Synthesis of hybrid nanocrystalline alloys by mechanical bonding through high-pressure torsion

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Abstract: This review provides an overview of the mechanical bonding of dissimilar bulk engineering metals through high-pressure torsion (HPT) processing at room temperature. A recently developed procedure of mechanical bonding involves the application of conventional HPT processing to alternately stacked two or more disks of dissimilar metals. A macro-scale microstructural evolution involves the concept of making tribomaterials and, for some dissimilar metal combinations, micro-scale microstructural changes demonstrate the synthesis of metal matrix nanocomposites (MMNCs) through the nucleation of nano-scale intermetallic compounds within the nanostructured metal matrix. Further straining by HPT during the mechanical bonding provides an opportunity to introduce limited amorphous phases and a bulk metastable state. The mechanically-bonded nanostructured hybrid alloys exhibit exceptionally high specific strength and an enhanced plasticity. These experimental findings suggest a potential for using mechanical bonding for simply and expeditiously fabricating a wide range of new alloy systems by HPT processing.

1. Introduction

The synthesis of hybrid metals and alloys having functionality, such as high specific strength and good ductility, is an increasing, yet technically challenging, trend for structural materials in the automotive, aerospace and electronic industries. For example, aluminum (Al) and its alloys are conventional lightweight metals which are widely used for structural applications. Copper (Cu) is also an important engineering metal especially in applications requiring high electric properties. Due to the sustainability and conditions of supply that place limitations on the required extensive growth in the consumption of these conventional metals, improvements in their mechanical properties and the development of additional functionalities are now becoming indispensable for enhancing their future impact in the worldwide search for critical resources[1].

In the Materials Science community, one of the promising developments over the last three decades is microstructural refinement and nanostrcuturing that may be achieved through the application of severe plastic deformation (SPD) to a wide range of engineering materials[2,3]. The bulk nanostructured materials (BNM) and ultrafine-grained (UFG) metals processed by SPD contain specially arranged grain boundaries leading to the formation of interface-controlled materials which generally exhibit significant superior mechanical properties and functionalities[4]. Nevertheless, there is an overall limitation in the ability of nanostructuring where the grain refinement often saturates at a coarser level than the nanoscale in the case of simple engineering metals and alloys having single-phase structures[5]. Consequently, there is a saturation in the maximum achievable mechanical properties in conventional metals and alloys even after SPD processing.

Among numerous examples of reported SPD techniques, one of the most effective methods for achieving grain refinement is through the use of high-pressure torsion (HPT) processing where a bulk metal, generally in a disk shape, is severely strained under extreme pressure with concurrent torsional straining[6]. Specifically, when a disk is processed in conventional HPT the equivalent von Mises strain, ε_{eq}, is given by a relationship of the form[7]:

$$\varepsilon_{\mathrm{eq}} = \frac{2\pi N r}{h\sqrt{3}} \qquad (1)$$

where N is the number of torsional turns and r and h are the radius and thickness of the disk sample, respectively, and the resultant shear strain, γ, is given by $\gamma = \sqrt{3}\varepsilon_{\mathrm{eq}}$ [8]. Because of the simple processing procedure that is used to introduce significant microstructural refinement, the HPT processing technique has been utilized for mechanical alloying in the bonding of machining chips and ribbons[9-13] and the consolidation of metallic powders[14-24]. Nevertheless, these procedures generally require high processing temperatures and often two-step processing involving cold/hot compaction of the small metal pieces prior to HPT in order to avoid severe damage to the HPT anvils.

In the last five years, a new approach of applying conventional HPT processing to the bulk-state reaction of engineering metals was studied through the mechanical bonding of dissimilar metal disks for the rapid synthesis of hybrid alloy systems at room temperature (RT). The concept behind this approach is to form light-weight alloys by maintaining constant or further lowering the material density and by extending the upper limit of mechanical properties, such as strength and ductility, during severe microstructural refinement and concurrent solid-state bonding in HPT. In practice, several different conditions of sample set-up were tried in a series of preliminary studies of the processing approach and recently a fixed procedure has been established by demonstrating unique microstructural and structural formation in several different metal combinations.

Accordingly, this paper is designed to review the procedures of the reported mechanical bonding of dissimilar metals by HPT and to summarize the developments in microstructure and mechanical properties of the synthesized hybrid alloy systems. Thus, the next two sections present an overview of the mechanical bonding of metals by HPT, the severe mixture of dissimilar metals and the formation of metal-matrix nanocomposites (MMNCs) during HPT processing. The following section describes the improved physical and mechanical properties of the synthesized hybrid alloy systems including density, hardness, specific strength, and plasticity. The final section before the conclusions describes the critical parameters needed to promote the mechanical bonding by HPT and the feasibility of the synthesis of metastable bulk nanostructured alloys by HPT-induced mechanical boning.

2. Mechanical bonding of dissimilar metals and alloys by HPT

Unlimited combinations of metals and alloys are possible for the mechanical bonding of dissimilar metals. Processing by clad-rolling[25,26] was developed earlier for a single step of roll-bonding at elevated temperatures and this produces a multi-layered plate from dissimilar metals but with micron-scale grains. To further refine the grain size and improve the mechanical properties of alloy sheets, the accumulative-roll bonding (ARB)[27] process has been applied tor the clad-rolling[28]. However, characterizations of the fine-grained metal plates after ARB often reveal anisotropic plastic behavior which varies significantly with the rolling direction and the through-thickness direction[29]. An earlier summery displayed a map as shown in **Figure 1**[30] of successfully bonded metal combinations for clad-bonding by cold rolling and/or ARB. It is apparent that *f.c.c.* metals are well studied and often easier to cold-bond since these metals exhibit less rapid work hardening than metals having other lattice structures. It is noted also that, for the processes involving rolling procedures which generally provide less straining than HPT, good bonding of dissimilar metals can occur only when the surface films or contaminations are removed properly by scratch brushing of the surfaces before roll-bonding in order to provide direct contact between the metals and thereby utilize a better operation of the interatomic attractive forces. Nevertheless, there are relatively limited reports of the successful cold-roll bonding of separate metals and, as will be described in the present review, a more powerful mechanical bonding of dissimilar metals is now feasible for the synthesis of hybrid metal systems through the application of HPT.

