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CHANGES IN THE BONDING ZONE OF EXPLOSIVELY WELDED SHEETS

ZMIANY W WARSTWIE POŁĄCZENIA PŁYT ZGRZEWANYCH WYBUCHOWO

In the presented paper two sets of Al/Cu and Ti/Ni sheets bonded through the explosive welding process was thoroughly analyzed. A particular attention was drawn to describe the changes in microstructure and chemical composition within the volumes close to the bonding interface. Optical microscopy as well as scanning (SEM) and transmission (TEM) electron microscopy were applied in the above studies whereas strain hardening across the welding zone was determined by Vickers microhardness measurements.

Optical microscopy revealed the coexistence of wavy (dominant) and flat interfaces in both sets of plates. Electron microscopy studies showed that the interface was characterized by sharp transition between two metals. For the Cu/Al sheets local melted zones were found mainly at the front slope of the waves where fluctuations in chemical composition were observed. Melting inside intermediate layers resulted in formation of intermetallic phases of the Cu_mAl_n -type while in the case of Ti/Ni sheets a continuous intermetallic layer of Ti_mNi_n -type delimitating both sheets was found.

Keywords: explosive bonding, Cu/Al and Ti/Ni plates, intermediate layer, intermetallic phase

W pracy analizowano zmiany struktury i składu chemicznego w zgrzewanych wybuchowo płytach w dwu układach blach, tj. Al/Cu i Ti/Ni wytworzonych z materiałów o czystości technicznej, ze szczególnym uwzględnieniem zmian, jakie następują w warstwach położonych w pobliżu strefy połączenia. Przeprowadzono wieloskalowe badania z wykorzystaniem mikroskopii optycznej, elektronowej mikroskopii skaningowej i spektrometru dyspersji energii promieniowania rentgenowskiego GENESIS oraz badania w skali transmisyjnej mikroskopii elektronowej.

Dla obydwu układów łączonych płyt, dla zastosowanych parametrów technologicznych procesu spajania obserwowano silne zmiany geometryczne na powierzchni łączonych blach. Szczegółowym badaniom poddano te platery, które zakwalifikowano, jako 'poprawnie połączone', tj. charakteryzujące się wymaganą stosownymi normami wytrzymałością połączenia. W najczęściej obserwowanych przypadkach uzyskane połączenie można sklasyfikować, jako płaskie lub faliste z fazą pośrednią. W wyniku przetopienia w strefie połączenia (w obydwu analizowanych układach) obserwowano tworzenie się faz międzymetalicznych typu Cu_mAl_n oraz Ti_mNi_n. Wskazuje to na zmieniającą się wzajemną koncentrację poszczególnych pierwiastków w zależności od analizowanego obszaru. Inną charakterystyczną cechą procesu spajania było wystąpienie silnego umocnienia po obydwu stronach warstwy fazy międzymetalicznej.

Zróżnicowanie pomiędzy łączonymi platerami, w najbardziej transparentny sposób ujawnia się poprzez zróżnicowaną grubość strefy pośredniej wypełnionej fazą międzymetaliczną. O ile w układzie Cu/Al występuje silna skłonność do jej pojawienia się dla bardzo szerokiego spektrum parametrów technologicznych procesu spajania, o tyle w przypadku układu Ti/Ni pojawia się ona raczej sporadycznie a uzyskane połączenie ma najczęściej charakter płaski. W tym ostatnim przypadku także i fluktuacje składu chemicznego w kierunku normalnym do powierzchni połączenia są znacznie mniejsze aniżeli dla układu Cu/Al.

1. Introduction

Explosive cladding is used in industry to join directly a wide variety of metals. This method is particularly useful in the case of those metals which can not be joined by any other welding or bonding technique [1-3].

