Research on fracture behaviour of the adhesive sealant based on energy failure criterion for TFT-LCD

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Abstract. The adhesive sealant is a crucial structure connecting color filters and thin film transistors in liquid crystal panels. Research on the fracture progress of the connection structure is heavily needed in reliability evaluation engineering. In this work, three types of adhesive sealants with different widths were tested by the uniaxial tensile experiment to obtain their fracture process curves, which conformed to the brittle fracture characteristics described by the bilinear cohesion zone model. Then, according to the theory of engineering fracture mechanics, the Dugdale-Barenblatt plastic zone model was employed to analyze the adhesive sealant with hole defects, and it was simplified to mode I fracture mechanics problem. Calculating with finite element numerical simulation, the numerical relationship between the stress field of the internal defect and the external stress of the material was obtained, and the brittle fracture behavior model was deduced as related to the defect size. Applying the model to the adhesive sealant, the average error of the model value after the correction was reduced from 7.98-12.13% to 6.84-7.53%, and the overall error was only within 15%. The model includes the material's basic characteristics and the defect's size that affect the fracture process, provides a theoretical basis for predicting the fracture of the sealant and improving the strength of bonded joints, thus is of great significance for material application and fracture analysis in engineering.

1. Introduction

The adhesive is widely used in the automotive, aviation, construction, and microelectronics industries due to its simplicity, lightweight, and good resistance to corrosion and fatigue [1-3]. For adhesively bonded joints to be used more widely and reliably, their fracture process and fracture behavior need to be studied more accurately.

Barenblatt and Dugdale first proposed the cohesive zone model, which was increasingly used to simulate fracture of adhesively bonded joints [4-5]. When the tensile force reaches a critical value, the crack grows. Meanwhile, the tensile force decreases until the material is entirely disconnected. Zhang Jun et al. [6] found that the Bilinear Cohesive Zone Model (B-CZM) was suitable for calculating the fracture processes of brittle adhesives with different adhesive by fracture experiments of the tensile, shear, and double-arm beam specimens. In another experimental investigation, Camacho and Oritiz [7]...
proposed B-CZM for brittle fracture and simulated the crack growth in material under the impact loading. Besides, Geubelle and Baylor [8] used B-CZM to simulate crack initiation, growth, and delamination of composites under low-velocity impact loading. In addition, cohesion zone models are widely used to analyze fracture failures in metals, polymers, and composites, and it can accurately analyze the plastic zone at the crack tip [9-13].

Based on the analysis of elastic fracture mechanics, it is clear that there is a stress singularity at the crack tip during material fracture. When an elastoplastic body containing a crack is subjected to an external force, a local plastic zone is created near the crack tip. Furthermore, the stress in the plastic zone is much lower than the stress intensity factor's stress prediction value. Irwin [15] and Barenblatt [16] independently studied and established the fracture process zone models and showed that the process is caused by deformation localization to form an aggregated plastic yield zone, which described the fracture behavior and processes in brittle materials containing defects.

There is extensive work dealing with brittle materials, and not so many in the adhesive sealant. Moreover, literature on theoretical research in the adhesive sealant seems to be limited so far. In this paper, the uniaxial tensile experiment was used to study the fracture behavior of three types of epoxy resin adhesive sealants under different widths. The fracture process curves were obtained, which conformed to the brittle fracture characteristics described by the bilinear cohesion zone model. Then, according to the theory of engineering fracture mechanics, the Dugdale-Barenblatt plastic zone was employed to simplify the fracture failure of the adhesive sealant with hole defects into a mode I fracture mechanics problem. Finally, combined with the finite element numerical simulation, the material mode I brittle fracture behavior model related to the defect size was derived. After applying it to the adhesive sealant, the corrected errors were only within ±15%, which shows accuracy.