The mechanical bonding of dissimilar metals was reported for HPT by applying several different sample set-ups. One of the first reports demonstrated the solid-state reaction of Al and Cu samples which were in the form of semi-circular disks by placing them between the HPT anvils to delineate a complete circular disk as shown in **Figure 2**(a)[31]. The deformed samples of the Al-Cu disk before and after 100 turns by HPT at 6.0 GPa are shown in Figure 2(b) and the processed disk demonstrated the mixture of nano-scale Al- and Cu-rich phases involving two intermetallic compounds of Al_2Cu and Al_4Cu_9. Another study of a solid-state reaction using Al and Cu was reported specifically for the architecturing of a metal composite structure and texture[32]. Thus, **Figure 3** shows (a) a schematic drawing of the initial quarter disks and (b) the sample set-up applying four quarter-circular disks of an Al-6061 alloy and pure Cu placed on the lower HPT anvil and processed at 2.5 GPa for 1 turn[32]. The computationally modeled disk is shown in Figure 3(c) and the calculated equivalent strain in the processed disk was estimated in the spiral hybrid texture as displayed in Figure 3(d).

Following these studies, a simpler procedure was introduced for HPT mechanical bonding by initially aiming to produce possible protective coating layers[33,34] and then by forming multi-layered nanocrystalline microstructures through the direct stacking of dissimilar metal disks without any surface brushing treatment. A first example was shown by the direct bonding of separate Al and Mg disks by HPT for up to 5-10 turns[35-37] and up to 20 turns[38] under 6.0 GPa at RT where the disks were stacked alternately in the order of Al/Mg/Al without any adhesive treatments as shown in **Figure 4** [35]. This procedure of HPT processing was further applied for several different metal combinations by stacking two disks of Al/Mg[39] and three disks of Al/Cu/Al[40], Al/Ti/Al and Al/Fe/Al[41], Cu/Al/Cu[42], Cu/ZnO/Cu[43], Zn/Mg/Zn[44], Fe/V/Fe[45,46], V-10Ti-5Cr/Zr-2.5Nb/V-10Ti-5Cr[47].

It should be noted that some of these listed combinations of dissimilar metals for the mechanical bonding by HPT use different sample volume by changing the disk thicknesses. This approach follows the successful cladding of Al and Cu plates by ARB when a thinner plate of hard Cu was placed between the thicker plates of soft Al[28]. Moreover, as shown by the schematic sample set-up in **Figure 5**, the mechanical bonding approach by HPT was further extended for the bonding of alternately stacked 19 Cu and 18 Ta thin foils to introduce a bulk hybrid alloy system[48]. This series of demonstrations confirms the feasibility and potential for using the unique HPT procedure for synthesizing hybrid alloy systems from conventional metals.

3. Structural evolution and the formation of metal-matrix nanocomposites

3.1 General microstructural evolution

When dissimilar metal disks are mechanically bonded by HPT as shown in Figure 4, the separate metal disks are generally well bonded without any visible segregations and the continuous interface of the dissimilar metal phases is clearly visible throughout the disk diameter when a vertical cross-section of the disk is revealed after the initial compression stage and in the very early stage of HPT for ~1 turn. A representative example is shown in **Figure 6**(a)[43] where a set of Cu/ZnO/Cu disks were compressed under 5 GPa without any torsional straining. The overall microstructure and some focused regions are shown in Figure 6(b) and (b1)-(b4), respectively, for the Cu-ZnO system after HPT for 5 turns under 5 GPa. Torsional straining by HPT produced necking and the fragmentation of the ZnO phase across the disk diameter but there was no clear information on any compositional mixture leading to a phase transformation between the Cu and ZnO.

The observation of phase fragmentation without any compositional mixing was also demonstrated when applying HPT to dissimilar metals of Al/Fe/Al after 20 turns under 1.0 GPa and Al/Ti/Al after 50 turns at 6.0 GPa as shown in **Figure 7**(a) and (b), respectively[41], where each alloy system is described by the cross-sectional microstructure (upper), a color-coded hardness contour map (lower) and an X-ray diffraction (XRD) profile taken at the disk edge (on right). The micrographs together with the hardness maps demonstrate the clear separation of two dissimilar metal phases at the disk centers at r <3-4 mm, while the hardness values are higher as Hv ≈ 330 and ~350 in the Al-Fe and Al-Ti systems, respectively, at the disk peripheries. The recorded high hardness at the edge of the mechanically-bonded disk is consistent with the saturation hardness values of Fe and Ti after grain refinement by HPT for 4 or higher turns when they are processed separately[49]. The XRD profiles for the mechanically-bonded Al-Fe and Al-Ti systems imply the presences of separate Al, Fe and Ti phases without any compositional mixture thereby nucleating new intermetallic compounds in these metal systems after HPT. It should be noted that the mechanical alloying and the consolidation process of Al and Ti powders by HIP[50] and HPT[51] demonstrated the formation of nano-scale intermetallic phases. The nucleation of intermetallic phases in these earlier presentations was reasonable due to the much shorter available diffusion lengths for the nucleation of such new phases by powder metallurgical processes by comparison with the HPT-induced mechanical bonding for the bulk reaction of metals.

It is worth noting that the vicinity of the metal interfaces under the HPT-induced mechanical bonding demonstrates unique flow patterns similar to those anticipated in the flow of liquids as shown in the earlier examples in Figure 7. This flow appears when a Kelvin-Helmholtz shear instability[52,53] leads to vortices at the interfaces under shear or sliding deformation. The specific region involving vortices adjacent to the interfaces denotes *tribomaterials*[54]. Thus, the interfaces in the HPT-induced mechanically-bonded metals receiving severe shear under high pressure introduce large volumes of tribomaterials during the phase mixing. A schematic display of the formation of tribomaterials is shown in **Figure 8** [54] which includes a lubricant at the metal interface whereas mechanical bonding through HPT avoids the use of any lubricants and/or special adhesive treatments.

