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Accelerated grain refinement during accumulative roll bonding by nanoparticle reinforcement

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Ceramic nanoparticles produced by comminution are introduced homogeneously during accumulative roll bonding (ARB) in aluminium AA1050A by airgun spraying from a stabilized aqueous suspension. In the so-produced nanoparticle-reinforced ultrafine-grained material, acceleration of microstructural evolution during the early ARB cycles as well as reduced grain sizes at higher cycles are observed, both caused by additional local straining around the embedded nanoparticles and accompanied by further strengthening. The presented method of nanoparticle application allows homogeneous as well as graded particle distribution.

Keywords: Particulate reinforced composites; Accumulative roll bonding (ARB); Ultrafine grained microstructure; Severe plastic deformation (SPD); Graded materials

It is generally known that severe plastic deformation (SPD) of metallic materials leads to the formation of an ultrafine-grained (UFG) microstructure with grain sizes clearly below 1 μm. Among the different methods of severe plastic deformation introduced in the past decades, accumulative roll bonding (ARB) [1] is the most established process for the production of UFG sheet metals. The ARB process can be easily implemented to standard rolling mills. The repeated stacking and rolling process offers the possibility of producing materials with tailored properties by producing sandwich-like laminates or introducing reinforcing phases like fibres, foils or particles [2,3]. In particular, improved mechanical properties beyond the conventional reinforcement effect of a metal matrix composite (MMC) are expected for particle-reinforced ARB materials [4,5]. Enhanced interlayer bond strength has already been demonstrated [6]. Nevertheless, the incorporation of well-dispersed nanoparticles is a major challenge. So far, particles have been employed as aggregated powder [2,6,7]. Uncontrolled agglomeration of nanoparticles leads to inhomogeneous spatial distribution of particles and strongly varying agglomerate structures and sizes, limiting the reproducibility of the reinforcement process. This problem can be overcome by employing stabilized particle suspensions and applying these directly from the liquid phase to the metal surface. For liquid phase deposition industrial standard technologies like airgun spraying can be applied, guaranteeing high controllability of the process.

The material used in this study is aluminium of commercial purity $\geqslant$99.5% (equal to AA1050A). Pure aluminium is used here as model material for ARB as solid solution strengthening and the formation of precipitates can be neglected. Before ARB processing the material was recrystallized at 773 K for 1 h and then quenched in water. Ceramic nanoparticle suspensions were produced by comminution in stirred media mills [8,9] down to a Sauter mean diameter (defined as the diameter of a sphere having the same volume to surface area ratio as the particle system) of 10 nm as determined by the nitrogen adsorption method (BET). One advantage of the nanomilling process is that suspensions with the appropriate volume concentrations can be produced by a scalable process and can be directly applied to metal sheets without any further treatment. The nanoparticles are electrostatically stabilized against agglomeration during

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comminution by adjusting the pH with nitric acid in the case of Al_2O_3 and ZrO_2 and with ammonia in the case of SiC. Furthermore, intrinsically stable SnO_2 in water is applied as a reinforcement in the ARB process. The principle and potential of the ARB process have been reported previously [1–3,10,11].

The ARB process was carried out using sheets with a size of 300 × 100 mm and a thickness of 1 mm. Acetone cleaning followed by wire brushing was applied as surface treatment, after which the particles were used in the ARB process as shown in Figure 1. Around 6 mm^3 ceramic particles were applied from suspensions with a 5 wt.% solid fraction using airgun spraying with a pressure of 0.4 MPa during each ARB cycle. After the application of particles a moderate warming of the sheets was carried out at 398 K for 300 s in order to remove the liquid suspension medium and to increase the deformability of the sheets in the following roll bonding step, where the sheets are rolled down to 50% of their initial thickness. The cyclic ARB process was carried out for up to 10 cycles. Mechanical characterization of the samples was done by uniaxial tensile testing at room temperature using an Instron 4505 universal testing machine at a strain rate of 10^{-3} s^{-1}. Microstructural characterization was done in a Zeiss Cross Beam 1540 EsB with backscattered electron contrast. A Philips CM-300 transmission electron microscope was used for further microstructural characterization.

