Manufacturing of nanostructured Al/WC<sub>p</sub> metal-matrix composites by accumulative press bonding

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Abstract. The accumulative press bonding (APB) process used as a novel technique in this study provides an effective alternative method for manufacturing Al/10 vol.% WC<sub>p</sub> metal matrix composites (MMCs). The results revealed that by increasing the number of APB cycles (a) the uniformity of WC particles in aluminum matrix improved, (b) the porosity of the composite eliminated, (c) the particle free zones decreased. The X-ray diffraction results also showed that nanostructured Al/WC<sub>p</sub> composite with the average crystallite size of 58.4 nm was successfully achieved by employing 14 cycles of APB technique. The tensile strength of the composites enhanced by increasing the number of APB cycles, and reached to a maximum value of 216 MPa at the end of 14th cycle, which is 2.45 and 1.2 times higher than obtained values for annealed (raw material, 88 MPa) and 14 cycles APB-ed monolithic aluminum (180 MPa), respectively.

Keywords. Accumulative press bonding; Metal matrix composites; Nanostructured materials; Mechanical properties.

1. Introduction
There are several traditional processing routes to prepare MMCs such as casting, powder metallurgy and spray forming [1, 2]. Each of these processes has its own drawbacks such as non-uniform distribution of the reinforcement, poor adhesion between the matrix and the reinforcement, and undesirable chemical reaction [3-5]. Many potential applications of MMCs prompted the present authors to invent and propose another solid state process named accumulative press bonding (APB) for manufacturing MMCs. The APB process was developed based on the principle of accumulative roll bonding (ARB) process, but can be readily installed in both a laboratory and industrial environment. The APB process can be applied to sheet materials and thick billet of relatively large-scale dimensions, while the ARB process can only be applied to sheet materials. For APB, a high-capacity rolling mill is not necessary; a conventional pressing machine is all that is required. Moreover, in the APB, the strain and strain rate can precisely be controlled by controlling the amount of deformation and the pressing speed. The conventional processes are rarely used for producing bulk Al/WC<sub>p</sub> composites because the density and melting point of WC (15.63 gr/cm<sup>3</sup>, 3143 K) is significantly higher than those of aluminum (2.70 gr/cm<sup>3</sup>, 930 K) [6]. The objective of the present study is to produce bulk nanostructured Al/10 vol.% WC particulate composite using APB process. Furthermore, the effect of number of APB cycles on the microstructure and mechanical properties of monolithic aluminum and Al/WC<sub>p</sub> composite were examined.

2. Experimental procedure
As-received commercial AA1050 aluminum strips with the dimensions of 100 mm× 50 mm× 1.5 mm were annealed at 623 K in ambient atmosphere for 1 h, and WC powder with average particle size of about 10 µm, were used as raw materials. The schematic of APB process for producing Al/10 vol.% WC<sub>p</sub> composite is shown elsewhere [1]. This process was performed in two stages. To achieve a good dispersion of WC particles between strips, an acetone-base
suspension was prepared and put under ultrasonic waves with frequency of 48 KHz for 30 min. After surface preparation, ultrasonicated WC powder in acetone was sprayed between the two aluminum strips with an atomizer. Then, WC particles deposited and acetone evaporated in air, so that the brushed surface of one strip uniformly covered with WC particles. Then, the two strips were put on each other and stacked. The cold press bonding process was performed on the stacked strips with no lubrication, employing a laboratory hydraulic press machine (Toni Technik Baustoffprüfung GmbH), with a loading capacity of 200 tons. The press bonding process was carried out with a specific amount of reduction equal to 50%. The deformation in a channel die provides a reduction in the thickness and elongation in the length of the sample without any lateral spreading. The deformation in a simple die provides a reduction in the thickness and elongation in the length of the sample without any lateral spreading. Then, the press bonded strips were cut in half. The same procedure was repeated up to five cycles in the first stage. In the second stage, the mentioned procedure was repeated again up to 14 cycles without adding WC powder. The same process was employed for the production of the monolithic aluminum, while the aluminum strips were APBed without adding WC powder in any stages of APB process. Scanning electron microscopy (SEM, Philips XL30) was employed to examine the distribution of WC particles through the different cycles of APB process. The X-ray pattern of the manufactured Al/WC composite was recorded with an X-ray diffractometer (XRD). The result was used for the microstructural characterization. The XRD experiment was conducted on the specimen with dimensions of 10 mm × 10 mm × 1.5 mm by a Philips X'Pert MPD X-ray diffractometer with CuKα radiation in the range of using a step size of and a counting time of 1 s per step. Consequently, XRD pattern was analyzed by using X'Pert HighScore software, and microstructural phases of the composite were characterized. The crystallite size of the specimen was calculated from the XRD patterns applying the Williamson-Hall method [7]. The tensile test specimens were machined from the APB-ed strips, according to the ASTM E8 / E8M standard.

3. Results and discussion
The particle distribution variations of Al/WC composites produced by different cycles of APB process are shown in Fig. 1. It can be seen from Fig. 1 that in the second stage of APB process, the laminate structure including aluminum and WC layers completely vanishes and changes to a particle-reinforced composite. In fact, WC particles distribute progressively in all part of aluminum matrix, and the length of particle free zones diminishes with increasing number of cycles. After the 14th cycle of APB process, the initial clusters and particle free zones are have virtually disappeared, and the composite contains a completely uniform distribution of WC particles. When the WC particles are introduced, some of them are grouped in clusters. The clusters characteristic parameters consist of the cluster shape, the particle volume fraction in the cluster, and the volume fraction of clusters in the matrix [8-10]. According to Fig. 1, by increasing the number of APB cycles, the WC particles in the clusters is dispersed from the interfaces to the rest of the aluminum matrix and the distribution of WC particles in the matrix improve. A number of mechanisms contribute to the removal of the lamination structure and the dispersion of WC particles. At the initial cycles of the APB process, the aluminum matrix extrudes into the clusters spreading the WC particles. By continuing deformation, the particles become uniformly dispersed in the matrix. In summary, by increasing the number of APB cycles the following positive results are obtained through the production of Al/WC composite: (a) the initial WC clusters disappear, (b) the porosity of composite is eliminated; and (c) an homogeneous distribution of particles with virtually no particle free zones is obtained.
Fig. 1. Typical SEM micrographs of the Al/WC$_p$ composites produced by the APB process in various cycles and magnifications: (a) first, (b) second, (c) third, (d) seventh, (e) tenth and (f) 14th cycles.