3.2 Formation of metal-matrix nanocomposites

Depending on the metal selections, several studies demonstrated the successful nucleation of nano-scale intermetallic phases by the mechanical bonding of dissimilar metals through HPT. In practice, intensive studies were conducted on the diffusion bonding of Al/Mg/Al and **Figure 9** describes the structural and hardness evolution of the Al-Mg mechanical bonding by HPT under 6 GPa for 1, 5, 10, 20, 40 and 60 turns. Specifically, a series of micrographs taken at the cross-sectional planes are shown in Figure 9(a)[35,38,55] where the dark phase represents the Mg-rich and the bright phase represents the Al-rich phase, the hardness variation at the disk diameter for the corresponding disks are shown in Figure 9(b)[55] where the dotted horizontal lines are the reference saturation hardness values observed for the Al-1050 [56] and ZK60 [57] alloys after HPT for 5 turns and an XRD peak profile is shown in Figure 9(c) at the disk edge of the Al-Mg system after 20 HPT turns[38].

It is apparent after 1 HPT turn that a multi-layered structure remains throughout the disk diameter with the fragmented Mg layers having thicknesses of ~200 µm and with no visible segregation at the Al-Mg interfaces. After increasing numbers of HPT turns to 5-20, a similar microstructure consisting of multi-layers of the Al and Mg phases was observed at the central regions at r <2.5 mm of the disk after 5 turns and this was reduced to r ≈1.0 mm after 10 and 20 turns. By contrast, the disk edge at r >2.5 mm after 5 turns contains homogeneous distributions of very fine Mg phases with thicknesses of ~5-10 µm to even a true nano-scale of ~100-500 nm within the Al matrix. Further processing to 10-20 HPT turns provided no evidence of visible Mg phases at the disk edges at ~3 < r <5 mm. Although some shear patterns appear as dark lines on the polished surfaces, the disks after 40 and 60 turns retained no visible Mg-rich phases at the disk peripheries. The multi-layered microstructure remained in small regions at the disk centers even after 40 and 60 HPT turns.

The hardness variation in Figure 9(b) shows the achievement of extreme hardness of Hv ≈ 330 at the disk edge after 20 turns and the hardness saturates and is maintained reasonably constant up to 60 turns. The low hardness values which are equivalent to the reference values for the UFG Al and Mg are observed for the Al-Mg system up to 20 HPT turns but there are gradual increases in hardness to Hv ≈ 115 and ~140 after HPT for 40 and 60 turns, respectively. These earlier studies demonstrated extreme hardness increases at the disk edges which resulted mainly from the significant grain refinement with only a small contribution from the formation of hard intermetallic compounds of Al_3Mg_2 and $Al_{12}Mg_{17}$ through the diffusion reaction shown in Figure 9(c) where the XRD analysis followed by Materials Analysis Using Diffraction (MAUD)[58] analysis computed the presence of $Al_{12}Mg_{17}$ at the disk edge of the Al-Mg system after HPT for 20 turns[38].

A similar behavior for the formation of intermetallic phases by mechanical bonding by diffusion beyond the complex metal mixture was shown between Al and Cu when the HPT mechanical bonding was conducted on disk stacks of Al/Cu/Al. The cross-sectional micrographs and the hardness variation along the disk diameters are shown in **Figure 10**(a) and (b), respectively, for the Al-Cu system after HPT for 10, 20, 40 and 60 turns at 6 GPa, and the XRD profiles for the system after 20 and 60 turns are shown in Figure 10(c)[40]. In the micrographs, the Al-rich phase is shown in bright color while the grey region at the disk center correspond to the Cu-rich phase and the dark color at the disk edges relates to a mixture of Al and Cu. As observed in the Al-Mg system, heterogeneous microstructural evolution is observed in the Al-Cu system where a multi-layered microstructure over a wide region from the disk center is present at $r \geq 3$-4 mm and a mixture of very fine Cu-rich phases at the disk edge after 10 HPT turns. Increasing the HPT processing to 60 turns significantly reduces the multi-layered region at the disk centers to $r < 2$ mm and instead there is a widening mixture of Al and Cu in the remaining disk regions at $r > 1$ mm.

These microstructural changes affect the hardness variations across the disk diameter. In practice, Figure 10(b) shows that all disks demonstrate a consistent trend of extreme hardness at the disk edges and low hardness at the disk centers, but the high hardness values at the disk peripheries increase with increasing HPT turns without saturation in the mechanical bonding of Al and Cu until 60 HPT turns under 6 GPa. The increased hardness without saturation is closely associated with the formation of hard intermetallic phases which are observed at the disk edges in the Al-Cu system after HPT for more than 20 turns and the Al-Cu intermetallic compounds of Al_2Cu estimated by XRD and associated MAUD analysis as shown in Figure 10(c). Thus, the total contents of the intermetallic phase increases with increasing numbers of HPT turns from 20 to 60 in the Al-Cu system.

The nucleation of intermetallic compounds with increasing numbers of HPT turns was also observed by XRD analysis in mechanically-bonded pure Mg and Zn with the disk stacking order of Zn/Mg/Zn through HPT processing. **Figure 11** shows the XRD line profile for the Zn-Mg system after HPT for 1, 15 and 30 turns[44]. In this study, newly appearing X-ray diffraction peaks indicate the presence of two intermetallic phases of Mg_2Zn_{11} and $MgZn_2$ in the Zn-Mg system with increasing numbers of HPT turns. Consequently, the processing of dissimilar metals by HPT demonstrates the successful mechanical bonding of the separate metals by the aid of diffusion and the formation of unique microstructural characteristics involving heterogeneous distributions of phases and their sizes. Moreover, the synthesized alloy systems often show extreme hardness at the disk edges attributed to an intensive mixture of the metallic phases and the formation of intermetallic compounds. Specifically, studies demonstrating a formation of nano-scale intermetallic phases defined the synthesized hybrid alloy systems as metal-matrix nanocomposites (MMNCs).

A critical factor for the formation of MMNCs during the mechanical bonding is an enhanced atomic diffusion of metallic atoms during HPT processing. Several earlier studies demonstrated experimental evidence for accelerated atomic diffusion by equal-channel angular pressing in a Cu-Pb alloy[59] and pure Ni[60]. The fast atomic diffusion was attributed to the introduction of extra free volumes by the excess numbers of lattice defects during nanostructuring by SPD processing. This proposal was also developed in an earlier review by noting the significance of the fast atomic mobility by acknowledging the increase in the vacancy concentration in UFG materials processed by SPD processing[61].

