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Characterization of Adhesion Properties by Delamination of Ceramic-Metal Interfaces in Four Point Bending

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Abstract. The adhesive strength of ceramic - copper interfaces was measured in four point bending using a central notch for crack initiation. According to our method, plastic deformation may occur during the delamination process. FEM simulations were employed in order to separate elastic and plastic contributions to the energy consumption of the experiment. In conclusion, a novel delamination criterion based on the stress intensity at the crack tip was established. Here, the stress invariant J_3 is used as indicator for delamination of the interface. Agreement between experiments and theoretical interpretation is demonstrated for copper layers directly bonded to aluminum oxide.

Introduction

Delamination of interfaces between dissimilar materials is a major concern in microelectronic reliability. In many applications metallic layers are attached to ceramic substrates, which simultaneously play the role of electric insulators and heat sinks. Owing to Joule heating, power electronic modules are exposed to harsh thermomechanical conditions leading to mechanical fatigue as consequence of high temperature swings. Nevertheless, lifetimes of up to 30 years are required under operation conditions. Consequently, a precise knowledge of the adhesive strength of interfaces is a prerequisite of optimized engineering design.

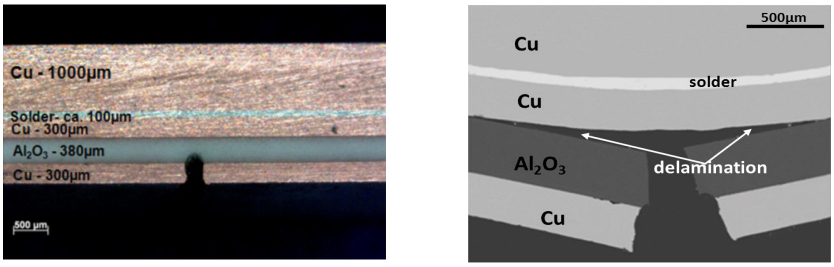
Nowadays, more than 200 methods of testing the adhesive strength between two materials are known [1]. The theoretical interpretation of adhesion properties splits up into two main branches, where the strain energy release rate represents the traditional measure of fracture mechanics, while modern interpretations are more focused on the true work of adhesion. The fracture mechanics approach is often utilized in FEM simulations for prediction of delamination.

A standard method for measuring the strain energy release rate (SERR) is provided by a four point bending setup with a central notch in the sample for initialization of delamination. This method was first proposed by Charalambides et al. [2], who developed a closed formula for the determination of SERR, which is based on the theory of elastic beams. This method was extended to multi-layered structures by Klingbeil and Beuth [3].

On the other hand, there are many sample types, which show considerable amounts of plastic deformation during delamination experiments. For example, ceramic substrates with copper metallization layers of several hundred microns are often used by electronic industry. If such samples with central notch are tested in a four point bending setup, then delamination is usually accompanied by plastic deformation. Even though the theory of elastic beams cannot be applied there, it should still be possible to extract values for the adhesive strength from the experiments.

Experimental

The samples tested in this investigation consisted of direct bonded copper layers attached to aluminum oxide. In order to detect possible geometry effects, samples of different dimensions were fabricated, whereby for one particular sample type an additional copper layer was soldered onto the Cu/alumina/Cu sandwich structure. Such a test specimen is depicted Fig. 1a. After mechanical testing, delaminated samples were inspected by Scanning Electron Microscopy. An SEM micrograph of a delaminated sample is depicted in Fig. 1b. Therefrom, it was confirmed that clean delamination of the layers has occurred without cohesive failure of the base materials.



a) b)

Figure 1: a) Selected test specimen; b) SEM micrograph of a delaminated sample.

The setup of the delamination experiments is depicted in Fig. 2, schematically. Crack initialization was achieved with use of a central notch in the sample, which was introduced prior to loading. The initial shape of the notch may be seen in Fig. 1a. During increase of the load, the notch broke and delamination between ceramic and copper was initiated. Loading of the structure was performed under displacement control, and the supplied load was measured by a load cell. A PC recorded the time dependent load-displacement curve, while the amount of crack propagation was monitored by a digital camera. Thereby, the length of the delamination crack could uniquely be related to the load level. Depending on the geometry of the composite layers, delamination usually stopped after propagating a distance between 300 µm and 2 mm.

