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Theory of transient liquid phase bonding process

Andi Johnson

Department of Research and Development, UOP, Santiago, Chile

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ABSTRACT

During the heating stage, the entire assembly placed in a furnace and its temperature increased from room temperature to below the eutectic temperature. When the assembly is heated, some solid-state penetration occurs between the interface layer and the base metal. The amount of penetration depends on several factors, including the surface roughness and cleanliness of the surface and the pressure exerted on the surface. In most cases, it expected that the amount of particle penetration of this stage is low. However, the amount of mass transferred in this step depends on the eutectic temperature, heating rate and diffusion coefficient. The longer the heating stage, the more penetration of elements that reduce the melting point in the base metal. If the heating rate is too low and the heating step is too long, the maximum concentration of the soluble element in the interlayer may be lower than its required value for melting, and therefore, by increasing the heating step too much, there may be no Do not form a liquid. After the dissolution and expansion phase, the melting point lowering elements permeate across the solid/melt interface in the base metal during an isothermal holding period. This infiltration process is similar to multiphase infiltration coupling. To maintain thermodynamic equilibrium, the melt composition at the solid/melt interface remains constant at CLα. Therefore, the composition of the melt assumed uniform throughout the width of the liquid. Since diffusion is faster in the melt than in the solid, the thickness of the melt is too thin for this assumption to consider accurate. The direction and rate of movement of the solid/melt interface should create a mass balance in the solid/melt interface, so that the direction of movement of the two solid/melt interfaces is towards the center of the connection.

Introduction

As mentioned in chapter one, the Transient Liquid Phase (TLP) bonding process is a combination of brazing and penetration bonding processes. The initial stages of this process are similar to brazing, i.e., the junction formed by a melt, but eventually the structure of the joint will have a structure similar to that of a penetration joint. In this chapter, we will examine the theory and parameters affecting this process [1-3].

TLP process

Transient liquid phase is a relatively new method of connecting materials using an interface layer. This connection method generally includes four steps:

- ✓ Apply and confirm the connection [4].
- ✓ Heating up to a specific temperature in order to create a melt in the joint area.
- ✓ Keeping the connection set at the connection temperature until freezing occurs at the temperature of the melt due to penetration.

Corresponding Author: **Andi Johnson (andi.johnson.uop@gmail.com - ORCID: 0000-0002-2326-6763*

- ✓ Homogenization of the joint area at the appropriate heat treatment temperature.

The joint assembly usually consists of placing an interface layer in the middle of the two base materials, but sometimes the interface layer material is placed outside the joint area to flow along the joint area using the capillary effect. As it has done in many soldering processes [5-20]. The choice of connection parameters is of vital importance in TLP connection. These parameters include pressure, storage temperature, time, surface roughness, and thickness of the interlayer and composition.

Dissolution and expansion stage

TLP bonding components heated to the bonding temperature and the interface layer melts and wets the base metal contact surface. Heating above the eutectic temperature causes the melt zone to expand. By increasing the temperature above the two points of figure (1) at the border of solid and molten phases, the equilibrium composition of solid and molten occurs [21].

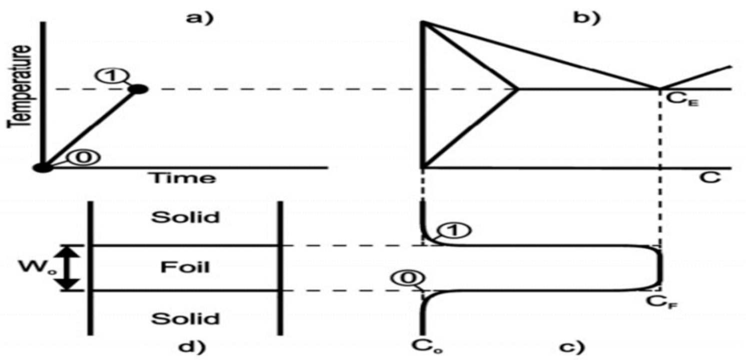

Figure 1. Schematic plan of heating stage a) Temperature profile b) Schematic phase diagram c) Connection concentration profile d) Connection schematic plan

To maintain the balance in the solid/melt interface, the base metal dissolved and as a result, the molten zone expands. Since diffusion is faster in the melt than in the solid, the melting point depressants will quickly permeate across the width of the melt at the solid/melt interface [22-30]. Dissolution kinetics depends on a number of factors such as the concentration of the dissolved element and the heating rate, so that the expansion of the molten zone in some cases will continue after reaching the junction temperature until reaching a stable state.