3.3 Severe mixture of dissimilar metals

A consistent finding of the rapid atomic diffusion due to the presence of intense numbers of vacancies, dislocations and grain boundaries was to calculate the diffusion length associated with the atoms needed for nucleating the intermetallic phases during the solid-state reaction of the semi-circular Al and Cu disks as shown in Figure 2[31]. Specifically, it was estimated that the diffusion coefficients were ~10^{12}-10^{22} times higher than lattice diffusion for the Al-Cu system during nanostructuring as well as the formation of intermetallic phases through HPT, and these estimated values are comparable to surface diffusion of the alloy system. **Figure 12** displays the change of diffusion coefficient against the Cu concentration in Al[31], where it visualizes that the estimated diffusion coefficients for the Al-Cu system mechanically-bonded by HPT are reasonably consistent with those for surface diffusion of the corresponding compositions. Thus, it is concluded that HPT-induced mechanical bonding is a powerful processing technique for severe metal mixing at RT.

Such severe mixture of the dissimilar metal phases during mechanical bonding by HPT are visualized in macro-scale micrographs as shown in Figures 7, 9 and 10. The regions of severely mixed phases appear at the disk edges in general and they extend with increasing numbers of HPT turns as is evident in the Al-Mg and Al-Cu systems in Figures 9 and 10, respectively. Taking a boundary radius, r_b, which is the distance from the disk center dividing the multi-layered structure from the severe mixture of metal phases at the disk edges, then **Figure 13** reveals a relationship between r_b and the estimated equivalent strain and the shear strain computed using equ. (1) for both mechanically-bonded Al-Mg[35,38,55] and Al-Cu[40] systems after HPT at 6.0 GPa[55]. It is apparent that there is an approximate linear relationship between r_b and strain for both alloy systems while the rate of change in r_b depends on the selection of dissimilar metals. This analysis implies, therefore, that there is a possibility to expand the regions with severe mixing of dissimilar metal phases in order to achieve homogeneous microstructure within the entire metal volume when applying much higher strains by HPT.

4. Advanced properties of the mechanically-bonded alloy systems after HPT

4.1 Strength-to-weight ratio

In terms of the structure and physical characteristics, the unique features of the mechanical bonding of dissimilar metals through HPT is that the processed bulk alloys are not the same as the base materials after HPT. Thus, by applying a lightweight metal as one of the dissimilar metals, the mechanical bonding process enables the processed alloy systems to demonstrate low densities due to the severe mixing of the metal phases as well as the nucleation of lightweight intermetallic phases in some conditions. Ultimately, because of the grain refinement process by HPT which improves the hardness and strength of the processed material, the mechanically-bonded alloys are anticipated to demonstrate a high strength-to-weight ratio (or specific strength).

Several earlier studies measured the densities of the mechanically-bonded alloys of the Al-Mg[38], Al-Cu[40] and Al-Fe systems[62] and the estimated strength-to-weight ratio for these alloy systems by applying their estimated yield strength by using the measured Vickers microhardness values. A summary of these results is shown in **Table 1** where, for comparison purposes, the tabulation gives the density, Vickers microhardness and specific strength for the base metals of the Al, Mg and Cu alloys after HPT for high numbers of turns. It should be noted that the density measurements for the mechanically-bonded alloys were conducted at the disk edges where the measured volumes and weights are for the severely mixed alloy systems and the forming of MMNCs for Al-Mg and Al-Cu systems.

It is apparent from Table 1 that the densities of the HPT-induced alloys show average or even lower values than the densities of the base metals. Moreover, these values tend to decrease with increasing numbers of HPT turns in the Al-Mg and Al-Cu systems due to the formation of intermetallics phases which reduce the fractions of Mg and Cu phases having higher densities. Due to the lower density but increasing hardness by nanostructuring through HPT, and the formation of hard intermetallic phases in the nanostructured matrix, the estimated specific strength becomes exceptionally high as ~455 MPa cm^3g^{-1} for the Al-Mg after 20 HPT turns, ~390 MPa cm^3g^{-1} for the Al-Cu after 60 HPT turns and ~300 MPa cm^3g^{-1} for the Al-Fe after 20 HPT turns.

In practice, the computed specific strength increases with increasing HPT turns for the Al-Mg and Al-Cu system. Moreover, the estimated specific strength for the HPT-induced alloy systems are significantly higher than the base materials of Al, Mg, Cu and Fe which are strengthening by grain refinement through HPT processing. In fact, the specific strength of the mechanically-bonded alloys are higher than many structural metals including steels which exhibit ~150 MPa cm^3g^{-1} and the engineering polymeric composites, ceramics and carbon fibers demonstrating ~200 MPa cm^3g^{-1} [63]. Thus, it can be concluded that HPT processing gives an excellent opportunity to bond dissimilar metals and further synthesize extreme strength-to-weight ratio alloy systems from conventional engineering metals.

4.2 Hardening mechanisms for the mechanically-bonded MMNCs and hybrid alloys

Extreme hardness was observed at the disk edges of the mechanically-bonded alloys and some results are shown in Figures 7, 9 and 10. An earlier report on the mechanically-bonded Al-Mg system discussed the mechanisms for the increased values of Vickers microhardness of the synthesized MMNCs at the disk edges after 5 and 10 turns[35]. Specifically, the hardness increase was evaluated by a combination of Hall-Petch[64,65] strengthening due to significant grain refinement, solid solution strengthening due to the accelerated diffusivity of Mg into Al-rich phases and precipitation hardening by considering the nucleated intermetallics as precipitates. Thus, the microstructure at the disk edges after HPT was evaluated to examine the grain sizes by TEM analysis and Mg contents and the phase fractions of intermetallic phases by XRD and MAUD analyses. These results were used to estimate each hardness contribution increase as separate and discrete effects and **Figure 14** displays the estimated total Vickers microhardness values from the three different strengthening mechanisms with increasing Mg content in an Al solid solution after HPT for 10 turns[35]. In practice, the ~5 wt.% of Mg in Al and the phase fraction of <0.5% of Al-Mg intermetallic phases, and the matrix grain and intermetallic phase sizes of 20 nm and 30 nm, respectively, give an estimate of Hv ≈ 269 ± 8. This value is in excellent agreement with the experimental value taken at the disk edge for the Al-Mg system after HPT for 10 turns as shown in Figure 9(b). Consequently, the evaluation confirms the simultaneous occurrence of these strengthening mechanisms that are available due to SPD at a low processing temperature which precludes the introduction of significant microstructural recovery.