2. Experimental section

2.1 Experimental materials

The experiment selected three types of adhesive sealants produced by J company, models A, B, and C, with the specific parameters shown in Table 1. As the force cannot be applied directly to liquid crystal panels, a square tape (with greater adhesive strength than the adhesive sealant) was designed to be applied to liquid crystal panels. Before the experiment, the surface of the liquid crystal panel was wiped with a dust-free cloth to remove surface impurities to ensure a firm adhesion.

| Performance                  | Unit | Environmental conditions | Model A | Model B | Model C |
|------------------------------|------|--------------------------|---------|---------|---------|
| Thixotropic Index            | (0.5rpm*5rpm) | / | 1.07 | 1.21 | 1.1 |
| Viscosity                    | Pa·s | 300 | 300 | 315 |
| proportion                   | g·cm² | 1.3 | 1.3 | 1.3 |
| Water absorption             | %   | 4.2 | 4.6 | 4 |
| Moisture permeability        | (g·m⁻²·24h) | / | 57 | 130 | 65 |
| Adhesive force               | kgf/cm | 2.7 | 3.6 | 3 |
| PI adhesive force            |      | 2.5 | 3.6 | 3.5 |
| Conductivity                 | μS·cm⁻¹ | 1.11 | 1.62 | 4.82 |
| Tg(DMA)                      | ℃    | 155 | 75 | 108 |

2.2 Experimental method

A universal material testing machine was used for the uniaxial tensile experiment [17], and the experimental curves of three types of adhesive sealants under different widths (widths are a and b mm, a<b) were obtained. According to industry standards, the experiment was conducted in an environment with a temperature of 23°C and humidity of 50%RH. The square tape was pasted and left for 48 hours
before the test to ensure the stability of attachment and the validity of the data. After conducting multiple sets of experiments, adequate experimental data was recorded, and the tensile force-strain curves of the adhesive sealant were obtained. The test image is shown in Figure 1.

![Test Image](image)

Fig.1 Test Image

2.3 Results discussion
The results of 20 samples are taken for the three models, respectively, as shown in Figure 2. It can be seen from Fig. 2(a) that the force changes with displacement in two stages: (1) The force increases gradually with the increase of displacement. At this stage, the defect underwent slight deformation and did not grow; (2) As the defect grows, the adhesive sealant fractured and failed, and the force drop to 0 N immediately. The apex is the critical condition for the adhesive sealant to fracture, and the maximum fracture strength is reached at this time. Additionally, the process curve conforms to brittle fracture characteristics and can be approximated by a bilinear cohesion zone model. The constitutive relationship is as follows [13]:

\[
\sigma_s = \begin{cases} 
\frac{\sigma_{sc}}{\delta_0} \delta & (\delta \leq \delta_0) \\
(1 - D) \frac{\sigma_{sc}}{\delta_0} \delta & (\delta > \delta_0)
\end{cases}
\] (1)

with \( D \) the damage factor is expressed as

\[
D = \begin{cases} 
0 & (\delta \leq \delta_0) \\
\frac{\delta_1 (\delta - \delta_0)}{\delta (\delta_1 - \delta_0)} & (\delta > \delta_0)
\end{cases}
\] (2)

with \( \sigma_s \) the normal stress, \( \sigma_{sc} \) the maximum stress value, \( \delta_0 \) the maximum stress opening displacement, and \( \delta_1 \) the maximum displacement.
It can be seen from Fig. 2(b) that under the same width, the tensile strength of the adhesive sealant of model A is the same as B. Moreover, model C is higher than others; when the width is different, the tensile strength of the adhesive sealant of the width a mm of model C is the same as that of model A and B of width b mm. It is preliminarily drawn that the adhesive sealant of model C has superior tensile performance under the same condition. Figure 2(c) shows the failure location. The fracture of the adhesive sealant mainly occurs inside the material, and the occurrence probability is as high as 80%. The average fracture tension, average fracture displacement, and average maximum displacement of the three materials under different widths are shown in Table 2.