Explosive welding is a solid state metal joining

process producing a weld joint by use of high velocity impact of one metallic mass onto another aided by controlled detonation with explosive charge. Following the explosion, the flyer plate collapses onto the parent plate and a high velocity jet is formed between two metal plates. At the same time high velocity oblique collision produces a high pressure, high temperature and

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high shear strain near the collision point in a very short time. Particularly, the pressure must be sufficiently high and reacting for enough length of time to achieve the inter-atomic bonding [2]. This causes a considerable local plastic deformation of the metals in the bond zone and leads to microstructural [4-11] and textural [12] changes. Contrary to classical methods of deformation bonding (e.g. the accumulative roll bonding or cladding by rolling), the oxide films are swept away from the interface by the jet. The bond is metallurgical in nature and it should be stronger than the weaker parent metal. From the morphological point of view the interface is very often wavy. Additionally, the welding process usually produces brittle intermetallics at the interface which strongly influences the bond strength [e.g.13].

The selection of explosive charge producing required detonation velocity is one of the most important parameters for obtaining a good weld [1]. However, the quality of the bonding strongly depends on careful control of other process parameters, e.g.: material surface preparation, the stand-off-distance and/or the inclination angle between the plates, etc. Another critical aspect for the bonding process, particularly important in the case of thick bonded plates, is the proper selection of the interlayer.

This study has been focused on the analysis of changes occurring in intermediate layers of two sets of sheets, i.e. the Cu/Al and Ni/Ti plates. After explosive welding, changes in microstructure, chemical composition and strain hardening across the interface were investigated in the initial and heat-treated samples. The cladding has been characterized by microscopic observations at different scales, energy dispersive X-ray spectrometry analysis and Vickers micro hardness measurements. A special attention has been drawn towards a detailed characterization of interfaces and possible interdiffusion between copper and aluminum as well as nickel and titanium sheets.

2. Experimental

Commercially pure annealed aluminium (AA1050) and copper (M1E-99.5%) as well as titanium (grade 1) and nickel (201) were used for manufacturing of Al/Cu and Ni/Ti plates. In the first case initial thickness of the base plate (Al) was 25 mm whereas the flyer plate (Cu) was 3 mm thick. In the second case both plates (base – Ni and flyer - Ti) were 1 mm thick. The bonding was achieved by constant stand-off explosive cladding technique.

To estimate the changes of mechanical properties close to the bonding zone, Vickers microhardness measurements were performed in a longitudinal section, i.e. along ND-RD plane, where: ND – the normal direction (perpendicular to the interface) and RD - the rolling direction which is parallel to the jetting direction. Loads of 10 G and 100 G were used for all indentations. The average value of 3 or 5 indentation measurements was used to evaluate microhardness.

Optical microscopy was applied to analyze microstructure over large areas of mechanically and chemically polished surfaces. Observations in microscale were performed by use of scanning (SEM) and transmission (TEM) electron microscopy. For this purpose specimens were cut from the welded sheets with the edges parallel to RD and ND. Chemical composition fluctuations across the interface were analyzed by use of SEM (Philips E-SEM XL30) equipped with energy dispersive X-ray spectrometer (EDAX Genesis 4000).

Specimens for TEM investigations were also cut-off from the bonded sheets perpendicular to the transverse direction (TD). Samples were mechanically ground with SiC papers followed by implementing the *twin-jet* technique (TenuPol-5 milling facility). Microstructure observations were performed on FEI Technai G^2 HREM microscope with a field emission gun operating at the accelerating voltage of 200 kV.

3. Results and discussion

3.1. Aluminium/copper plates

3.1.1. Microstructural changes

In the case of 'well-welded' sheets¹), the SEM backscattered electrons (BSE) micrograph revealed that a significant part of the interface showed strong geometrical changes when observed in the longitudinal section (i.e. ND-RD). These changes were due to formation of waves in the contacting layers and occurrence of melted zones near the top of the waves. Height and length of the waves were in the range of 40-60 μ m and 300-400 μ m, respectively (Fig. 1). A hard intermetallic phase in the bonding zone is a source of micro/macro cracks leading to a weakening of the bond strength. Cracks are limited to the melted zone and do not show the tendency for propagation across the base materials either in copper or aluminium sheets.