In practice, defining the strengthening mechanisms for the complicated microstructure in the mechanically-bonded nanocrystalline metal systems and MMNCs is not a trivial task due to the complexities of their microstructures. For example, thin layers of the intermetallic compounds, as observed in the mechanically-bonded Al-Mg[35,38] and Al-Cu[31,40] systems, may strengthen the materials by the load transfer (load bearing) effect occurring between the soft and compliant matrix to the stiff and hard particles under an applied external load[66,67] instead of by precipitation strengthening. However, the specific experimental parameters and results provide a negligible effect on the load bearing to the Al-Mg system[35]. It is also supported by an earlier report where several Mg nanocomposites, reinforced by less than 5 vol.% of either Al_2O_3 or Y_2O_3, showed that load-bearing strengthening is too small to be considered as a significant contributor to the overall strength [68].

Moreover, a difference in the coefficient of thermal expansion (ΔCTE) between the matrix and the reinforcement is also an important strengthening mechanism for composite materials since it leads to the generation of dislocations at the interfaces[69-71]. The contribution of ΔCTE to the total strength is produced when there is a quenching effect with a temperature change during the solution treatment or processing, and composite materials including ceramic particulates such as SiC generally show large ΔCTE. However, the intermetallic-based Al MMNCs synthesized by mechanical-bonding though HPT provides a significantly small ΔCTE between the Al matrix and the small content of intermetallic compounds estimated by the rule of mixtures. It is reported that for composites in which the processing is at RT then the Orowan strengthening has higher values than the CTE mismatch effect, while for those with a processing temperature higher than RT (as at ~300°C) the CTE mismatch effect becomes more significant than Orowan strengthening[68].

Finally, within the limited reports determining the strengthening mechanisms of MMNCs, it is suggested that Orowan strengthening plays a significant role in the strengthening of MMNCs when very fine (~100 nm) insoluble particles are present in a metal matrix[68,72-74]. Also, reductions in both the diameter and the interparticulate spacing of the fine second phase influences the strength of MMNCs and can be correlated with the increase in the extent of Orowan strengthening[74]. Nevertheless, further studies are needed to provide a more comprehensive understanding of the strengthening mechanisms for different alloy systems bonded mechanically by HPT.

4.3 Micro-mechanical response and plasticity

There is no well-defined method for examining the improved mechanical properties at a selected region of interest within the HPT samples where these samples are often small with a diameter of 10 mm. Especially, the mechanically-bonded metal systems processed by HPT demonstrate significant heterogeneities in their microstructures across the disk diameters as seen in Figures 7, 9 and 10. Thus, the novel technique of nanoindentation provides a useful testing procedure[75] for detecting and understanding the plastic yield at the incipient plasticity focusing on the very early stages of deformation where the transition of elastic to plastic flow occurs using a small volume of the sample[76]. In practice, nanoindentation was applied for a wide variety of metallic materials processed by different SPD techniques to understand their micro-mechanical responses by measuring and computing hardness and strain rate sensitivity of the nanostructured materials[75].

A series of nanoindentation measurements were reported at the disk edges[35,38] and centers[77] on mechanically-bonded Al-Mg systems after HPT up to 20 turns. As was expected from the increased hardness at the disk edges of the Al-Mg system whose strengthening mechanisms were explained in an earlier section, the results taken by nanoindentation demonstrated an increased hardness but reduced plasticity by computing the decreased strain rate sensitivity, m, with increasing numbers of HPT turns. One approach to improve the reduced plasticity was demonstrated by applying a post-deformation annealing (PDA) on the mechanically-bonded Al-Mg system after 20 turns[38]. The PDA treatment provided a reasonable amount of microstructural relaxation for enhancing plasticity by compromising the hardness of the MMNC structure.

For the mechanically-bonded Al-Cu system, increasing values of strain rate sensitivity for up to 60 HPT turns are demonstrated as shown in **Figure 15** [55] while the hardness also increases at the disk edges after HPT though 60 turns as shown in Figure 10. In practice, the strain rate sensitivity of $m \approx 0.03$ was estimated at the disk edge of the Al-Cu system after HPT for 20 turns. However, due to the plastic instability attributed to less mixture of Al and Cu phases, wide error bars were observed from 15 or more measurements at each nanoindentation strain rate. Thus, the m value for the Al-Cu disk after 20 HPT turns is induced by the m values of ~0.02 and ~0.04 for the base metals of Al and Cu after HPT for 10 turns, respectively. By contrast, the HPT-induced mechanical bonding after 60 turns introduced an MMNC at the disk edge in the Al-Cu system so that the alloy system acquired an improved m value of ~0.08 with narrower error bars than after 20 turns. The results demonstrate a significant enhancement in the strain rate sensitivity, and thus a potential for achieving improved ductility, on the MMNC at the disk edge of the Al-Cu system with increasing numbers of HPT turns.

In order to fully visualize the significant improvement of the *m* value on the HPT-synthesized MMNC in the hybrid Al-Cu system, it is reasonable to compare the estimated *m* values for this system with the available data for various UFG Al and UFG Cu samples processed by different SPD techniques. **Figure 16** shows the variation of the *m* value with grain size for UFG Al[78-98], UFG Cu[89,99-115] and for the HPT-synthesized hybrid Al-Cu system. The encircling ovals provide a simple visualization of the general trends for the UFG Al and Cu and for the hybrid Al-Cu system. It is recognized from inspection of these ovals that, although there are a few points lying outside of the ovals, there is a consistent trend of enhanced strain rate sensitivity with grain refinement through SPD processing in both the UFG Al and UFG Cu. In practice, an increase in the *m* value with a reduction in grain size in Al and Cu is well summarized in a recent review on the enhancement in strength and ductility in terms of the micro-mechanical behavior analyzed using nanoindentation in various UFG metals and alloys after SPD[75]. By contrast, the Al-Cu system consisting of an MMNC shows excellent improvement in the *m* value with a very significant microstructural refinement capability, as indicated by the arrow in Figure 16 for increasing HPT turns. Together with the high hardness as shown in Figure 10(b), the observed excellent plastic response at the disk edges of the Al-Cu system leads to the conclusion that the mechanical bonding and concurrent grain refinement through HPT is an excellent strategy for fabricating lightweight hybrid metal systems and with forming a variety of MMNCs which show extraordinary physical and mechanical properties.