The XRD pattern and Williamson-Hall plot of the 14 cycles APB-ed composite are shown in Fig. 2.

The Williamson-Hall method was used to calculate crystallite size from the XRD pattern of the produced MMC. As it can be seen from Fig. 2a, the diffractions of the four crystallite planes ((1 1 1), (2 0 0), (2 2 0) and (3 1 1)) are more intensive the rest ones in the aluminum matrix. Therefore, the diffractions of these four peaks are considered to estimate the crystallite size of the composite. From the intercept of the Williamson-Hall plot (Fig. 2b), the calculated mean crystallite size is 58.4 nm ($D = 0.91\lambda/\beta = 0.91 \times 0.154056 \text{ nm}/0.0024 = 58.4 \text{ nm}$). That is, the 14th cycles of APB process successfully developed a nanostructured Al/WC$_p$ MMC. Although Williamson-Hall method has many application in nano-technology including characterization of powders and deformed polycrystalline metals, some researcher reported that the grain size measured by XRD (using Williamson–Hall methods) is somewhat smaller compared to TEM (transmission electron microscopy) observations [9]. The mechanism of the nanostructure formation during APB process is an important issue, and needs further
supplementary investigations. Fig. 2a also illustrates that the manufactured Al/WCₚ composite consists of only aluminum and WC phases, and no effect of Al₄C₃, W₂C, and Al-W intermetallic become apparent. The variations of the tensile strength of Al/WCₚ composite and monolithic aluminum versus the number of APB cycles is shown in Fig. 3.

Fig. 3. The tensile strength of the monolithic aluminum and the Al/WCₚ composite produced by the APB process versus number of cycles.

It can be seen from Fig. 3 that by increasing the number of APB cycles, the tensile strength of the both aluminum and Al/WCₚ composite improves. It has been widely reported [11-13] that strength variations in severely deformed materials are governed by two main strengthening mechanisms: Grain refinement and strain hardening by dislocations. Strain hardening (or dislocation strengthening) plays an important role in enhancement of the strength in the initial stages of severely deformed metal processes, while at final stages higher strength is achieved by grain refinement [10, 13]. Since both the materials in the present investigation (monolithic aluminum and Al/WCₚ composite) have been produced by the same process (APB technique), the two mentioned mechanisms exist during the deformation of the both materials. However, as regards the composite material, there are some extra strengthening mechanisms due to the presence of the WC particles: i) the strengthening role of the WC particles: WC particles are obstacles for dislocation glide as predicted by Orowan mechanism and cause the generation of additional dislocation around the WC particles [9], decreasing the mobility of dislocations during plastic deformation; ii) the uniformity of the WC particles distribution: By increasing the number of APB cycles, the WC particles in the aluminum matrix are dispersed more uniformly (Fig. 1). This produces two effects: first, the distance between the Al/WCₚ interfaces increases, so during plastic deformation the cracks initiated in the interfaces will propagate and link up with other cracks later, increasing the ductility of Al/WCₚ composite. Additionally, the absence of particle free zones increases the resistance of the APBed Al/WCₚ composite material [14, 15]; iii) Porosity: The presence of porosity in MMCs results in a lower tensile strength and elongation. However, when the number of APB cycles increases, the porosity across the specimens decreases due to the good formability of the aluminum matrix and press pressure (Fig. 1). Thus, tensile strength enhances. As it is well known, conventional MMCs production processes suffer from (a) poor distribution of the reinforcement particles in the matrix, (b) high level of porosity in the composites, (c) undesirable chemical reaction between the reinforcement and matrix, (d) weak bonding quality between the reinforcement and matrix, and (e) disability of producing some of the MMCs such as Al/WCₚ. However, as it can be seen from the results of the present investigation, the accumulative press bonding can be a much more practical procedure for
producing high strength, without significant porosity, homogeneous bulk metal matrix composites.

4. Conclusions
A novel process for manufacturing metal matrix composite was invented, based on accumulative press bonding process (APB). Using WC_p powder and AA1050 strips, bulk nanostructured Al/WC_p composites were successfully produced through 14 cycles of the APB process. The microstructure and mechanical properties of the composite during various cycles of the process were investigated by SEM, XRD and tensile test. The conclusions drawn from the results can be summarized as follows:
1) When the number of APB cycles increased, the uniformity of WC particles in the aluminum matrix and bonding quality between them improved.
2) The manufactured Al/WC_p MMC by 14 APB cycles showed a uniform distribution of WC_p throughout the aluminum matrix, strong bonding between particles and matrix, and a microstructure without any porosity and undesirable phases.
3) The crystallite size of composite processed by 14 cycles of APB obtained by XRD analysis was 58.4 nm.
4) The tensile strength of the composites increased by increasing the number of APB cycles, and reached to a maximum value of 216 MPa at the end of the 14 cycle, which was 2.45 and 1.2 times higher than that obtained for annealed and 14 cycles APB-ed monolithic aluminum, respectively.

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