastic deformation is possible in the aluminium sheets. Because of the extensive plastic deformation and the limited stress concentration at the crack tip due to the unbroken fibres in the wake of the crack, load increase does not have to lead to crack extension in the aluminium sheets (as long as the fibres stay intact and therefore aramid is superior to carbon). In the matrix material, high shear stresses are present near the crack tip (this shear stress distribution will be a function of both the matrix and fibre properties and therefore differ for the different fibre types - in general, the higher the fibre stiffness, the higher the shear stress peaks) and with increasing load, delamination extension is possible. First delamination extension is expected in the case of the aramid specimen. Since the crack resistance of the aramid fibre/matrix interface is poor and the shear stress peaks occur at the interface, the delamination will preferentially grow along the fibre/matrix interface of fibres situated in the vicinity of the initial delamination. Since G at the crack tip will not differ too much with increasing crack length, debonding can occur along a significant part of the fibre length but without resulting in catastrophic failure. For the carbon and glass specimen, delamination growth will be preferentially in the fibre direction.

At a certain load (displacement), some fibres will fail in the wake of the crack which not only releases a large amount of energy but also
rapidly increases the stress concentration at the crack tip in the aluminium sheets and will result in further crack growth. Whether or not this leads to catastrophic failure depends on the ability of neighboring fibres to carry the extra load and the energy dissipation capacity of the aluminium sheets due to plastic deformation. Resuming, in an ARALL configuration, the highest toughness can be achieved for a material with:

- high fibre stiffness (decreased stress concentration at the crack tip in the aluminium sheets)
- high strain to failure of fibre (high energy storage)
- tough aluminium sheets (large plastic deformation capacity)

5.4: Impact loading

It is obvious that the damages discussed in the previous cases A and B were not the result of impact loading. Still, also damage as the result of impact loading is directly coupled with an energy balance concept since again both initiation and propagation of damage for certain mechanisms will be a function of $G$ and $R$. At the moment of impact, $G$ will be locally high and a lot of different mechanisms will be activated. Propagation of the cracks (delaminations) will be controlled by the $(G - R)$ relation occurring in the different mechanisms (the decrease of $G$ after impact is of course also a function of the external applied load, $R$ is a material constant). The danger of impact loading will be dominantly the elimination of the energy barriers for the initiation of certain mechanisms. Subsequent external loading will then cause early delamination (or crack) extension.
Chapter 6: Conclusions

In the present investigation experiments were carried out on two types of adhesively bonded specimens, viz. a width tapered double cantilever beam (W.T.D.C.B.) specimen and a thick adherend specimen. The prime purpose was to study delamination in the bond line as affected by unidirectional fibres. In the W.T.D.C.B.-specimen delamination occurs under mode 1 loading (tension) and in the T.A.-specimen it occurs under mode 2 loading (shear). The main variables of the test programme were:

- different types of fibres (aramid, carbon, glass)
- fibre orientation (parallel and perpendicular to the length direction of the specimen)
- environmental conditions (air and distilled water, different temperatures and pre-exposure)

The test results are presented as $G_{1c}$-values for the W.T.D.C.B.-specimens and as $G_{2c}$ and $\gamma$ (shear strength) for the T.A.-specimens. The fracture surfaces were examined visually and in the scanning electron microscope. The experimental results are summarized below.

- The introduction of fibres in the adhesive decreases both $G_{1c}$ and $G_{2c}$ as compared to the $G_{1c}$ and $G_{2c}$-values of the adhesive itself. The decrease is limited if the fracture mechanism remains matrix dominated (glass and carbon fibres) but is significant if the introduction of the fibres results in a fibre/matrix interface dominated fracture type (aramid fibres).

- Fibre orientation is an important parameter for prepreg systems with a matrix dominated fracture type (fibre orientation perpendicular to the crack growth direction largely increases $G_{1c}$ for a carbon prepreg) but prepregs with a fibre/matrix interface dominated failure mechanism are more or less insensitive for the fibre orientation (aramid fibres).