Experimental results of delamination tests are shown in Fig. 2. For the case of 3-layered sandwich structures, our results could be compared with similar studies published recently [4, 5]. However, the addition of a copper substrate had a strong influence on the load level at which delamination was observed.

FEM Simulation and Theoretical Interpretation

The closed analytical formula evaluated by Charalambides et al. [2] makes use of the elastic theory of beams, and therefore it can only be applied to the elastic deformation regime. On the other hand, Finite Element Analysis is in general not limited by such restrictions. Consequently, one may try to extend the principles of linear elastic fracture mechanics to plastic deformations using FEM. A general approach for evaluation of cracks surrounded by plastic materials was first proposed by Irwin [6]. According to this approach, elastic and plastic contributions to the energy are separated on a theoretical level. Thus, the strain energy release is related to the recoverable elastic energy contribution. However, one should take into account that the geometries considered by Griffith and Irwin are not in perfect agreement with the four point bending setup used in the present study. Nevertheless, we at first make an attempt to extend the measurement standard for the SERR proposed by Charalambides et al. to the plastic deformation regime.

For this purpose, the experiments described in the previous section were simulated with use of elasto-plastic material models for the metallic layers. The crack lengths assumed in the simulation were correlated to the corresponding load levels determined experimentally. Due to symmetry, it was sufficient to simulate a quarter of every sample.

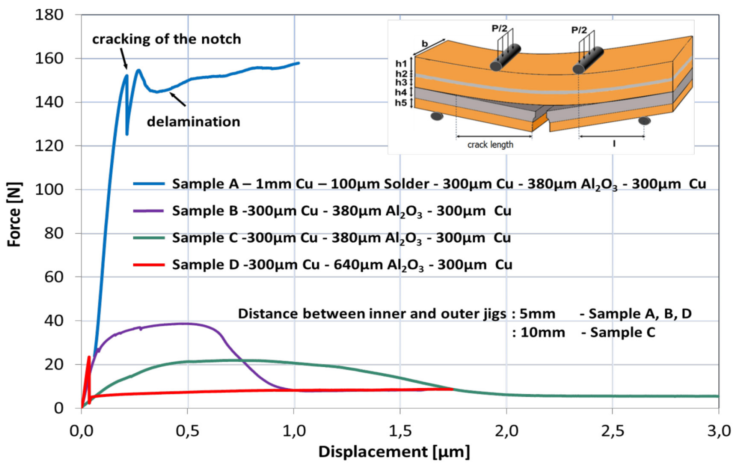


Figure 2: Force-displacement curves for selected delamination tests. The sample widths ranged from 3,7 to 6,6 mm. The experimental setup is depicted in top right position.

Following Charalambides et al. [2], the energy release of crack propagation is per definition the difference between the elastic energies of the sample at the end and at the onset of delamination. This energy release must be related to the delaminated interface area. The separation of recoverable elastic energy from the plastic energy contribution was achieved with use of FEM simulations. In principle, there are several methods how this information may be extracted from simulation data. The simplest way is to unload the specimen on theoretical level in order to derive the stored elastic energy from a load - displacement curve. For the Cu/alumina/Cu sandwich structures, a strain energy release rate of approximately 50 J/m^2 was obtained, whereas the addition of a copper substrate resulted in a strain energy release rate of 122 J/m^2. However, the delaminated interfaces of either structure had the same adhesive strength. Therefrom, it is concluded that the extension of the test standard, which has been tried here, is not perfectly validated. Therefore, a different delamination criterion is hereafter elaborated.

It is the main purpose of the present investigation to develop a delamination criterion, which may be applied to interface structures, regardless whether they deform elastically or plastically. The novel criterion shall be related to the stress distribution in the vicinity of the crack tip. In contrast to existing calculations primarily applied to the fracture stress of welded metal interfaces [7], it is here assumed that the adhesive strength between dissimilar materials is not governed by the elastic moduli of the adjacent materials alone. Instead, the adhesive strength of metal and ceramic is considered as an independent material property, and in many cases this strength appears to be weaker than the cohesive strength of adjacent bulk materials. Nevertheless, a phenomenological investigation suggests that a critical stress intensity required for crack propagation can be found.

