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M E T A L L U R G Y

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THE INFLUENCE OF THE ASYMMETRIC ARB PROCESS ON THE PROPERTIES OF AI-Mg-AI MULTI-LAYER SHEETS

WPŁYW ASYMETRII W PROCESIE ARB NA WŁAŚCIWOŚCI WIELOWARSTWOWYCH BLACH AI-Mg-AI

The paper presents the results of the experimental study of the three-layer Al-Mg-Al sheets rolling process by the ARB method. The tests carried out were limited to single-pass symmetric and asymmetric rolling processes. An Al-Mg-Al package with an initial thickness of 4 mm (1-2-1 mm) was subjected to the process of rolling with a relative reduction of 50%. To activate the shear band in the strip being deformed, an asymmetry factor of $a_v=2$ was applied. From the test results, an increase in the tensile strength of the multi-layer Al-Mg-Al sheets obtained from the asymmetric process was observed. Microhardness tests did not show any significant differences in aluminium layer between respective layers of sheets obtained from the symmetric and the asymmetric process. By contrast, for the magnesium layer, an increase in microhardness from 72 HV to 79 HV could be observed for the asymmetric rolling. The analysis of the produced Al-Mg-Al sheets shows that the good bond between individual layers and grain refinement in the magnesium layer contributed to the obtaining of higher mechanical properties in the multi-layer sheets produced in the asymmetric process compared to the sheets obtained from the symmetric process.

Keywords: ARB rolling process, asymmetry rolling, Al-Mg-Al multi layer sheets, magnesium alloy, aluminium

W pracy przedstawiono wyniki badań doświadczalnych procesu walcowania metodą ARB trójwarstwowych blach Al-Mg-Al. W przeprowadzonych badaniach ograniczono się do jednoprzepustowego symetrycznego i asymetrycznego procesu walcowania. Procesowi walcowania poddano pakiet Al-Mg-Al o grubości początkowej 4 mm (1-2-1 mm) walcowany z gniotem względnym 50%. W celu aktywacji pasm ścinania w odkształcanym paśmie zastosowano współczynnik asymetrii a_y =2. Na podstawie wyników badań zaobserwowano wzrost wytrzymałości na rozciąganie wielowarstwowych blach Al-Mg-Al otrzymanych w procesie asymetrycznym. W wyniku przeprowadzonych badań mikrotwardości w poszczególnych warstwach dla blach otrzymanych w procesie symetrycznym i asymetrycznym nie stwierdzono istotnych różnic w wartościach mikrotwardości dla warstw aluminiowych. Natomiast dla warstwy magnezowej można było zaobserwować zwiększenie mikrotwardości z 72 HV do 79 HV - dla walcowania asymetrycznego. Analiza mikrostruktury otrzymanych blach Al-Mg-Al wskazuje, iż dobre połączenie pomiędzy poszczególnymi warstwami oraz rozdrobnienie ziarna w warstwie magnezu miały wpływ na uzyskanie wyższych właściwości mechanicznych w wielowarstwowych blachach otrzymanych w procesie asymetrycznym w porównaniu do blach wytworzonych w procesie symetrycznym.

1. Introduction

Ultrafine-grained and nano-crystalline structure materials exhibit very interesting properties and still not fully understood physicochemical properties, including, above all, very hard strength compared to materials obtained traditionally. The ultrafine-grained structure of metals can be obtained, among other methods, by applying SPD (Several Plastic Deformation) processes [1-4, 5-8]. To date, the most popular methods of accomplishing large plastic deformations are ECAP (Equal-Channel Angular Pressing) [1, 4] and ARB (Accumulative Roll Bonding) [5-8].