- Moisture, present because of pre-exposure or introduced during testing, decreases both $G_{1c}$ and $G_{2c}$ for all evaluated systems (adhesive, aramid prepreg, carbon prepreg, glass prepreg). The decrease is only dramatic if the moisture induces a change in fracture path e.g. from cohesive failure in the matrix to failure at the fibre/matrix interface (carbon fibres).

An evaluation of the experimentally obtained results in function of the fracture toughness behaviour of composite materials showed that both introduced test methods (W.T.D.C.B.- and T.A.) can be used to examine fracture toughness although the mutual relation is not always self-evident. It is shown that in some cases, de-
pending on loading condition, fibre orientation, matrix and fibre properties, a low $G_{IC}$-value (as obtained with the W.T.D.C.B.-test) will result in good fracture toughness properties while in other cases an opposite relation should be aimed at (high $G_{IC}$-value for good fracture toughness). In this context a few recommendations are presented to choose an appropriate combination of fibres and matrix system to obtain good fracture toughness properties (again of course a function of the loading condition).
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Table 1A: Number of specimens used per subject in the W.T.D.C.B.-test series
Number of specimens between brackets:
L: fibres oriented in the length direction of the specimen
W: fibres oriented in the width direction of the specimen
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Table 1B: Number of specimens used per subject in the T.A.-test series

Number of specimens between brackets

L: fibres oriented in the length direction of the specimen
W: fibres oriented in the width direction of the specimen
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Table 2: The strain energy release rate (shear mode) and average shear stress of an aramid, carbon and glass prepreg compared with an adhesive (in the prepregs, the adhesive is used as matrix material)

L: specimen with fibres in width direction
W: specimen with fibres in length direction

$$G_{2c} = \frac{1}{b' h E} \cdot \frac{P_c^2}{c}$$

$$\bar{\tau} = \frac{P_c}{b l}$$

where $P_c$: failure load
b: specimen width
h: adherend thickness
l: overlap length
E: Young’s modulus adherend
Fig 1: Width tapered double cantilever beam specimen
Fig 2: Thick adherend specimen
Fig 3: $G_{1c}$ test performed with a W.T.D.C.B.-specimen

Fig 4: $G_{1c}$ test performed in an environmental cell
Fig 5: The strain energy release rate versus displacement rate of an aramid prepreg
Fig 6: The strain energy release rate of a carbon and aramid prepreg compared with an adhesive system (in the prepreg, the adhesive is used as matrix material).
Fig 7: Fracture surface of the adhesive after $G_{1c}$ testing magnification: x50

Fig 8: Yielded character of the adhesive fracture surface magnification: x750
Fig 9: Fracture surface of the aramid prepreg after $G_{1c}$ testing
magnification: x5

Fig 10: Aramid fibre surfaces after $G_{1c}$ testing
Scanning electron microscopy: magnification x1360
Fig 11: Fracture surface of the carbon prepreg after $G_{lc}$ testing
  magnification: x5

Fig 12: Carbon fibre surfaces after $G_{lc}$ testing
  Scanning electron microscopy: magnification x1360
**Fig 13:** Failure of the carbon fibres after $G_{1c}$ testing

Scanning electron microscopy: magnification x1420
Fig 14: Influence of the fibre orientation on the energy release rate of an aramid and a carbon prepreg
Fig 15: Crack growth in a carbon prepreg with fibre oriented in the width direction of the specimen (W.T.D.C.B.-test)
Fig 16: Step-wise fracture path of specimens with carbon fibres in width direction
magnification: x5
Fig 17: Influence of distilled water exposure (90°C) on the energy release rate of a carbon and aramid prepreg.