3. Fracture mechanics model of the adhesive sealant

From the experimental analysis, it can be known that the fracture failure of the adhesive sealant occurred inside. In order to explore the cause of failure, further analysis is needed. According to the process, the adhesive sealant should be defoamed before use. Then as the characteristics of the material, there are still defects after treatment. The fracture of the adhesive sealant in bulk specimens is caused by the presence of inherent or pre-existing defects, which act as stress concentrators. FIB image and SEM
image are shown in Figure 3. The internal defects are regarded as micro-crack defects. When measuring the strength, it is simplified as a mode I open the crack in fracture mechanics, and the internal complex defect distribution is normalized into a single hole. The displacement directions of the defect surfaces are opposite each other, and they are all perpendicular to the propagation direction of the crack. The glass substrate is regarded as a rigid body, and the two-dimensional fracture mechanics model is shown in Figure 4.

![Figure 3 FIB image and SEM image of the adhesive sealant](image)

![Figure 4 Two-dimensional fracture mechanics model](image)

with \( b \) the width of the defect, \( a \) the length of the defect, \( E \) the elastic modulus of the sealant, \( \nu \) the Poisson's ratio, \( \sigma_\infty \) the external stress, and \( \sigma_0 \) the stress at the edge of the defect.

4. Fracture behavior model of the adhesive sealant

According to the experimental curve, it is necessary to establish a fracture behavior model to find the relationship between external stress and displacement. Therefore, based on the energy failure criterion, the plastic zone theory in fracture mechanics is used to theoretically analyze the two-dimensional fracture mechanics model of the adhesive sealant containing defects.

4.1 Theoretical model of mode I fracture plastic zone

Energy-based failure criterion means that for brittle or semi-brittle materials with notches or finite length cracks, if the stress intensity factor of notch or crack tip reaches or exceeds the material's critical value, the crack will grow and cause material damage. A local plastic zone forms near the crack tip when the elastoplastic body with cracks is subjected to the external force. The stress in the plastic zone is much lower than the stress intensity factor's stress prediction value. For brittle materials, the plastic zone may be small or even negligible. In this case, the analysis method of linear elastic fracture mechanics can be used. If the size of the plastic zone is much smaller than the K-field control zone, most of the stress field
in the singular zone depends on the stress intensity factor. The case becomes a small-scale yield. It can be processed by modifying the stress intensity factor derived from linear elastic mechanics [18].

According to the analysis of linear elastic mechanics, the adhesive sealant can be used as an infinite plane with a mode I penetrating crack with a length of 2a, as shown in Figure 5.

![Fig.5 Plastic zone model](image)

with R the length of the plastic zone, r and θ the polar coordinate parameters with the origin at the tip of the defect.

When tensile stress was applied, Irwin gave the near-field solution of the stress field and displacement field near the tip of the defect as [15-16]:

\[
\begin{align*}
\sigma_{x_1} &= -\frac{K_I}{(2\pi r)^{\frac{1}{2}}} \cos\left(\frac{\theta}{2}\right)[1 - \sin\left(\frac{\theta}{2}\right) \sin\left(\frac{3\theta}{2}\right)] \\
\sigma_{x_2} &= \frac{K_I}{(2\pi r)^{\frac{1}{2}}} \cos\left(\frac{\theta}{2}\right)[1 + \sin\left(\frac{\theta}{2}\right) \sin\left(\frac{3\theta}{2}\right)] \\
u_x &= \frac{K_I}{2E} \left(\frac{r}{2\pi}\right)^{\frac{1}{2}} (1 + \nu) [(2\kappa - 1) \cos\left(\frac{\theta}{2}\right) - \cos\left(\frac{3\theta}{2}\right)] \\
u_y &= \frac{K_I}{2E} \left(\frac{r}{2\pi}\right)^{\frac{1}{2}} (1 + \nu) [(2\kappa + 1) \sin\left(\frac{\theta}{2}\right) - \sin\left(\frac{3\theta}{2}\right)]
\end{align*}
\]

(3)

(4)

The mode I displacement is symmetrical to the defect and its extension line. On the upper and lower surfaces of the defect, the opening displacement of the mode I problem can be rewritten as the following formula:

\[
u_y = \frac{K_I}{2E} \left(\frac{r}{2\pi}\right)^{\frac{1}{2}} \frac{1}{(2\pi)} (\kappa + 1)
\]