Magnesium is characterized by low density and

excellent ability to dampen vibrations. However, it exhibits poor strength and low deformability due to the fact that its crystal lattice has a limited number of slip systems at ambient temperature [5]. Reducing the grain size until the ultrafine (100 nm < d < 500 nm) or the nanoscales would thus make these alloys competitive in terms of strength with respect to other heavier materials, such as aluminum alloys. Additionally, small grain sizes often allow superplasticity at the appropriate temperatures and strain rates. Thus, fine-grained Mg alloys would be susceptible to be formed into complex parts in one single operation by superplastic forming. Another obstacle to its broader use in technology is also the fact of its relatively poor corrosion resistance and considerable abrasive wear.

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Hence, a prospective solution is to produce an Al-Mg sheets and bars that will provide increased corrosion resistance compared to magnesium sheets and bars produced by different method.

For this purpose, it is necessary to produce Al-Mg sheets and bars, in which the outer layer will be aluminium or aluminium alloy, with the individual layers being bonded together. Aluminium and its alloys exhibit better deformability compared to Mg alloys [6, 7]. Therefore, it can be foreseen that an Al-Mg bars or sheets would combine the advantages of the both materials, Al and Mg.

The techniques used to produce Al cladding layers include: heating Mg-based components in contact with Al powder [8, 9], electrodeposition [10], diffusion coating in molten salts [11, 12], cold spray coupled with heat treatment [13], physical vapour deposition [14] and laser processing [15, 16]. The above-mentioned methods provide an aluminium coating with a maximum thickness of $\sim 10 \ \mu\text{m}$ and may result in a porous cladding surface, which might in some instances lead to a worsening of the corrosion resistance [17]. It therefore seems justifiable to protect magnesium with aluminium coatings using methods that ensure an outer layer with the adequate thickness and bond quality to be obtained. Layers of this type are possible to be obtained using metal forming processes, such as extrusion [18-20], hot-rolling [21-23], coldrolling [24], explosive cladding [25], twin-roll casting [26] and ARB (Accumulative Roll Bonding) [21].

The ARB method used in the paper ensures an Al-Mg-Al multi-layer product to be obtained, which is distinguished by a fast bond between individual layers and an adequate aluminium layer thickness that effectively protects the magnesium against corrosion.

In recent years, the ARB process has been successfully applied to the manufacturing of different types of ultrafinegrained structure. Many papers [6, 7, 21, 27, 28] studied the influence of the ARB process on the mechanical properties and microstructure development of various alloys differing in crystallographic structure. The investigations carried so far have shown that the ARB process substantially enhances the plastic strength and tensile strength of alloys worked [6, 7, 21, 27-29]. For several years now, the ARB process has also been applied in the production of multi-layered sheets from different starting materials, such as Al-Ni [30], Al-Mg [21] and Cu-Zr [28]. The use of this type of sheets is owing to a relatively simple technological process and low costs of their manufacture. The investigations concerning the application of the ARB process to the production of multi-layered sheets, carried out so far, have determined the microstructure development and mechanical properties of the sheets [6, 7, 21, 23, 27-29].

Asymmetric rolling includes a shear strain during rolling decreasing the texture intensity and enhancing the elongation to failure [5, 10, 31]. Hence, the aim of the study was to determine the effect of roll rotational speed asymmetry on the quality of bond between individual components of multi-layer Al-Mg-Al sheets and on the refinement of their structure, as against the symmetric rolling process. The appropriate value of asymmetry factor, a_{y} , which is defined as the upper to lower roll rotational speed ratio, should cause additional shear stresses to occur in the strip being deformed, which will have

the effect of activating the shear bands in the deformed strip. It should be noted that in the studies of the asymmetric sheet rolling process carried out so far the asymmetry factor did not exceed the value of 1.2 [14, 15]. In studies [34-36], for rolling aluminium and magnesium sheets, a considerably greater asymmetry factor was employed, which in extreme cases ranged form 1.8 to 2.2. The studies cited above have concluded that using enhanced asymmetry factor values results in an increase in shearing stresses in the deformed material, which favourably influences the microstructure, but, at the same time, causes a large flexure of the band upon exit from the roll gap, which considerably hinders the rolling process.

