X-RAY MAPPING OF METALLIC ELEMENTS IN ROLL BONDED METAL LAMINATES

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Introduction

Metal laminates have experienced rapid development in functional and high performance engineering applications. These laminated materials generally possess enhanced mechanical properties, leading to improved service performance. Roll bonding is recognised as an effective method in manufacture of metal laminates. In the roll bonding process, the metals are first bonded under appropriate rolling conditions. Sintering heat treatments are then applied to the bonded metals to enhance their bond strength.

Analyses of the solid state bonding of metals suggests that under the combined action of pressure and heat over short periods, a three-stage process which involves (i) development of physical contact between the metals (ii) activation of the surfaces in contact and (iii) interaction between the joined materials may occur. A mechanical bond usually forms between the metals during the rolling stage and a strong metallurgical bond eventually develops in the sintering process.

This paper is to report results obtained from an examination of atomic movement of the metallic elements across the interfacial areas via compositional profile and x-ray mapping analyses. The distribution of the metallic elements and the development of the various metallic phases in the interfacial areas were identified and located, and their effects on the property development of the metal laminates were studied.

Experimental

Metal laminates of copper and aluminium were produced by roll bonding at 430°C with a 40% rolling reduction in a single pass, Figure 1. These rolling conditions were selected to provide an optimum bond strength development. Post-rolling heat treatments (sintering) at 450°C were then applied to the bonded samples for various periods up to 3 hours. Location of the examined area was marked such that the interfacial development of the same area could be traced after each sintering heat treatment. Fine polishing was applied to the sample after each heat treatment to remove all contaminants on the surface before examination.

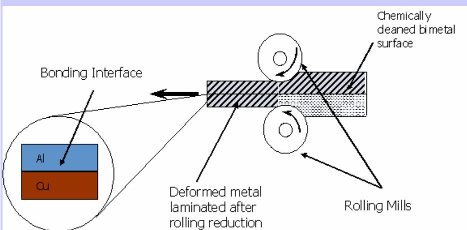

Figure 1: Schematic of the roll bonding process.

Results

Microstructural characterisation and interface development

The interfacial development of the bonded metals is shown in Figure 2. A defined interface boundary appeared in the as-rolled sample, Figure 2a. As the sintering time increased, a multi-layer interface area developed, Figure 2b. A string of voids became evident along the copper side of the bonded area after sintering for 2 hours (Figure 2c) and void formation became more serious after sintering for 3 hours (Figure 2d).

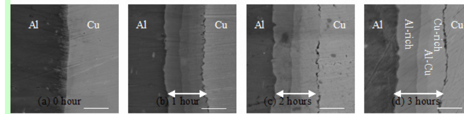

Figure 2: Secondary electron images showing interfacial development of a roll bonded copper/aluminium metal laminate after sintering at 450°C for (a) 0 hour (as-bonded); (b) 1 hour; (c) 2 hours and (d) 3 hours. Width of field (WOF) = 45μm.

The movement of the interfacial fronts was determined after each sintering heat treatment with reference to the marked locations in the sample. It was apparent that the diffusion of copper into aluminium occurs much faster than aluminium into copper, resulting in a shift of the interfacial region into the aluminium metal. The thickness of the interfacial region was also determined, Figure 3. A steady growth in the thickness of the interface area was observed with increasing sintering time. As the sintering time increased, the interfacial thickness continuously increased from 1.0 μm in the as-bonded condition to 25.7 μm after sintering at 450°C for 3 hours.

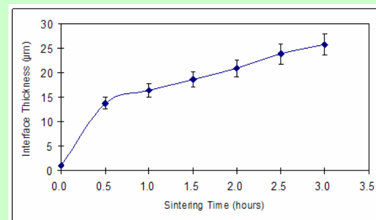

Figure 3: Growth of interfacial thickness versus sintering time at 450°C.

Interfacial composition profiles

The interfacial composition profiles of the sintered sample determined by energy dispersive x-ray spectroscopy (EDS) are shown in Figure 4. The interfacial composition profiles of the metallic elements across the interfacial area changed substantially with increasing sintering time and reflected the complexity of the interfacial development in roll bonded metal laminates. In the as-rolled material, a sharp compositional change was observed across the interface, Figure 4a.