5. Feasibility of HPT-induced mechanical bonding

5.1 Numbers of disks and metal varieties for severe phase mixtures

It is well defined in numerous fundamental studies on HPT processing that the critical processing parameters of HPT include compressive pressure[116] and the numbers of torsional turns[117,118]. These parameters are also important for HPT-induced mechanical bonding of dissimilar metals and the importance of torsional turns is indicated in Figure 13. In addition to these parameters, the numbers of stacked disks is a unique parameter providing a difference in the mechanically-bonded materials. In practice, mechanical bonding by HPT can be applied from upwards of two disks of dissimilar metals such as Al/Mg[39].

A recent study showed a preliminary result on the influence of the number of stacked disks to the evolution of microstructure at the macro-scale on the Al-Mg system. A total of five disks of Al and Mg, stacked in the order of Al/Mg/Al/Mg/Al, were processed by HPT under 6.0 GPa for 10 turns and **Figure 17** shows (a) schematic illustrations of the different sample set-ups and (b) a comparison of the macro-scale microstructures with five disk stacking (upper) and three disk stacking (lower) for the Al-Mg system under 6.0 GPa after HPT for 10 and 20 turns, respectively [41]. It should be noted that each separate disk for both the five and three stacking set-ups used consistent disk diameters and thicknesses of 10 and 0.8 mm, respectively.

The microstructural appearance at the cross-sections shows that both disks include a severe and complex mixture of Al and Mg phases at the disk edges with $r_b \approx 2.0$ mm and there is a reasonable similarity in the microstructural evolution. Thus, this preliminary study provides information on the effect of large numbers of alternatively stacked dissimilar metal disks for achieving a rapid metal mixture. A faster rate of diffusivity of Al and Mg can be achieved during the HPT-induced mechanical bonding when more disks, thus having a thicker initial total thickness, are prepared for the HPT processing. This mainly results in the failure of maintaining the initial large volume of the set of disks during processing by HPT as is evident from Figure 17(b) where the five disk stacking has a reasonably similar final sample thickness as the three disk stacking. High volumes of samples are compressed and flow out between the anvils to provide a higher compressive pressure when larger numbers of disks are processed simultaneously.

In this respect, applying mechanical bonding by HPT is feasible also for more than two dissimilar metals. A recent trial on the bonding and mixing of 5 different commercial purity metals of Al, Mg, Cu, Fe and Ti is shown in **Figure 18** with (a) a schematic drawing of the sample stacking in the order of Al/Mg/Cu/Fe/Ti/Al and (b) cross-sectional micrographs of the alloy system after HPT under 2.0 GPa for 1 and 33 turns, respectively. The micrographs show that after 1 HPT turn there is a clear space at the Mg/Cu interface and segregation at the Cu/Fe interface. These separations and segregations around the boundaries for the Cu-rich phase were eliminated at the macro-scale after 33 HPT turns. Moreover each separate metal phase necked during the mechanical bonding so that all interphases of the dissimilar metal phases became wavy across the disk diameter whereas no severe phase mixture was visible at the disk edge. Further analyses are now needed to understand the nature of the metal mixing, the required amount of torsional straining and the preferred metal stacking when more than two different kinds of dissimilar metals are used. Nevertheless, this mechanical bonding raises a potential for using HPT processing in the synthesizing of new types of metal systems.

5.2 Scale-up of the HPT-induced mechanical bonding

When evaluating the capability of the mechanical bonding of dissimilar metals by HPT, it is important to examine the possibility of scaling up the sample size. A recent report provided a guideline where the rotational speed during HPT should be reduced for large disk samples in order to minimize heat generation from friction with the anvils[119]. Considering the torsional speed, a recent study processed three disks of Al and Mg having a diameter of 25.0 mm and a thickness of 2.5 mm in each disk in the order of Al/Mg/Al for 10 and 20 turns under 1.0 GPa at 0.4 rpm. This rotational speed was selected to apply a consistent shear strain rate at r = 12.5 mm with 1 rpm at r = 5.0 mm for the 10 mm diameter disk. The micrographs taken at the vertical cross-sections and the corresponding hardness variations are shown in **Figure 19** for the scale-up of disks having 25 mm diameter after HPT for 10 turns (upper) and 20 turns (lower) [120] and these may be compared with the microstructure and hardness for the mechanically-bonded Al-Mg system having a 10 mm diameter as shown in Figure 9(a) and (b); the bright phase denotes the Al-rich phase and the dark phase denotes the Mg-rich phase in Figure 19.

It is apparent that the final thickness of the processed alloy is ~2.5 mm which is one-third of the total height of the initial sample set-up for processing, but a consistent ratio of thickness reduction is observed by the 10 mm diameter disk sample set-up. Microstructural evolution at the disk edges of the 25 mm diameter disk shows small to large fragmented Mg phases after 10 turns and these phases are even smaller after 20 turns. Thus, the observed severe phase mixture at the disk edges is also consistent with the 10 mm diameter disk set-up. On the contrary, the disk center in the 25 mm diameter disk showed less or almost no Mg phases which generally remain at the disk centers in the disks having 10 mm diameter after high numbers of HPT turns to 60 as in Figure 9(a). However, large Mg phases are apparent at the mid-radius of the 25 mm diameter Al-Mg disks after 10-20 turns and a series of analyses by micro-XRD and TEM provided no evidence for the formation of intermetallic compound phases at the disk centers, the mid-radii positions or the edges of the scale-up of the Al-Mg disks[120]. Therefore, it is difficult to consider the complete dissolution of Mg atoms only at the disk center. A high hardness of Hv ≈ 250 was observed within a limited region of the disk periphery after 20 HPT turns and it was anticipated from the small grains with an average grain size of ~380 nm at the disk edge[120]. Overall, the mechanical bonding of the scaled-up Al and Mg disks of 25 mm diameter was successful and slower but nevertheless there was a microstructural evolution that was reasonably consistent with the evolution observed in disks having diameters of 10 mm.