¹⁾ The weld is macroscopically acceptable when the clad plate is expected to be stronger than the softer of two initial metallic components.





Sheets before cladding were characterized by quite distinct mechanical properties. Such a differentiation results in formation of characteristic 'waves' observed in the intermediate layer of harder material, i.e. Cu (flyer plate). Most likely, the waves were formed due to translocation of near-surface volumes in the copper sheet and were directed along the explosive jet. Aluminium, as a softer material, deforms more uniformly, without tendency of non-uniform flow.

Microstructures observations were performed in the TEM in the areas located about 0.5 mm from the interface. After the bonding, a well-developed cellular dislocation structure, with a relatively low dislocation density was observed in both materials (Figs. 2a and b). However, primary grain boundaries were still clearly visible. For copper, the mechanism of deformation by slip was supplemented in some areas by deformation twinning, i.e. very thin twins occurring against the background of dislocation cells (Fig. 2c).

The TEM and SEM observations were supported by microhardness testing in order to evaluate the distribution of strain hardened zones close to the interface. The microhardness of fully recrystallized sheets before cladding was ~84 HV for copper and ~31 HV for aluminium. Vickers microhardness measurements were performed along the line scans perpendicular to the bonding surface, in the longitudinal section, both in the areas without melted volumes and with intermetallic layers. In the case of the interface without melted zones, measurements showed a systematic increase of microhardness when approaching the welded zone. Maximal values were obtained in the nearest neighborhood of the interface and were ~ 135 HV for Cu and ~ 50 HV for Al. For the areas lying ~ 1.0 mm from the welded surface, the values of microhardness were 20% (for Al) and 40% (for Cu) higher compared to the values obtained for recrystallized material. Such an increase might be correlated with strong structure refinement in intermediate layers, especially well visible in copper, as observed earlier by Paul et al. [12].



Fig. 2. TEM images observed in the areas \sim 0.5 mm away from the interface, in aluminium (a) and copper sheets (b); micro twinning in copper (c). Bright field images in section perpendicular to TD



Fig. 3. Vickers microhardness line scans across the Cu/Al interface in areas without melted volumes. Dotted lines mark the average microhardnes of fully recrystallized (initial) sheets



Fig. 4. SEM images in the BSE mode showing different types of interfaces observed in the Cu/Al composite (a), (c) and (e) with corresponding chemical composition variations along the line scans marked on the BSE images (b), (d) and (f). Section perpendicular to TD

A different behavior was observed in the areas occupied by wide and very hard intermetallic layers. Inside intermetallics, microhardness were recorded very often exceeded 700 HV being several times higher than the values recorded in strongly refined (and hardened) layers of the Cu plate close to the interface.

The base material in the volumes close to intermetallics is always softer than the one observed in the areas without melted zones, as reported earlier by e.g. Fu et al [9].

3.1.2. Chemical composition variations

Chemical composition variations inside intermetallic layers were characterized by the X-ray microanalysis performed along line scans parallel to ND, i.e. across the interface. Three characteristic profiles of Al and Cu concentration changes inside intermetallic phase are presented in Figs. 4a-f. For most of observed cases, the average ratio of Cu to Al (in atomic percentage) varies from 1:1 to 1:2.7. Inside intermetallic phases a higher concentration of copper was observed in the layers close to copper plate contrary to lower concentration close to aluminium sheet. Chemical fluctuations of both elements within particular intermetallic layers were also observed. Diffusion of Al and Cu atoms to the volume of neighboring plates was not recorded in the interfacial zone free of melted metals.

Some areas of the interface appeared to be flat without melted zones. However, Figs. 4b and c show the regions where the melted volumes were 'locked' inside harder parent metal, i.e. copper, creating the volumes resembling 'multilayered sandwiches' – intermetallic layers separated by parent material.