The motion of the solid/melt interface is shown schematically, part c [31-45]. The isothermal solidification stage is complete when the two interfaces intersect at the center of the junction and there is no residual liquid phase. The movement rate of the interface depends on the diffusion coefficient and concentration gradient of the soluble element in the base metal. The time required to complete isothermal solidification is strongly dependent on the initial volume of the liquid phase [46].

Homogenization stage

When the isothermal solidification step is complete, a concentration peak of the soluble element remains at the CαL concentration at the junction centerline, part c. In order to eliminate this peak and homogenize the concentration, the set kept at a high temperature so that the concentration peak gradually decreases over time through permeation. Homogenization of the joint will continue until an acceptable amount of soluble element, which depends on the material and use of the joint, remains in the joint zone. The concentration should be less than the amount that harmful phases formed in the solid-state during cooling [47-60]. Therefore, sediments and harmful phases reduce the mechanical properties of the connection.

Connection pressure

Usually, a pressure applied to the joint assembly to keep the base material and interface layer aligned and increase the bond. In Table 1-3, some commonly used applied pressures are classified with their closest magnitude. Sometimes the base materials placed at a fixed distance until pressure applied to them, but this tends to create porosity in the final joint. As mentioned in the scientific literature, pressure has a great effect on TLP connection. However, there is a limitation in applying pressure. If the pressure is high, as soon as the molten phase formed, it may be pushed out of the connection area, and therefore, the reduction of the melt cause a limitation in the connection. Nevertheless, pressure remains an important parameter.

Table 1. Fixture pressure used during the TLP connection process.

(%) Repetition times	The closest value
۸	۱Kpa
۵	۱۰ Kpa
۱۶	۱۰۰ Kpa
۳۶	۱Mpa
۳۱	۱۰ Mpa
۴	۱۰۰ Mpa

Heating

Heating the connection area and homogenizing the connection is done with different equipment and can be done with the following methods.

- ✓ Radiation.
- ✓ Guidance.
- ✓ Radio-frequency induction.
- ✓ Resistance.
- ✓ Laser.
- ✓ Infrared.

The connection process done in a limited space. Also, a noble gas atmosphere such as argon can be used. TLP bonding process done in different atmospheres such as nitrogen, hydrogen or free air. The vacuum pressure used in the aforementioned tests usually varies between the minimum value of 0.1 mm Hg and the maximum value of 34 mm Hg. During the heating stage, the entire assembly is heated under vacuum or a neutral atmosphere to near and lower than the eutectic temperature. The spread of influence is small in most cases, as expected. Also, the amount of mass transfer during the heating stage depends on the eutectic temperature, the heating rate and the penetration coefficient of the elements [61-70]. A higher heating rate causes more penetration of the interface layer into the base metal. However, if the heating stage is very long due to very low heating rate, the maximum composition of the interface layer solution may be less than the solidus composition at the junction temperature Tb, and no more melt will be formed in the continuation of heating. According to the mentioned article, the thickness of the interlayer has a direct effect on the heating rate and then on the strength of the connection [71-80]. Therefore, along with other parameters, it is necessary to optimize this parameter.

Connection temperature

Connection temperature is another important parameter in TLP connection. The importance of this parameter is related to the temperature required to form the melt in the joint area. Increasing the joining temperature increases the penetration of the solution into the base metal. When the eutectic is formed in the joining zone, wetting of the base metal occurs. Hong et al reported that in TLP bonding of aluminum composites reinforced with SiCp particles, the shear strength increases up to a temperature of 600°C, heating beyond this temperature causes a decrease in shear strength.

Connection time

The connection time depends on the type of materials to be connected and the required properties in the connection area. An increase in the joining temperature leads to an increase in the volume of dissolved atoms present in the base metal, which has shown to improve the joining strength compared to the strength of the base metal [81-90]. The dependence of connection time on other parameters such as pressure, temperature and surface roughness has been widely studied. The results show that there is an initial and major interaction between connection time and the mentioned parameters. The effect of one parameter cannot be compensated by another parameter [91-100]. For example, it is not possible to increase the heating time at temperatures lower than the required temperature in order to reach the same connection obtained from the previous conditions, and it is necessary to mention that all the parameters in the TLP connection are interdependent and it is not possible to keep the test parameters constant in Considering a parameter optimized.