The spatial distribution of nanoparticles in the finally produced UFG sheet is highly controllable by the applied method. This controllability offers the possibility of functionalization, grading and structuring of the reinforcement material on a small scale. Hence, tailored local material properties can be designed by this method. In this study a homogeneous distribution of particles in the sheet plane was created, while perpendicular to the sheet plane a strongly inhomogeneous distribution is observed (see Fig. 2a). This variation in the particle density perpendicular to the sheet plane is related to the introduction of particles in the ARB process. In the ARB process, after N cycles, 2^N layers, which are cold welded by $2^N - 1$ bond

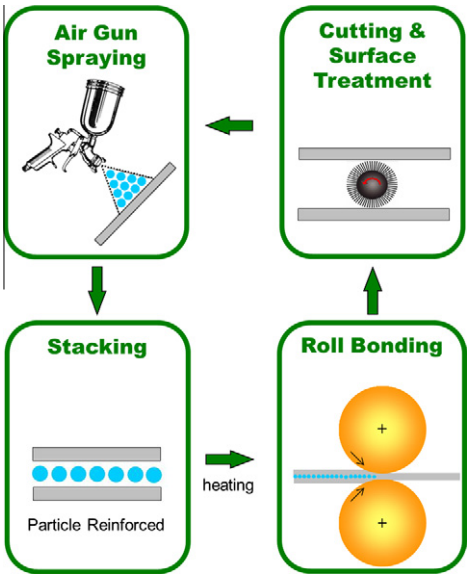

Figure 1. Nanoparticle reinforcement during the ARB process.

layers filled with particles, are formed. In this study, during each cycle the same amount of particles was introduced but, due to repeated cutting and stacking, the volume of particles introduced in a bond layer formed in the nth of N cycles is divided by $2^{(N-n)}$, leading to a strongly heterogeneous distribution in the sheet normal direction. As demonstrated in Figure 2b, the ceramic nanoparticles applied to the sheet surfaces before rolling are displaced inside the bulk material during rolling and are distributed within a distance of around 4 μm at both sides of the bond layer. This penetration depth equals the distance of bond layers produced by the described method after eight ARB cycles.

ARB itself leads to a strengthening of the material by grain refinement. In the following, the additional strengthening by nanoparticles is discussed. Compared to the recrystallized starting material, the ARB process leads to a strengthening by a factor of about 2–4 after eight cycles [10]. By reinforcement with only 0.1 vol.% of different ceramic materials of various particle sizes between 7 and 170 nm, considerable additional strengthening was achieved as compared to the unreinforced ARB sample after eight ARB cycles (see Fig. 3). Due to the high purity of the applied batch of AA1050A, the strength level achieved is quite low for UFG aluminium. A UFG microstructure was obtained for all specimens. According to the well-known Orowan mechanism [12], particle strengthening is caused by the resistance of closely spaced hard and incoherent particles to the passing of dislocations. Following Eq. (1) for 0.1 vol.% particles with a diameter of 25 nm, the resulting particle strengthening $\Delta\tau_{PS}$ amounts to 13 MPa, representing a relative strengthening of 8.5% compared to the unreinforced UFG aluminium. The Orowan mechanism assumes an equally distributed particle spacing 2λ as well as fully intragranular particles, although, as demonstrated in Figure 2, the distribution of particles in the sheet normal direction is strongly heterogeneous and the particles are mainly located at the grain boundaries.

$$\Delta\tau_{PS} = \frac{Gb}{2\lambda} = \frac{Gb}{r}\sqrt{\frac{f_v}{2}} \quad (1)$$

G is the shear modulus of aluminium (25.4 GPa), b is the Burger's vector of aluminium (0.286 nm), 2λ is the mean distance between particles, r is the mean particle radius (12.5 nm) and f_v is the volume fraction of particles (0.1%).

The contribution of the particles to a volumetric strengthening mechanism following a linear rule of mixture of strength is only relevant at higher volume fractions than present in the materials discussed here. It amounts to 0.35 MPa, and is therefore negligible in the present case. As demonstrated in Figure 3, the observed additional strengthening caused by nanoparticle reinforcement is 4.4%. In earlier experiments that applied a less aged suspension and a different batch of AA1050A with different impurity atoms, the strengthening effect of the nanoparticle reinforcement was found to reach 12.6%. Hence, the observed strengthening caused by nanoparticle reinforcement is decidedly beyond a simple linear rule of mixture of strength as the volume fraction of particles is far too low. Compared to the theoretical strengthening calculated by the Orowan model, the strengthening observed for this batch is lower, while