Since the delamination criterion should be independent of the coordinate system, we claim that an invariant of the stress tensor leads to an appropriate model description. In three dimensions there are 3 independent fundamental invariants of a second rank tensor. Therefore, we look for some decomposition into independent invariants, where 2 invariants are independent of delamination, while the third invariant can be used as indicator for possible delamination. It is therefore suggested to decompose into the invariants I_1, J_2 and J_3 of the stress tensor σ_{ij} [8]:

$$I_1 = tr(\sigma_{ij}) = -3p \quad (1), \qquad J_2 = \frac{1}{2} tr\left(s_{ij}{}^2\right) \quad (2) \quad \text{and} \quad J_3 = \frac{1}{3} tr\left(s_{ij}{}^3\right) = \det\left(s_{ij}\right) \quad (3),$$

where p is the hydrostatic pressure stress and

$$s_{ij} := \begin{pmatrix} \sigma_{11}+p & \sigma_{12} & \sigma_{13} \\ \sigma_{12} & \sigma_{22}+p & \sigma_{23} \\ \sigma_{13} & \sigma_{23} & \sigma_{33}+p \end{pmatrix} \qquad (4)$$

is the deviatoric stress tensor. The invariant I_1 is related to hydrostatic stress. From the knowledge of tensile tests performed with solder joints [9, 10], one may conclude that a triaxiality of stress does not induce delamination behavior. In fact, solder joints exposed to hydrostatic stress can carry loads above the tensile strength of bulk solder materials. Consequently, this invariant of stress is not expected to initiate delamination behavior.

The invariant J_2 is widely used in plasticity theory. Therefrom, one may conclude that high values of J_2 cause large plastic deformation. In principle, the appearance of J_2 does not exclude the possibility of delamination. But if one intends to achieve delamination of a metallic structure without huge deformations of the base material, then one needs to avoid high values of J_2, because high J_2 would rather result in cohesive failure.

Thus, we are left with the invariant J_3. Indeed, this invariant of stress leads to deformations which severely violate the symmetry of a crystal lattice. In the following we demonstrate how this stress invariant can be related to the mode angle dependence, which has frequently been reported in context with delamination experiments [11, 12]. Let us consider a crack propagating under mixed mode loading in plane stress approximation. Thus, the stress tensor takes the form

$$\sigma_{ij} = \begin{pmatrix} 0 & \sigma_{12} & 0 \\ \sigma_{12} & \sigma_{22} & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{5}$$

In the case that the von Mises stress equal to $\sqrt{3 \cdot J_2}$ takes a normalized value, one derives a dependence of the J_3 stress invariant on the angle

$$\Psi = \tan^{-1}\left(\frac{\sigma_{xy}}{\sigma_{yy}}\right) \tag{6}$$

of mode mixture. If one now defines a criterion in the sense that delamination will propagate, when the stress invariant J_3 reaches a critical value, then one obtains a relation between mode angle and von Mises stress required for delamination. A diagram representing this relation is depicted in Fig. 3. Indeed, this diagram reproduces a phenomenological mode angle dependency, which was established on the basis of numerous delamination experiments [11, 12]. Before our criterion is applied to experiments, it is necessary to consider that the stress distribution ahead of a sharp crack shows the characteristics of a stress singularity. In consequence, the delamination criterion must be treated in analogy to the critical fracture toughness K_{Ic}. Since the fracture toughness has the dimension of $Pa \cdot \sqrt{m}$, the same dimensionality is required for the critical quantity of our criterion. In consequence, we expect that in the vicinity of the crack tip the stress related to J_3 takes values, where at a distance *d* to the tip the product of stress and square root of *d* converges. However, J_3 has a dimension of Pa^3, since it has been defined through the determinant of the deviatoric stress. Therefore, we hereafter use an invariant of the form $r := \sqrt[3]{J_3} \cdot n$, where *n* is a dimensionless normalization factor. The value of *n* will later be determined through the condition that delamination shall be described by a quantity, which is comparable to the fracture toughness K_{Ic}. Further, we here define the stress intensity $R_d := \lim_{d \to 0}\left(r \cdot \sqrt{2\pi \cdot d}\right)$, which will hereafter be used as critical quantity of our delamination criterion. In conclusion, we claim that delamination at a crack tip will propagate, when the stress intensity R_d reaches a critical value of R_{dc}. Hereupon, we call R_{dc} the delamination toughness. Now, we are in the position to evaluate $n = \sqrt[3]{\frac{27}{2}}$ from the value of $\sqrt[3]{J_3}$ for a stress state, where $\sigma_{11} = 1$, and all other components of σ_{ij} are zero. After this normalization, R_{dc} and K_{Ic} are comparable quantities.