Optimum connection temperature

Sometimes the joining temperature is completely limited due to the instability of the microstructure of the base material. If, however, the base material is flexible enough to choose an optimal bonding temperature, a minimum solidification time at the same temperature and therefore bonding time at a specific temperature will be obtained. If the phase diagram and permeation data are available in the problem for the material system, the isothermal solidification time can be determined with respect to the temperature. However, this is a common situation where experiments are the only way to find this relationship. In general, the relationship is elliptical, which is the result of the minimum isothermal freezing temperature at an intermediate temperature between the melting point of the interface layer and the base material [101-123]. Still, in some cases, system variables are still obtained.

- ✓ A uniform increase in time, in which case the optimal junction temperature is just above the melting point of the interface layer.
- ✓ A uniform reduction in time, in which case the optimal junction temperature is as high as the base material allows.

The behavior of this process depends a lot on the system and the results in part of the interactions of the penetration rate and the fuzzy diagram of the system. As the temperature increases, the penetration rate increases exponentially. In addition, it is often possible to prevent the creation of an intermetallic zone that tends to reduce the penetration rate by increasing the temperature. In addition to intermetallic compounds, the shape of solidus and liquidus lines can play an important role in TLP bonding kinetics and optimal bonding temperature. The partial distribution coefficient in the diagram is defined as follows:

Equation: 1-3

$$K=Cs/Cl$$

Where Cs and CL are the solid and liquid compounds of the interface layer. Systems a and b have the same convex liquidus line and according to experience, it results in the same amount of melt return.

- ✓ **First:** The convex line of the liquidoid form (a,b) prevents the increase of melt return value, while the convex line of the liquidoid form (d,c) causes more melt return.
- ✓ **Second:** isothermal freezing occurs faster in systems with higher partial coefficients (a,b). For mass conservation, the volume of the solidified melt is equal to the change in the concentration profile of the solid region. The width of isothermal solidification in an arbitrary period of time is equal to the volume rate, which is obtained from the difference between the liquidus and solidus compositions, or it is as follows:

Equation: 2-3

$$V_{liq}=(C_L-C_s)$$

Also, systems a and b have the same partial coefficient. Because the fractional coefficient of solidus and liquidus compounds of system c is closer to zero, they are also closer to each other and this causes isothermal freezing to happen a little faster. If the base material has a sensitive microstructure, this microstructure can be damaged by melt return. Therefore, fuzzy diagrams like c and d should be avoided. In systems b and d, isothermal freezing occurs later and increases the cost of connection operation. Systems a and c have the same freezing rate and temperature. Because the solidus composition of system c is close to the completely homogenized composition. The homogenization of the dissolution peak after isothermal freezing is likely to happen faster than system a. However, because the solidus line of system a is convex, the increase in remelting temperature due to homogenization probably does not happen faster and is larger in this system.

Interface layer

The interface layer is used in different ways. Including:

- ✓ Thin foil (rolled sheet that rolling is one of the methods of shaping materials with waxing ability. At low temperatures and close to room temperature, only metals and some polymers show proper waxing ability).
- ✓ Amorphous foil (amorphous or solid structure in which the constituents, unlike crystalline materials, do not have long-range order and only have short-range order).
- ✓ Fine powder.
- ✓ Pressed powder (made by sintering, cold isostatic pressing).
- ✓ Electroplating.
- ✓ Evaporation of an element from the substrate material to form glazed surface powders with or without bonding agent.

The thickness of the interface layer

The thickness of the interface layer has a great effect on the connection time and quality. The first consideration that should be made in this regard is that the thickness of the interface layer should be greater than the critical thickness.

Critical thickness

The thickness of the molten phase formed when the assembly reaches the melting point of the interface layer is less than the initial thickness of the interface layer. Now, if the penetration rate is high or the heating rate is slow before reaching this temperature, a large part of the interface layer penetrates into the base metal, and if these conditions are accompanied by a very low initial thickness of the interface layer, it may occur before reaching The melting point of the interface layer, the entire interface layer penetrates into the base metal and as a result, no liquid phase is formed at the interface, and since in this process, the formation of liquid phase is required to create a connection, in this condition, a proper connection will not be created. became.