(5)

with E the modulus of elasticity, ν the Poisson's ratio, \(K_I\) the mode I fracture stress intensity factor, and the \(\kappa\) parameter is as follows:

\[
\begin{align*}
\kappa &= \frac{(3 - \nu)}{1 + \nu} \quad \nu' = 0, \nu'' = \nu \quad \text{(Plane stress)} \\
\kappa &= \frac{(3 - 4\nu)}{1 + \nu} \quad \nu' = \nu, \nu'' = 0 \quad \text{(Plane strain)}
\end{align*}
\]

(6)

It can be seen from equation (3) that stress singularity appears at the defect tip, and the stress field changes with \(r^{\frac{1}{2}}\). There is cohesion at \(a < |x_1| < R + a\), and the medium outside the strip shape is linear elastic. Considering the solution of the central defect with a half-length of \(\frac{R + a}{2}\) under the far-field stress, the stress intensity factor and defect tip opening displacement are obtained:

\[
\begin{align*}
K_I &= \sigma_{\infty} \sqrt{\pi(R + a)} \\
\delta &= \frac{4\sigma_{\infty}}{E} \sqrt{(R + a)^2 - a^2}
\end{align*}
\]

(7)

Based on Dugdale's research on particular cases of ideal plastic condition zone, the crack tip field's opening displacement and plastic zone model are obtained [5]:
After sorting out the above formula, the relationship between the defect displacement, defect size, external stress, and stress in the cohesive zone of a single material containing a defect in the adhesive sealant can be calculated, as shown in the following formula:

\[
\delta = \frac{8\sigma_s a}{\pi E} \ln \left( \frac{R + a}{a} \right)
\]

\[
R = \left( \sec \frac{\pi \sigma_{\infty}}{2\sigma_s} - 1 \right) a
\]

(8)

4.2 The relationship of internal and external stress of the adhesive sealant

The B-CZM constitutive relationship only described the relationship between the stress at the defect and the fracture displacement. Also, it was necessary to calculate the relationship between the edge stress of the internal defect and the external stress. Therefore, the simulation model was established using the finite element numerical analysis method to calculate the stress field of the defect on the loading state[19-23], and the corresponding area and numerical relationship were shown in Figure 6. It was assumed that the displacement outside the material was the same as the defect. The material parameters were set to elastic modulus 2.94GPa, 3.4GPa, and 3.6GPa, respectively, and Poisson's ratio was 0.4. In order to improve the calculation accuracy, mesh refinement was performed on the vicinity of the defect and the material's surface until the mesh size did not affect the calculation result. The stress distribution curve around the defect was derived, and the relationship between \(\sigma_{\infty}\) and \(\sigma_s\) is obtained.

![Fig.6 (a) Plastic zone ; (b) Fracture curve ; (c) Stress distribution from the defect to the surface of model C; (d) The stress ratio of \(\sigma_{\infty}\) and \(\sigma_s\)](image)

Figure 6(a) shows a plastic zone diagram. Due to the displacement in the Y direction, a local plastic zone and a cohesive zone like C appear around the defect. Figure 6(b) is a comparison diagram of the experiment, numerical simulation, and cohesion curve of model C. It can be seen that the numerical simulation curve of the fracture process of the sealant has the same characteristic as the experiment and model and shows an acceptable agreement between the results of the experiment and the numerical modeling of the finite element. As the tensile displacement increases, the stress gradually increases until the maximum strength is reached. Figure 6(c) shows the stress distribution from the defect to the surface of the material. There is stress concentration around the defect. In Figure 6(d), the ratio of \(\sigma_{\infty}\) to the...
stress $\sigma_s$ gradually smoothes out as the mesh is divided, and the result indicates that the ratio is infinitely close to 0.18 for all three models.