Because for the exposed specimens, $G_{1c}$ is a function of the crack length, the crack lengths at the beginning and ending of the measurement period are noted.
Fig 18: The decrease of the influence of the deteriorated part with increasing crack length (deterioration of the specimen edges due to environmental exposure)
Fig 19a: Fracture surface of the aramid prepreg after environmental exposure (160 days distilled water at 90 °C) followed by $G_{IC}$ testing; magnification: x5

specimen 160 days in distilled water at 90 °C

Fig 19b: Comparison of the fracture surfaces of an aramid prepreg with and without environmental exposure
Fig 20: Fracture surface of the carbon prepreg after environmental exposure (160 days distilled water at 90 °C) followed by $G_{1c}$ testing

magnification: x5
Fig 21: The influence of environmental exposure on the energy release rate of two prepreggs and an adhesive (in the prepregg, the adhesive is used as matrix system)
- test performed in a distilled water environment at two different temperature levels (20 °C and 50 °C)
- Because for the exposed specimens, $G_{IC}$ is a function of the crack length, the crack lengths at the beginning and end of the measurement period are noted.
Fig 214: Crack growth rate of a specimen (adhesive interfacial layer) during $G_{IC}$ testing in an environmental cell (50°C distilled water exposure)
Fig 22A: Fracture surface of the adhesive after $G_{IC}$ testing in an environmental cell (distilled water exposure - temperature 20 °C)
magnification: x5

Fig 22B: Fracture surface of the adhesive after $G_{IC}$ testing in an environmental cell (see fig 22A)
Scanning electron microscopy: magnification x20
-3 typical fracture zones-
Fig 23: Magnification of zone A in fig 22B - cohesive failure in the adhesive - Scanning electron microscopy: magnification x85

Fig 24: Magnification of zone C in fig 22B - interfacial failure at the adhesive/adherend interface; adhesive side - Scanning electron microscopy: magnification x300
Fig 25: Magnification of zone B in fig 22B - interfacial failure at the adhesive/adherend interface; adherend side - Scanning electron microscopy: magnification x300

Fig 26: Aramid fibre surfaces after $G_{lc}$ testing in an environmental cell (distilled water exposure - temperature 20 °C ) Scanning electron microscopy: magnification x1360
Fig 27: Carbon fibre surfaces after $G_{\text{ic}}$ testing in an environmental cell (distilled water exposure - temperature 20 °C) Scanning electron microscopy: magnification x2020
Fig 2C: The energy release rate (shear mode) and the average shear stress of two prepreg and an adhesive (in the prepregs the adhesive is used as matrix system)

L: specimen with fibres in the length direction
W: specimen with fibres in the width direction
Fig. 30: Brittle fracture character of the local interface failure at the adhesive/adherend interface - adhesive side - Scanning electron microscopy: magnification x300
Fig 31: Fracture surface of the aramid prepreg (fibres in width direction) after $G_{2c}$ testing
magnification: x5

Fig 32: Fracture surface of the carbon prepreg (fibres in length direction) after $G_{2c}$ testing
magnification: x5
Fig 33A: Fracture surface of the glass prepreg (fibres in length direction) after $G_{2c}$ testing
magnification: x5

Fig 33B: Fracture surface of the glass prepreg (fibres in width direction) after $G_{2c}$ testing
magnification: x5
Fig 3: Influence of environmental exposure on the shear strength of the T.A.-specimen (aramid prepreg interfacial layer; fibres oriented in the width direction of the specimen)
Fig 3.5: Use of the slender beam theory for the calculation of $Q$ (part due to bending).

- Boundary conditions and definition of $v$ (displacement of load).
Fig 36: $G_{2c}$ specimen as used by Ripling and Mostovoy (ref 8,9)
Fig 37: Influence of the overlap length on the shear stress distribution in a thick adherend specimen
Fig 30: Comparison of the Young's moduli of an aramid fibre and epoxy adhesive
Possible failure modes in a fibre reinforced material:
1-fiber/matrix fracture, 2-fibre pull-out, 3-crack bridging, 4-matrix microcracking, 5-ductile fibre failure
6-fibre fracture at a weak flaw matrix crazing, 7-
plastic tensile strain distribution at tip of main crack,
8-plastic shear strain distribution leading to interfacial
failure, 9-longitudinal matrix failure.

Fig 39: Possible failure modes in a fibre reinforced material
as described by Arronsson (ref 11)
Fig 40: Case A: through crack
fibres perpendicular to the loading direction
Fig 41: Case B: through crack
fibres parallel to the loading direction
Fig 42: case C: ARALL configuration
Fig 43: Comparison of the elastic energy capacity of the different fibre types.