Chemical composition of the interface layer

The chemical composition of the interface layer should be selected in such a way that its melting temperature is lower than the joining temperature and the penetration of its elements into the base metal occurs at a faster rate. One of the most important steps in designing an optimal connection is the selection of the interface layer, which also affects other process parameters. The first characteristic required for the interface layer is its melting temperature, which must be lower than the base metal so that the connection can be made without completely melting the base metal. Lowering the melting temperature of the interface layer is done either by adding melting point reducing elements such as boron, phosphorus, etc., or the interface layer is selected from a combination that, despite its high melting point, by forming a eutectic system with the base metal, the liquid phase at a temperature formed below If melting point reducing elements are used in the interface layer, these elements should have a high penetration coefficient in the base metal as much as possible, and it is better to have a high solubility limit in the base metal in the solid state, so that the penetration speed increases and the solidification time is at the same temperature. And also reduce the time required to homogenize the connection. Therefore, in general, the chemical composition of the interface layer should be similar to the composition of the base metal as much as possible. Because in this case, the structure becomes more homogeneous quickly and the connection time is reduced.

The critical thickness of the interface layer

During the initial heating, the elements of the intermediate layer penetrate into the base material. According to the heating rate and the thickness of the interface layer, the amount of penetration can greatly reduce its width. In fact, to obtain a high total penetration rate, slow heating or a thin interface layer, all the interface layer material can be penetrated into the base material before reaching its melting point. Also, this is a rare event. Because the connection of TLP requires the formation of a liquid phase bulk. In order to create a solid band without holes, when we want to increase the penetration rate, the interface layer must be greater than its minimum or critical value. In addition to the mentioned parameters, the critical thickness of the interfacial layer depends on variables such as the applied force of the clamp, the tension of the solid/liquid interface, the surface roughness of the base material, the formation of intermetallic materials. In short, the experiments should be directed for each integrated set of materials in a direction that shows the critical thickness of the interface layer experimentally. On the other hand, the analytical models of TLP connection show that the time of the isothermal freezing process is approximately proportional to the square of the thickness of the interface layer. Experimental data often prove this tendency. Therefore, in order to minimize the connection time, the interface layer with a little more thickness than the critical interface layer is ideal.

TLP process kinetics

It is assumed here that the interface layer and the base metal are pure elements that have complete solubility in the solid state. As the temperature increases from the ambient temperature to the melting temperature of the interface layer T3, there is some penetration from the interface layer to the base metal and vice versa, which causes the melting point of the areas near the boundary to rise in the interface layer, and as a result, the thickness of the initial molten layer is formed. and at temperature T3, it should be less than the initial thickness of the interface layer. During the process, the thickness of the central layer first decreases and then increases. As the temperature rises to the junction temperature T5, penetration continues and this penetration causes the melting point to decrease regularly in the areas adjacent to the interface in the base metal, and as a result, these areas melt. This phenomenon is called reverse melting. Therefore, when the temperature of the part reaches the joining temperature, in addition to the interface layer, parts of the base metal that are near the joining interface are also melted. As a result, the thickness of the molten layer at this temperature is greater than the initial thickness of the interface layer. By remaining the part at the penetration junction temperature, the concentration of the interlayer component in the molten/solid interface region is reduced, which results in an increase in the melting point of these regions and their freezing at the same temperature. Therefore, the freezing front moves towards the center line of the joint. If the part remains at the joint temperature long enough, the solidification front eventually reaches the center line of the joint and here the isothermal solidification is complete. At this time, the piece can be cooled or homogenized at another temperature so that penetration is complete and the chemical composition is uniform throughout the entire joint area. In this way, with homogenization, the temperature of the solidus of the connection zone increases regularly until it reaches the solidus temperature of the base metal in limiting conditions.

Some advantages and disadvantages of TLP connection

Advantages:

- ✓ Unlike the penetration welding process, TLP joining creates an isothermal solidification, which is ideal for materials that are sensitive to hot cracking or heat treatment cracks after welding.
- ✓ It is possible to remove harmful intermetallic phases in the connection line. Therefore, the structure and mechanical properties in the joint area become similar to the base metal.
- ✓ The TLP connection is resistant to surface oxides and is stable on the contact surface, and unlike penetration connection, it requires less surface preparation and applied pressure during connection.
- ✓ The mentioned advantages have made the TLP method to be used to connect a wide range of materials such as nickel-based superalloys, alloys with dispersed oxides, composite base materials and intermetallic structures. In addition, this method can be

used to connect dissimilar materials. In contrast to the wide range of applications of this type of connection, there are also disadvantages for it, which will be discussed briefly below.