One critical requirement for the scaling up of the SPD-processed samples is that the processing facility must have a capability to apply a sufficiently severe hydrostatic pressure, thereby creating a high compressive pressure in the case of HPT processing. The above discussed experiments on the 25 mm diameter disks were conducted under a compressive pressure of 1.0 GPa due to the use of a laboratory-scale HPT facility. Additional experiments are now required to further understand the influence of compressive pressure on the mechanical bonding of these large-scale samples.

5.3 Formation of bulk metastable alloys

This last section describes recent findings regarding the feasibility of using the HPT-induced mechanical bonding to produce not only hybrid materials with intermetallic phases but also the formation of amorphous phases and a bulk metastable state. In practice, the HPT-induced mechanical bonding of vanadium and Zr alloys in the order of V-Ti-Cr/Zr-Nb/V-Ti-Cr demonstrated a partial formation of amorphous phases in the Zr alloy layers within the multi-layered V alloy and Zr alloy after HPT for 5 turns under 6 GPa[47]. A high-resolution TEM micrograph showing the amorphous phase in the Zr alloy phase is shown in **Figure 20**[47]. In addition, ultra-severe plastic deformation of HPT for 1200 turns led to the formation of amorphous phases in several powder mixtures of Mg immiscible alloys[121], but the locations, sizes and the volume fractions of the amorphous phases were not provided in these reports and the structural homogeneity/heterogeneity is not clear.

By contrast, a recent report on the continuous HPT processing to an efficient amount of high shear strain demonstrated, for the first time, a bulk nanostructured metastable Al alloy in a supersaturated solid solution state[122]. Specifically, a set of Al and Mg disks arranged with the stacking order of Al/Mg/Al was mechanically bonded by HPT for 100 turns under 6.0 GPa at RT. The cross-sectional microstructure and the TEM micrographs taken at the disk edge and center as well as the corresponding hardness distribution at the cross-section are shown in **Figure 21**(a) for the synthesized Al-Mg alloy disk after 100 HPT turns. It is apparent that this high level of straining during the mechanical bonding synthesized a homogeneous microstructure throughout the entire volume of the Al-Mg disk and the TEM micrographs confirm consistent equiaxed grain distributions with average grain sizes of ~35-40 nm at both measured locations. The homogeneous nanostructure yielded a uniform distribution of Vickers microhardness of ~350-370 that is, by comparison with the alloy system with lower numbers of turns as shown in Figure 9(b), a record high hardness achieved in the Al-Mg alloy system synthesized by HPT mechanical bonding.

Figure 21(b) shows the XRD profiles of the mechanically-bonded Al-Mg system after HPT for 100 turns and HPT processing followed by natural aging for 60 turns to evaluate the structural stability: also included are reference XRD profiles of as-received Al and Al after HPT for 5 turns[122]. This evaluation provides an interesting finding that the Al-Mg system after 100 turns and additional annealing shows *f.c.c.* peaks for Al without any peaks for Mg or intermetallic phases, while the material volume remains unchanged after 20 HPT turns. Further analysis of the XRD results using MAUD quantified the supersaturated solubility of Mg in the Al matrix with a maximum of ~38.5 at.% and an average of ~15 at.% all over the disk. Thus, this study demonstrated the unique production of a uniform nanocrystalline microstructure having excellent structural stability of a bulk nanocrystalline metastable Al-Mg alloy with a heterogeneous supersaturated solid solution state when Al and Mg are mechanically bonded under severe plastic straining at ambient temperature.

These reported studies on the mechanical bonding of dissimilar metals by HPT provide significant developmental opportunities for use in solid-state recycling techniques[123] and excellent contributions to current manufacturing techniques in diffusion bonding, welding and mechanical joining.

6. Summary and conclusions

1. Mechanical bonding of dissimilar engineering metals has been studied by utilizing conventional HPT processing for introducing unique alloy systems at room temperature. Currently, a variety of metal combinations are studied using a procedure of stacking dissimilar metal disks directly without any adhesive treatment.

2. Using two dissimilar metals, all HPT-induced mechanically-bonded alloy systems bonded successfully without any segregations. Some selected metal combinations demonstrated the formation of metal matrix nanocomposites by nucleating nano-scale intermetallic compound phases in the nanocrystalline metal matrix.

3. This HPT processing decreased the density of lightweight alloys and improved the hardness leading to exceptional specific strength at the disk edges of the alloy systems introduced by the HPT-induced mechanical bonding.
4. The excellent strength of these mechanically-bonded alloys is due mainly to the significant grain refinement with the additional accommodation of several factors including solid solution strengthening and precipitation hardening.
5. The use of HPT-induced mechanical bonding may be promoted by selecting optimal numbers of stacking disks in addition to high compressive pressures and large numbers of turns. The results from this processing show the feasibility of introducing amorphous phases and a bulk metastable nanostructured material.
6. Accordingly, the results demonstrate there is a considerable potential for making use of the mechanical bonding of dissimilar metals both as a manufacturing technique and for the development of new and functionally significant hybrid nanomaterials.

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Conflict of Interest

The authors declare no conflict of interest.

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References

[1] O. Vidal, F. Rostom, C. François, G. Giraud, *Elements* **2017**, *13(5)*, 319.

[2] R.Z. Valiev, Y. Estrin, Z. Horita, T.G. Langdon, M.J. Zehetbauer, Y.T. Zhu, *JOM* **2006**, *58(4)*, 33.

[3] R.Z. Valiev, Y. Estrin, Z. Horita, T.G. Langdon, M.J. Zehetbauer, Y.T. Zhu, *JOM* **2016**, *68*, 1216.