3.2. Nickel/titanium plates

3.2.1. Thin intermediate layer

This part of the work was focused on microstructure changes occurring close to the boundary zone after application of different heat treatments simulating the real conditions of the work. Variations in microstructure and chemical composition close to the interface in the 'properly bonded' materials were analyzed on the annealed samples and compared with those of just after the bonding. The thickness of bonded sheets was 1 mm. The cases described below differ from the previously analyzed mainly due to applied different detonation energy. In the case of the Ni/Ti plates microstructures observed by use of optical and scanning electron microscopy revealed quite different character of the bonding zone compared with that of observed in the Cu/Al plates. BSE images show the occurrence of a thin (of about 5 μ m), continuous layer, often found in intermediate layers of cladded composites (Fig. 5). Such an interface is typically observed in 'well-welded' Ti/Ni systems. Microstructure observed just after bonding close to the interface and corresponding X-ray microchemical analyses across the intermediate layer are presented in Figs. 5a and b, respectively. It is well-visible that within the whole area of melted zone the Ni content is always higher than the Ti content. From practical reason, it is worth noting that very important changes, in the terms simulating the real conditions of the work, occur close to the interface. Therefore, as-deformed samples were annealed at different conditions (modeling real conditions of the work). Applied heat treatments were as follows: 610°C/1h (Figs. 5c and d), 500°C/13h (Figs. 5e and f), 610° C/1h + cooling + 510° C/13h (Figs. 5g and h). The Ni content inside the intermetallic zone is always lower than the Ni content observed in as-deformed samples independently from applied heating conditions. It was also stated that different heating schemes led to similar changes, i.e. the average Ni content was always higher than the Ti content within the whole area of the intermetallic zone (Fig. 5c-h).

The SEM/EDS investigations were completed by microhardness testing in order to evaluate deformation zones throughout the thickness of cladded composites. Figure 6a shows the microhardness profile measured in the area without melted zone. Sheets in the initial state were characterized by microhardness of ~155 HV for Ti and ~ 165 HV for Ni (marked by dotted liens in Fig. 6a). A localized shock hardening at the interface occurred as indicated the values of ~ 250 HV for Ti and ~ 240 HV for Ni plates measured in the regions adjacent to the interface. It is evident that the stress concentration close to the interface developed during the process led to strong increase of hardness. In bonded sheets the size of strain hardened zone close to the interface was $\sim 600-800 \ \mu m$. Microhardness increased drastically inside intermetallic zone. (Typical measured values were ranged from 830 to 960 HV inside melted zones). However, the strength of parent materials layers adhering directly the melted zones decreased significantly (Fig. 6b). A typical Vickers microhardness profile of the Ni/Ti clad plates after heating is shown in Fig. 6c. No significant changes in microhardness distribution were observed as compared with of those observed in the samples just after cladding, independently of the applied heat treatment. As expected, microhardness and the width of strain hardened zone close to the interface decreased after annealing. The decrease was larger for Ni than for Ti. The increase of hardening in the basal plate close to the interface is well illustrated and often discussed in the literature. However, another hardened zone produced by the explosive charge was observed by Mamalis et al [6] at surface layers near the edges of titanium plate, i.e. the flyer plate.



Fig. 5. SEM microstructures and corresponding chemical composition variations along the line scans marked on the BSE images in the Ni/Ti composite: as-deformed sample (a) and (b), annealed at 610° C for 1h (c) and (d), annealed at 500° C for 13h (e) and (f), after scheme: annealing 610° C/1h + cooling + 510° C/13h (g) and (h). Section perpendicular to TD



Fig. 6. Vickers microhardness line scans across the interface in the Ni/Ti composite in the areas: with no melted zone (a), close to melted zone (b), line scan across the interface in the area without melted zone after annealing at 610°C for 1h (c). Note a strong decrease of microhardness in Ni sheets. Measurements performed in section perpendicular to TD. Dotted lines mark average microhardness of initial sheets