Disadvantages

- ✓ High heating speed in the heating stage is needed in order to prevent the penetration of elements that reduce the melting point from the interface layer to the base metal and prevent the formation of intermetallic compounds.
- ✓ Long bonding time and post-bonding homogenization operations are required to achieve uniform microstructure, chemical composition, and mechanical properties.
- ✓ It is very difficult to choose the combination of the interface layer and the melting point lowering element, and in some cases the joining process is highly dependent on this choice.
- ✓ According to the advantages and disadvantages stated for the TLP process, this method is very efficient for joining superalloys and is used by researchers to join superalloys.

partial transient liquid phase

Partial transient liquid phase is a type of TLP that is mainly used for joining ceramics. PTLP uses both TLP gap-wide and TLP Active techniques together. Also, the presented articles state that this method was established and used before those two methods. In the following sections, the differences between PTLP and TLP will be discussed.

- ✓ **PTLP process:** In this method, the interface layer includes thin layers of metals or alloys with a low melting point, which are placed on both sides of thinner layers of refractory metals or alloys. In some cases, a thin multilayer on each side of the delay core can be used, but the general principles of the method remain the same. By heating up to the joining temperature by natural melting or eutectic reaction, a melt is formed with each of the thin layers of the refractory core. The formed melt, along with penetration into the refractory solid core, wets each of the ceramic substrates. Like TLP bonding, the molten regions are frozen at the same temperature and the homogenization process leads to a refractory bonding. The thickness of the refractory core is preferably between 20 to 30 micrometers or 100 to 127 micrometers. If it can be selected in the range of 200 to 1000 micrometers. The refractory core element is usually Ni, and at the same time, the rest of the elements or alloys used include Si, Pd, Pt, Ni-Cr, Nb, Cu, Co, Au, V, Ta, and Ti elements. In most cases, the interface layer is used in the same way as it is used in TLP, and it is often in the thickness range of 1-10 micrometers.

The second and fourth assumption is the important and big difference between TLP and PTLP connection

- ✓ **First:** The multiple interface layer used in PTLP is self-defined. This is because the molten phase must penetrate into the rc instead of the much larger base metal material to cause isothermal freezing to prevail.
- ✓ **Second:** the molten phase must wet the base metal to create a strong bond.

It seems that this issue is difficult due to the neutral property of ceramics and it usually requires the use of active elements such as Zr, V, Ti, Ta, Sc, Ni, Nb, Hf, Cr or Al. Also, when analyzing the critical thickness of the layer thin interfaces, a molten part formed from those layers react with the ceramic base metal and add to the critical thickness. Both binary systems show complete dissolution. As soon as the temperature reaches the melting point of each of the thin layers, both thin layers penetrate into rc. However, the amount of melt that is initially formed from tlB is due to the high diffusion coefficient. On further heating, the melt refreezes rc strongly due to the concave shape of the liquidus line. This freezing continues again until the set temperature reaches the connection temperature. In other words, the melt formed from tlA (CP4A) slowly expands until it reaches the same width as the original thin layer due to the convex liquidus line of the system. At this point, isothermal freezing occurs on both sides of the multiple interface layer. This happens faster for tlB due to its high partition coefficient and co-broadcast. In fact, isothermal solidification for tlB (CP5B) is complete despite significant melt solidification rc when other regions of the melt are only partially frozen. The melt formed from tlA suddenly freezes isothermally.

On the other side of the junction region, the peak of the dissolved material is flattened due to homogenization and the melting temperature increases again to TR, P. Further homogenization heat treatment makes the remaining thermal gradient in tlB disappear. In this way, the remelting temperature of the joint area next to the base metal B is followed by the final value of TR, F. An increase in melting temperature similar to (TR, P1) simultaneously occurs on the other side of the junction due to flattening of the solute peak. Extending the homogenization time continues until the remelting temperature on the left side of the junction rises.

The remelting is applied to TR, P2, which is more than the right side of TR, F, and homogenization has a weak effect on increasing the remelting temperature. From the point of view of optimization, homogenization should be stopped in this situation, but experience and reality sometimes dictate that the temperature of homogenization is higher or lower than its optimal temperature due to various reasons.

Superiority and non-superiority of the PTLP connection process

The very nature of the interface layer made of multiple PTLP connection layers creates barriers and advantages to privacy.