2. Materials and testing methodology

Materials used for the tests were 2 mm-thick AZ31 magnesium alloy sheet and 1 mm-thick sheet of aluminium in 1050A grade.

AZ31 magnesium alloy feedstock was prepared in two stages. In the first stage, a 75x120x250 mm cast ingot was rolled on a DUO D300 rolling mill to a thickness of 4 mm. The second stage consisted in rolling magnesium sheet on a DUO D150 laboratory rolling mill to the final thickness of 2 mm. Next, the obtained magnesium sheets were subjected to bending process and then they were mechanically cleaned in order to make Al-Mg-Al packages with individual layer thicknesses of (1-2-1 mm).

Chemical composition of the materials used for the tests is given in Table 1.

Material	Chemical composition, % mass.							
	Mg	Al	Fe	Mn	Ni	Si	Zn	Cu
AZ31	95	3.5	0.01	0.4	0.01	0.1	0.8	0.05
1050A	0.05	99.5	0.4	0.05	0.01	0.25	0.7	0.05

TABLE 1 Chemical composition of the materials used for the tests

25 mm-wide and 150 mm-long specimens were cut out from magnesium and aluminium sheets. Prior to putting the aluminium and magnesium specimens together to form a package, each layer had been mechanically cleaned to remove oxides, dulled and then degreased. So prepared packages were heated up to a temperature 420°C in a chamber furnace, and then put through the rolling process. The rolling temperature was chosen based on the analysis of the investigation results reported in studies [37, 38], which showed that the quality of bond in Al-Mg-Al multi-layer materials rolled by the ARB method depended on the rolled material temperature. The investigation showed that a high-strength bond in Al-Mg-Al materials was possible to be obtained at a temperature of 400°C. Tests of rolling Al-Mg-Al specimens by the ARB method at a lower temperature of 300°C did not result in the bonding of individual components.

The rolling was carried out using, respectively, the classic ARB method with identical roll rotational speeds and the modified ARB method for which a roll rotational speed asymmetry factor of $a_v = 2$ was employed.

It should be remembered that a slip occurs between the deformed band and the rolls in the rolling process, which is associated with the delay and advance zones in the roll gap. When using a large asymmetry of roll peripheral speeds in the rolling process, individual slip zones become either extended or shortened, which may result in a metal slip across the entire roll gap length. Hence, the asymmetry factor resulting from the speed of metal plastic flow on the surfaces in contact with the upper and lower rolls is smaller that the a_v resulting from the difference between the upper and lower roll peripheral speeds [39].

The ARB process was perfomed on a laboratory DUO D150 two-high mill available at the Czestochowa University of Technology's Institute of Metal Forming and Safety Engineering, which is equipped with individual working roll drives. The test was limited to a single pass. The rolling of Al-Mg-Al packages was carried out with a relative reduction of 50% for both variants. The rolling speed in the symmetric process was 0.2 m/s. By contrast, in the asymmetric process, the rotational speed of one of the rolls was increased by two times. Specimens for static tensile tests were cut from the rolled Al-Mg-Al sheets. The test were carried out on an Instron - 5969 testing machine, and specimens were prepared for microhardness tests to be performed in individual layers using an FM-700 microhardness tester supplied by FutureTech. The microhardness test was done by the Vickers method under a load of 50 g. The examinations of the structure of individual materials used for the tests and of the multi-layer sheets obtained from the rolling process were carried out using a Nikon Eclipse MA-200 microscope. The microstructure of the AZ31 alloy was revealed using a reagent with the following composition: 20 ml ethyl alcohol + 2 ml acetic acid + 2 g picric acid, while the aluminium was etched with 5% HF. The identification of the phases in the AZ31 alloy was done on JSM 5400 scanning microscope furnished with an EDS OXFORD INSTRUMENTS ISIS 300 X-ray microanalyzer.