3.2.2. Thick intermediate layer

In the case of Ni-Ti plates with continuous intermetallic zone a fundamental parameter which decides about the welding quality is the width of melted zone. One can observe a direct correlation: if the width of melted zone increases²⁾, the tendency for macro crack occurrence increases as well leading directly to decrease of bond strength. Thus, a widely accepted statement that the fraction of melted zone is being strongly reduced at low collision energies, e.g. Mausavi and Sartangi [13], is supported by this work. For the plates bonded with very high energies a broad zone of coagulated melt is formed leading to decrease of strength properties and, in some cases, giving material of insufficient strength. Such a case is shortly described below.

Figure 7a shows the microstructure of very broad (of $\sim 50 \ \mu$ m) intermetallic layer close to the interface. Observations at high magnifications reveal more or less clearly visible network of micro/macro cracks (the source of decrease of bond strength). Here, the clad plate shows insufficient strength. Though the material was classified

as faulty, however it was used successfully for further detailed analysis of changes occurring inside a very broad intermediate layer. The EDS microanalysis from such an intermetallic layer for the as-deformed state is presented in Fig. 7b. From the plot presenting the line scan across the interfacial, it was calculated that the interlayer consists of approximately 45% wt. of Ti and 55% wt. of Ni. More detailed analyses within selected areas of interfacial zones indicate that intermetallic layers consist of fine mixture of TiNi phases, characterized by different proportions of both elements, namely TiNi₃, TiNi and Ti₂Ni, what is in a good agreement with work of Mamalis et al [6].

Another interesting phenomenon observed close to the interface is the formation of a thin zone with fine equiaxed crystallites transforming into fine columnar grains. Since they have been always formed between intermetallic and nickel sheet, it strongly supports the thesis that grain formation is due to differences in heat conductivity between Ni and Ti (Ni reveals higher heat conductivity than Ti), as reported by e.g. Yang et al [12].



Fig. 7. SEM images in the BSE mode showing microstructure of broad melted zone (a) and corresponding chemical composition variations along the line scan parallel to ND (b) in the Ni/Ti composite bonded at a very large energy of detonation. Section perpendicular to TD

²⁾ In the case of metals unveiling the tendency to form continuous intermediate layers, the higher energy the broader zone occupied by coagulated melt and the stronger tendency for macrocracks occurrence.



Fig. 8. Optical micrographs recorded at low (a) and high (b) magnifications showing a thin layer of equiaxed and columnar fine grains forming a new zone between intermetallic and nickel

4. Summary

Two sets of plates, composed of Al/Cu and Ti/Ni sheets were bonded through the explosive welding process. A particular attention was paid to the changes occurring within the volumes close to the bonding interface. The coexistence of wavy (dominant) and flat interfaces in both combinations of metals was revealed. First type of the interface is characteristic for the Cu/Al plate whereas the second one – for the Ni/Ti plate.

From phase transformation point of view, microstructural changes in the interfacial zone can lead to two types of bonds, i.e. metal-metal and metal-coagulate metallic liquid. For the Cu/Al plates local melted zones with chemical composition variations were found mainly at the front slope of waves. A formation of intermetallic phases of the Cu_mAl_n -type was observed as a result of melting close to the interface. In the case of the Ni/Ti plates a formation of continuous layer delimitating both sheets was detected. The layer was composed of different Ni-Ti intermetallic phases, i.e. TiNi₃, TiNi and Ti₂Ni-types. As the very hard intermetallic phases are formed, they are also a source of micro/macro cracks; such a situation strongly influences the global quality (strength) of the bond.

In all analyzed cases stress concentration in parent metals close to the interface developed during the processing led to strong increase of hardness. In the case of composites with a thin intermediate layer, the work hardening increases and the existence of very hard continuous layer of intermetallics without cracks may influence the overall increase of strength in the cladded composites.

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