Its special advantages are as follows:

- ✓ The dual nature of the interlayer made of multilayers combines the beneficial properties of the two processes of soldering and penetration bonding and provides some flexibility in the design of the connection.
- ✓ A lower joining temperature can reduce thermal conduction stresses and reduce or prevent intermetallic reactions.

Because penetration occurs on a smaller scale, the connection uses elements that penetrate slowly and at reasonable times.

The important disadvantages are:

- ✓ The molten interface layer should wet the ceramic.
- ✓ Matching the coefficients of thermal expansion of the ceramic base metal and the metal elements of the interface layer is sometimes necessary to prevent thermal conduction stress and crack formation.
- ✓ The formation of intermetallic elements is necessary, but a thick reactive layer tends to become brittle and reduce the bond strength.

However, most of the disadvantages of PTLP can be overcome with proper designs. Finally, brittleness is a limiting factor in special ceramic materials.

TLP process modeling

Analytical and numerical models are used to model the TLP connection process. These models are based on fake penetration laws. Since these models make it easy to understand the effects of different parameters on the process, they are very useful. Most of the research in the field of TLP process modeling has been done analytically. In analytical models, the connection process is considered in several separate steps, which is not the case in reality. In addition, approximate methods may be used to solve the differential equations governing the problem analytically in each of the mentioned steps. While in numerical models, it is possible to analyze the process steps sequentially. It is possible to solve the problem in two or three dimensions with complicated boundary conditions by numerical method. In the following, the available numerical models for modeling the TLP connection process are described.

Numerical methods for simulating the TLP process in one dimension

- ✓ **Fuzzy field model:** In this method, two equations of fuzzy field and penetration are used. For an alloy and a mixture of solid and melt, the following equations apply.

Equation:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}[\frac{D(\emptyset)}{f_{cc}}\frac{\partial f_c}{\partial x}$$

Equation:

$$f(c,\emptyset) = h(\emptyset)f^s(c_s) + \left(1-h(\emptyset)\right)f^l(c_l) + W_g(\emptyset)$$

θ is a phase field (in melt θ is zero and in solid θ is one)? f^s and f^l are free energy data of solid and melt. The subscripts θ and C denote the partial derivative.

Equation:

$$6h(\emptyset) = \emptyset^3(15 - 10\emptyset - 6\emptyset^2)$$

Also, W, M, ‹ε are the parameters of the fuzzy field, which are determined in the following way:

Equation:

$$\varepsilon = \sqrt{\frac{6\lambda}{2.2}\sigma}$$

Equation:

$$W = \frac{6.6\sigma}{\lambda}$$

Equation:

$$M^{-1} = \frac{RT}{V_m}\frac{\varepsilon^2}{\sigma}(\frac{1-k}{m}\beta + \frac{\varepsilon}{D(\theta)\sqrt{2W}}\xi(C_s^e, C_l^e)$$

Equation:

$$\xi\left(C_s^e, C_l^e\right) = f_{cc}^{S}(C_s^e) f_{cc}^{L}(C_l^e)\left(C_s^e, C_l^e\right)\int_0^1 \frac{h(\phi)[1-h(\phi)]}{[1-h(\phi)]f_{cc}^{S}\left(C_s^e\right)+h(\phi)f_{cc}^{L}(C_l^e)\phi(1-\phi)}$$

σ energy of interface, λ thickness of interface region, R is gas constant, T is absolute temperature, V_m is molar volume, k is separation coefficient and β is kinetic coefficient. M is the phase field parameter that relates to the growth or dissolution kinetics of the solid/melt interface.

Conclusion

The results of both the fuzzy field model and the interface movement model are the same, but the calculation time in the fuzzy field model is much more than the interface movement model. Because in the fuzzy field model, in addition to the penetrating field equations, the fuzzy field equations also be calculated. In addition, a finer meshing in the fuzzy field model used for normal calculations of the interface area. Most of the simulations performed regarding TLP bonding are limited to the bonding of two similar materials, and limited reports are available on the bonding of two dissimilar materials and its simulation. Ganim has presented a two-dimensional model for TLP bonding, which is also applicable to heterogeneous materials. In this model, the infiltration in the grain boundaries is also considered, and in addition to the infiltration equations governing each half, the infiltration equations for the grain boundaries also solved. In the next section, this model described in detail.

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