4.3 Brittle fracture behavior model of the adhesive sealant
Under the condition of small-scale yielding, take $\sigma_\infty$ as a small amount, and take the first term of (7) concerning the $\frac{\pi \sigma_\infty}{2\sigma_s}$ gradual progress formula, and sort out formulas (1)-(2), (8)-(9), the material brittle fracture behavior model related to the defect size is obtained.

$$\sigma_\infty = \begin{cases} \frac{E \sigma_{sc}}{\pi \delta_0} \delta & (\delta \leq \delta_0) \\ \frac{-\sigma_{sc} \delta T \delta}{\pi \delta (\delta_1 - \delta_0)} + \frac{\sigma_{sc} \delta_1 \delta}{\pi \delta (\delta_1 - \delta_0)} & (\delta > \delta_0) \end{cases}$$ (10)

4.4 Model modification
The theoretical model ignores the influence of other stress, and the ratio of the stress at the defect to the external stress obtained by the numerical simulation has an inevitable error with the experiment, so the correction coefficient of $b$ is introduced, and $\sigma'_\infty = b \sigma_\infty$ is used to perform the above model modification:

$$\sigma'_\infty = b \sigma_\infty = b \begin{cases} \frac{E \sigma_{sc}}{\pi \delta_0} \delta & (\delta \leq \delta_0) \\ \frac{-\sigma_{sc} \delta T \delta}{\pi \delta (\delta_1 - \delta_0)} + \frac{\sigma_{sc} \delta_1 \delta}{\pi \delta (\delta_1 - \delta_0)} & (\delta > \delta_0) \end{cases}$$ (11)

The data of $\sigma_{sc}$, $\delta_1$, and $\delta_0$ are respectively averaged and brought into the model. According to the numerical simulation, model and experimental data, and Bessel correction formula, the $b$ is obtained. Then compared with the data of another 20 samples and the model, the specific values are shown in Table 3 and Figure 7. It can be seen from the table that the model correction coefficient values of the three types of adhesive sealants are all close to 1, indicating that the theoretical and experimental results correspond well. After correction, the average error was reduced from 7.98-12.13% to 6.84-7.53%, which means the average errors were reduced by 5%, 4%, and 1% for the three types of adhesive sealant, respectively. Figure 8 shows the specific model error distribution of the data of another 20 samples. It can be found that most of the errors are greater than 5%, but the overall errors are controlled within ±15%, indicating that the accuracy of the model is relatively high. The brittle fracture process will be affected by the defect size, fracture displacement, material properties and other parameters in the model. For different materials, the curves will have specific differences.

| The model of the adhesive sealant | Correction factor/b | Average error before correction $a_{error}$/% | Average error after correction $a'_{error}$/% | Error reduction $a_{error} - a'_{error}$/% |
|---------------------------------|---------------------|---------------------------------|--------------------------------|---------------------|
| A                               | 0.8430              | 12.1325                         | 7.5371                         | 4.5954              |
| B                               | 0.8798              | 11.9481                         | 7.0387                         | 4.9094              |
| C                               | 0.9323              | 7.9817                          | 6.8450                         | 1.1369              |

Notes: Error calculation formula: $a_{error} = \frac{\sigma_\infty - \sigma_{oc}}{\sigma_{oc}} \times 100\%$, $a'_{error} = \frac{\sigma'_\infty - \sigma_{oc}}{\sigma_{oc}} \times 100\%$. 

8
5. Conclusion

In this work, the fracture behavior of the adhesive sealant with defects in the TFT-LCD industry has been studied. Uniaxial tensile experiments obtained the fracture process characteristic curves, fracture strength, and fracture displacement of three types of adhesive sealants. The fracture process conforms to the brittle fracture characteristics described by the bilinear cohesion zone model. The Dugdale-Barenblatt plastic zone model was employed to simplify the fracture failure process of the adhesive sealant with hole defects, which was regarded as the mode I fracture mechanics problem. The finite element numerical simulation was used to calculate the numerical relationship between the external stress of the material and the stress field of the internal defect, and the brittle fracture behavior model related to the size of the defect was deduced. Applying this model to adhesive sealant and comparing with experimental data, the average errors were reduced by 5%, 4%, and 1%, respectively, and the overall errors were only within 15%. The model can be used to study the fracture behavior of mode I crack materials and bonding structures and provide a theoretical basis for predicting the fracture of the adhesive sealant and improving the strength of the bonded joints. It has guiding significance for material application and fracture analysis in engineering.

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