3. Testing results and their analysis

Figure 1 shows the initial microstructure of the magnesium alloy and aluminium, which were used as individual components for rolling multi-layer Al-Mg-Al sheets.



Fig. 1. Microstructure of the materials used for the tests: a) AZ31, b) 1050A

After the rolling process, the AZ31 alloy (Fig. 1a) is characterized by a fine-grained structure without any primary grain areas visible. Such a structure is typical for the process of hot rolling of magnesium alloys [40]. Precipitates were also observed in the obtained microstructure, which formed small chains arranged in line with the rolling direction. The quantitative analysis result, which is 19.73 atm% Mg, 51.11 atm% Al, and 29.16 atm% Mn, suggests that these are particles of the intermetallic phase Al-Mn-Mg. Figure 1b displays a microstructure of aluminium sheet on the longitudinal section, where large grains elongated in the rolling direction can be observed.

The first stage of testing was to determine the tensile strength of multi-layer Al-Mg-Al sheets obtained from the symmetric and asymmetric processes. The results yielded by the static tensile test for both rolling variants are represented in Figure 2.



Fig. 2. Tensile strength of stock materials and Al-Mg-Al sheets produced in the symmetric and asymmetric rolling processes

It can be seen from the data in Figure 2 that the tensile strength of the multi-layer Al-Mg-Al sheets obtained from the symmetric process was 130 MPa. By contrast, the tensile strength of the Al-Mg-Al sheets produced in the asymmetric rolling process was 180 MPa. Such a considerable difference in obtained tensile strength values, which amounted to approximately 30% for respective Al-Mg-Al sheets, can be explained by greater strain hardening of the magnesium layer in the asymmetric rolling process, compared to that of the magnesium layer obtained from the symmetric process. The tensile strength of multi-layer Al-Mn-Al sheets produced in the asymmetric rolling process was higher by approximately 60% than that of the initial aluminium and by approximately 30% lower than that of the initial magnesium. Whereas, for multi-layer Al-Mn-Al sheets obtained from the symmetric rolling process, these values were, respectively, 40 and 50%.

The next testing stage was to determine the microhardness of rolled Al-Mg-Al sheets. The average values obtained in individual layers of Al-Mg-Al sheets after symmetric rolling amounted to, respectively: 42 HV for aluminium 1050A and 72 HV for the AZ31 magnesium alloy. The microhardness values obtained upon asymmetric rolling, on the other hand, amounted to, respectively: 43 HV for aluminium and 79 HV for magnesium. No significant differences in obtained microhardness values for the external aluminium layers were observed between the both rolling variants. By contrast, an 9.70% increase in the microhardness of the middle AZ31 magnesium layer was noted between the Al-Mg-Al sheets produced in the symmetric and asymmetric processes, which may suggest greater grain refinement of the magnesium layer obtained from the asymmetric rolling process. The obtained tensile strength values for individual Al-Mg-Al sheets are dependent, *inter alia*, on the quality of bond between individual Al-Mg-Al components. Figure 3 shows macroscopic photographs of the longitudinal sections of Al-Mg-Al sheets as rolled, respectively, symmetrically and asymmetrically.



Fig. 3. Longitudinal section of multi-layer Al-Mg-Al sheets after rolling: a) symmetric process, b) asymmetric process

Lack of bond between the lower aluminium layer and the magnesium over 1/2 of the specimen length can be observed in Figure 3a. Small localized discontinuities were observed between the upper aluminium layer and the magnesium. Figure 3b represents the longitudinal section of Al-Mg-Al sheet produced in the asymmetric rolling process. In this case, small discontinuities can be observed at the side edges between the lower aluminium layer and the magnesium (the left-hand specimen side) and between the upper aluminium layer and the magnesium (the case of asymmetric rolling, a much better quality of bond between the aluminium and magnesium layers was achieved, which contributed to obtaining higher tensile strength values.