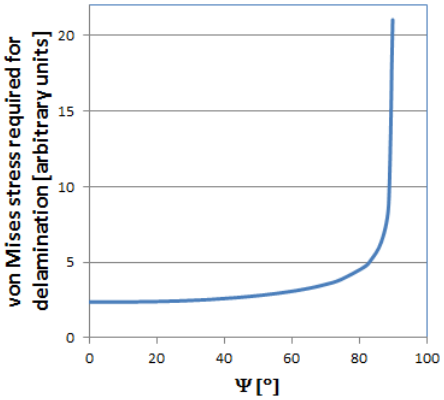


Figure 3: Mode Angle dependence, schematically.

Table 1: Comparison of R_{dc} values.

Composite DBC structure	R_{dc}
Cu/solder/Cu/Al_2O_3/Cu	3,3 MPa $\sqrt{m}$
Cu/380 µm Al_2O_3/Cu	2,7 MPa $\sqrt{m}$
Cu/630 µm Al_2O_3/Cu	2,8 MPa $\sqrt{m}$

Finally, it is demonstrated how the new criterion is successfully applied to the delamination experiments. For this purpose, FEM simulations of the experiments were performed with fine mesh size at the crack tips. The load levels were chosen in agreement with experimental data for samples of corresponding crack length. In fact, the plots of the stress invariant r depicted in Fig. 4 showed stress concentrations at the tip of the delaminating crack.

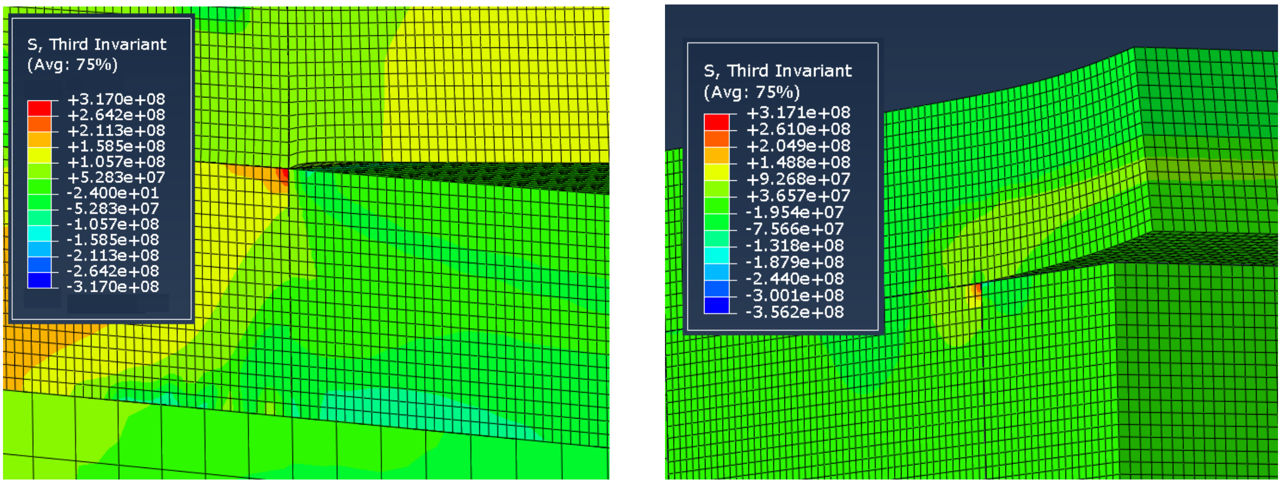


Figure 4: Plot of the third invariant of stress for different sample types. (In contrast to our definition, ABAQUS uses a normalization of $\sqrt[3]{4{,}5 \cdot J_3}$). The element size near the tip of the delamination crack was 20 µm. Left: Sample A occurring in Fig. 2. Right: Sample D.