With increasing sintering time, several transitional changes in the compositional profile developed, Figures 4b and c, showing the development of a multi-layer interface comprised of different compositions.

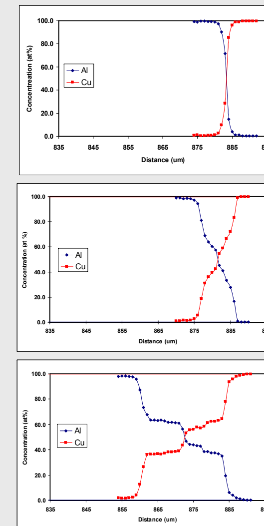

Figure 4: Interfacial composition profiles showing composition changes across the interface of the roll-bonded metals after sintering at 450°C for a) as-rolled b) 0.5 hour and c) 3 hours.

X-Ray mapping (XRM) and chemical phase mapping (CPM)

A thorough analysis was performed through use of x-ray mapping to determine i) the phases present (through CPM), ii) the composition of the phases and iii) the location of these phases in the interfacial area of the metal laminate. Figures 5a-d show the secondary electron image and quantitative x-ray maps for the copper-aluminium rolled bonded material after sintering for 3 hours. Figures 5e-g show the corresponding averaged scatter diagrams for the three respective elements present.

As shown in Figure 6, the selection of a particular cluster can give detail where a phase exists on the image. Consequently, the chemical composition of the phase can be quantified by summation of the pixels spectra from the cluster. This involves adding the spectra collected from each pixel, where the pixel is determined by the cluster. Development of the phases was found to be dependent upon the sintering conditions. With a short sintering time, a $CuAl_2$ phase was observed to form, followed by the Cu_9Al_4 phase. For sintering with a longer period, CuAl and Cu_3Al_2 phases were also found to develop.

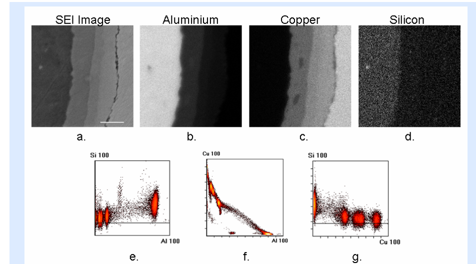

Figure 5: Copper – aluminium roll bonded metal laminate after sintering at 450°C for 3 hours. a) SE image of the interface between the two metals and elemental x-ray maps of b) aluminium, c) copper and d) silicon, and the scatter diagrams showing different clusters in the bond interface region: e) silicon versus aluminium f) copper versus aluminium g) silicon versus copper. Maps collected at 20keV, 512x512 pixel, 100msec/pixel and 7kcps. Width of field (WOF) = 45 μm.

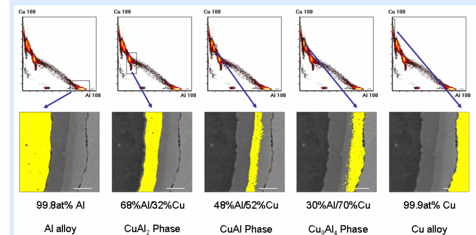

Figure 6: Copper-aluminium scatter diagrams showing many different clusters for the copper – aluminium roll bonded metal laminate after sintering at 450°C for 3 hours. The images below the scatter diagrams are secondary electron images with information from the different clusters of the scatter diagram superimposed over the image. The different yellow regions represent different phases.

According to the results of x-ray mapping analysis performed in this investigation, it appears that the development of the peak bond strength for this metal laminate is associated with the evolution of $CuAl_2$ and Cu_9Al_4 phases in the interfacial area. Existence of these interfacial phases has also been confirmed by measurements via conventional x-ray diffractometry. While the x-ray diffractometry provide the general information in identification of the interfacial phases, x-ray mapping shows its advantages and, with the help of scatter diagrams, locates the exact positions of the individual phases in the interfacial area.

Conclusion

Through the use of x-ray mapping and chemical imaging techniques, a better understanding of the distribution of the metallic elements and the phase development in the interfacial area has been achieved for the roll bonded copper-aluminium. A multiphase development of the $CuAl_2$ and Cu_9Al_4 phases was identified and located in the interfacial area.