Figure 4 shows the microstructure of the middle layers (the AZ31 alloy) in Al-Mg-Al sheets produced in the symmetric and asymmetric rolling processes.



Fig. 4. Microstructure of the magnesium layer (the AZ31 alloy) after the rolling process: a) symmetric, b) asymmetric

In Figure 4a, shear bands are visible in the Mg layer of symmetrically rolled Al-Mg-Al sheet, which run at an angle, suggesting the plastic flow of the magnesium during this process. Strong grain refinement and Mg-Al-Mn phase particles are observed in those bands, which are oriented in line with the direction of plastic flow of the layer under consideration. Outside of the bands, the grains are much larger in size. Figure 4b represents the microstructure of the Mg layer of Al-Mg-Al sheet after the asymmetric rolling process. The applied asymmetry to the rolling process has significantly increased the intensity of occurrence of shear bands, compared to the symmetric process. As a result, regions with a very fine grain size are predominant in the microstructure.



Fig. 5. Microstructure at the Mg-Al joint boundary in Al-Mg-Al sheets after the rolling process: a) symmetric, b) asymmetric

Figure 5 illustrates the transition between the magnesium layer and the aluminium layer in multi-layer Al-Mg-Al sheets after symmetric and asymmetric rolling, respectively. In this case, only the aluminium layer was etched. For symmetrically rolled specimens, observations were conducted in locations, where a good bond between the both metals was obtained. No diffusion layer was observed to have been formed at the Mg-Al joint boundary in Al-Mg-Al sheets produced in either the symmetric or asymmetric rolling process. The rolling was conducted at a temperature lower than the eutectic temperature which, for the Mg-Al system, is 437°C [5, 41, 42]. The test results reported in study [41] demonstrate that it is possible to obtain a diffusion bond between the magnesium layer and the aluminium layer by soaking the package at a temperature of 430°C for a duration of 20 minutes. In the study presented herein, the bond between the metals is of the adhesion type; the rolling temperature was too low for reactions to occur between magnesium and aluminium, as a result of which metallurgical bonding of individual layers in the strip would have been obtained.

No major differences in microstructure were found in the Al layers; for Al-Mg-Al sheets obtained both in the symmetric and asymmetric process, large non-recrystallized grains elongated in the rolling direction could be observed. The obtained grain arrangement was the result of applying large deformation and considerable overcooling of the outer aluminium layers due to the contact with the rolls. As a result of the contact with the working rolls and the action of ambient temperature, the outer aluminium layers underwent rapid cooling, which prevented the recrystallization from occurring. Hence, large grains strongly elongated in the rolling direction were observed in the outer layers.

4. Summary

Based on the analysis of the obtained results of investigations it has been found that the introduction of asymmetry to the process of rolling multi-layer Al-Mg-Al sheets by the ARB method has contributed to increasing their tensile strength. In Al-Mg-Al sheets produced in the asymmetric rolling process, a much better bond has been obtained between the aluminium layers and the magnesium layer, compared to Al-Mg-Al sheets obtained from the symmetric rolling process. The microhardness of the Mg layer in Al-Mg-Al sheets rolled asymmetrically was by 9.7% higher than that of the Mg layer in symmetrically rolled Al-Mg-Al sheets. No significant effect of process asymmetry on the microhardness and structure of the outer Al layers has been observed. The analysis of the microstructure of the Mg layer in produced Al-Mg-Al sheets has shown that asymmetry introduced to the rolling process has considerably increased the intensity of occurrence of shear bands, comparing to the symmetric process. In the case of asymmetric rolling, a finer grain size has been obtained within the entire magnesium layer. The good bond between the layers and the grain refinement in the magnesium layer are the factors that have contributed to the obtaining of higher mechanical properties in the multi-layer Al-Mg-Al sheets produced in the asymmetric process compared to the Al-Mg-Al sheets obtained from the symmetric process.

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