The simulations confirmed that for the simulation of every sample $\lim_{d \to 0}\left(r \cdot \sqrt{2\pi \cdot d}\right)$ actually converges against a value R_{dc} at the load level, where delamination occurred experimentally. The R_{dc} values obtained for composite layers of different geometry are summarized in Table 1. Indeed, the R_{dc} values agree well for interfaces of equivalent adhesive strength. Therefore, the delamination toughness R_{dc} may be considered as a material parameter representing the strength of an interface.

Discussion

The delamination toughness R_{dc} may now be compared to experimental values of the fracture toughness K_{Ic} of homogeneous materials. In particular, we are interested in a comparison regarding the two adjacent materials, i.e. copper and aluminum oxide. The K_{Ic} of copper is in the range from 40 - 100 $MPa\sqrt{m}$, while the K_{Ic} of aluminum oxide is between 4 and 5 $MPa\sqrt{m}$. In fact, the delamination toughness measured in this study is somewhat smaller than the fracture toughness of aluminum oxide and much smaller than the value for copper. In view of these data, it appears logical that we have observed a clean delamination without cohesive failure of the base materials. Even though the experimental data of our investigation are restricted to a specific pair of base materials, one may try to predict a more general behavior. Insofar, delamination is possible for structures, where the delamination toughness of an interface is smaller than the fracture toughness of the adjacent materials. In the case that the critical value R_{dc} occurs at the location of the interface before the critical fracture toughness is reached in the base materials, delamination will actually occur. If R_{dc} is, however, larger than the fracture toughness of base materials, the sample will rather fail inside the base material in cohesive manner. This analysis suggests that the delamination toughness is in general an independent material property, which cannot be evaluated from the material properties of the pure base materials.

Summary and Conclusions

The adhesion properties of copper direct bonded to aluminum oxide were characterized. A first attempt was carried out with use of the strain energy release rate, whereby the four point bending test method was extended to the plastic deformation regime. However, the quantification of the interface strength along those principles was disturbed by size and geometry effects. Therefore, a novel delamination criterion has been suggested, which is based on the J_3 invariant of the stress tensor. In this context, a critical stress intensity R_{dc} was defined, which has the same dimension as the fracture toughness. A delamination crack will propagate, when the stress intensity related to the J_3 invariant of stress reaches a critical value. This criterion has been validated on the basis of a series of delamination experiments, where samples with equivalent adhesion properties were tested in combination with different geometries.

Acknowledgements

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References

[1] Volinsky, A. A., Moody, N. R., and Gerberich, W. W. (2002), Acta Mater., 50, 441–466

[2] Charalambides P. G., Lund J., Evans A. and McMeeking R., (1989), Journal of Applied Mechanics, 56, 77– 82

[3] Klingbeil N. W. and Beuth J. L., (1996), Eng. Fract. Mech. 56, pp. 113-126

[4] Tuan W. H. and Lee S. K., Ceramic Transactions, Vol. 219, (2009), 9-14

[5] Ben Kabaar A., Buttay C., Dezellus O., Estevez R., Gravouil A., Gremillard L., (2017), Microelectronics Reliability 79, pp. 288-296

[6] Irwin G. R, (1956), Onset of fast crack propagation in high strength steels and aluminum alloys, 2nd Sagamore Ordnance Materials Conference

[7] Rice, J. R., and Sih, G. C , (1965), ASME Journal of Applied Mechanics, 32, pp. 418-423.

[8] Ottosen N. S. and Ristinmaa M., The Mechanics of Constitutive Modeling (2005), Elsevier

[9] Khatibi G., Lederer M., Byrne A., Betzwar Kotas A., Weiss B. and Ipser H. (2013), Journal of Electronic Materials 42, pp. 294-303

[10] Lederer M., Khatibi G. and Weiss B. (2012), Int. J. Solids Structures 49, 3453-3460

[11] Hutchinson J. W. and Suo Z., (1992), Advances in Applied Mechanics 29, pp. 63–89

[12] Pape, H., Maus, I., Nabi, H. S., Ernst, L. J., Wunderle, B., and Ag, I. T., (2012), 62nd Electronic Components and Technology Conference (ECTC) pp. 1213–1222