[4] R.Z. Valiev, Y. Estrin, Z. Horita, T.G. Langdon, M.J. Zehetbauer, Y.T. Zhu, *Mater. Res. Lett.* **2016**, *4,* 1.

[5] R. Pippan, F. Wetscher, M. Hafok, A. Vorhauer, I. Sabirov, *Adv. Eng. Mater*. **2006**, *8*, 1046.

[6] A.P. Zhilyaev, T.G. Langdon, *Prog. Mater. Sci.* **2008**, *53*, 893.

[7] F. Wetscher, A. Vorhauer, R. Stock, R. Pippan, *Mater. Sci. Eng. A* **2004**, *387-389*, 809.

[8] R.Z. Valiev, Yu.V. Ivanisenko, E.F. Rauch, B. Baudelet, *Acta Mater*. **1996**, *44,* 4705.

[9] J. Sort, D.C. Ile, A.P. Zhilyaev, A. Concustell, T. Czeppe, M. Stoica, S. Suriñach, J. Eckert, M.D. Baró, *Scr. Mater.* **2004**, *50*, 1221.

[10] A.P. Zhilyaev, A.A Gimazov, G.I. Raab, T.G. Langdon, *Mater. Sci. Eng. A*, **2008**, 486, 123.

[11] K. Edalati, Y. Yokoyama, Z. Horita, *Mater. Trans.* **2010**, *51*, 23.

[12] M. Ibrahim Abd El Aal, E.Y. Yoon, H.S. Kim, *Mater. Sci. Eng. A* **2013**, *560*, 121.

[13] M.M. Castro, P.H.R. Pereira, A. Isaac, R.B. Figueiredo, T.G. Langdon, *J. Alloy. Compd.* **2019**, *780*, 422.

[14] A.V. Korznikov, I.M. Safarov, D.V. Laptionok, R.Z. Valiev, *Acta Metall. Mater.* **1991**, *39* 3193.

[15] V.V. Stolyarov, Y.T. Zhu, T.C. Lowe, R.K. Islamgaliev, R.Z. Valiev, *Mater. Sci. Eng. A* **2000**, *282*, 78.

[16] X. Sauvage, P. Jessner, F. Vurpillot, R. Pippan, *Scr. Mater.* **2008**, *58*. 1125.

[17] K. Kaneko, T. Hata, T. Tokunaga, Z. Horita, *Mater. Trans.* **2009**, *50*, 76.

[18] K. Edalati, Z. Horita, H. Fujiwara, K. Ameyama, *Metall. Mater. Trans. A* **2010**, *41A*, 3308.

[19] K. Edalati, Z. Horita, *Scr. Mater.* **2010**, *63*, 174.

[20] A. Bachmaier, M. Kerber, D. Setman, R. Pippan, *Acta Mater.* **2012**, *60*, 860.

[21] J.M. Cubero-Sesin, Z. Horita, *Mater. Sci. Eng. A* **2012**, *558*, 462.

[22] Y. Zhang, S. Sabbaghianrad, H. Yang, T.D. Topping, T.G. Langdon, E.J. Lavernia, J.M. Schoenung, S.R. Nutt, *Metall. Mater. Trans. A*, **2015**, *46A*, 5877.

[23] A.P. Zhilyaev, G. Ringot, Y. Huang, J.M. Cabrera, T.G. Langdon, *Mater. Sci. Eng. A* **2017**, *688*, 498.

[24] Y. Huang, P. Bazarnik, D. Wan, D. Luo, P.H.R. Pereira, M. Lewandowska, J. Yao, B.E. Hayden, T.G. Langdon, *Acta Mater.* **2019**, *164*, 499.

[25] A. Macwan, X.Q. Jiang, C. Li, D.L. Chen, *Mater. Sci. Eng. A* **2013**, *587*, 344.

[26] K.S. Lee, Y.S. Lee, Y.N. Kwon, *Mater. Sci. Eng. A* **2014**, *606*, 205.

[27] Y. Saito, N. Tsuji, H. Utsunomiya, T. Sakai, R.G. Hong, *Scr. Mater.* **1998**, *39*, 1221.

[28] M. Eizadjou, A.K. Talachi, H.D. Manesh, H.S. Shahabi, K. Janghorban, *Comp. Sci. Technol.* **2008**, *68*, 2003.

[29] B. Beausir, J. Scharnweber, J. Jaschinski, H.-G. Brokmeier, C.-G. Oertel, W. Skrotzki, *Mater. Sci. Eng. A* **2010**, *527*, 3271.

[30] L. Li, K. Nagai, F. Yin, *Sci. Technol. Adv. Mater.* **2008**, *9*, 023001.

[31] K. Oh-ishi, K. Edalati, H. S. Kim, K. Hono, Z. Horita, *Acta Mater.* **2013**, *61*, 3482.

[32] O. Bouaziz, H. S. Kim, Y. Estrin, *Adv. Eng. Mater.* **2013**, *15,* 336.

[33] X. Li, W. Liang, X. Zhao, Y. Zhang, X. Fu, F. Liu, *J. Alloys Compd*. **2009**, *471*, 408.

[34] K. Spencer, M.-X. Zhang, *Scr. Mater*. **2009**, *61*, 44.

[35] B. Ahn, A.P. Zhilyaev, H.-J. Lee, M. Kawasaki, T.G. Langdon, *Mater. Sci. Eng. A* **2015**, *635*, 109.

[36] B. Ahn, H.-J. Lee, I.C. Choi, M. Kawasaki, J.-I. Jang, T.G. Langdon, *Adv. Eng. Mater.* **2016**, *18*, 1001.

[37] M. Kawasaki, B. Ahn, H.-J. Lee, A.P. Zhilyaev, T.G. Langdon, *J. Mater. Res.* **2016**, *31*, 88.

[38] J.-K. Han, H.-J. Lee, J.-i. Jang, M. Kawasaki, T.G. Langdon, *Mater. Sci. Eng. A* **2017**, *684*, 318.

[39] X. Qiao, X. Li, X. Zhang, Y. Chen, M. Zheng, I.S. Golovin, N. Gao, M.J. Starink, *Mater. Lett.* **2016**, *181*, 187.

[40] J.-K. Han, D.K. Han, G.Y. Liang, J.-i. Jang, T.G. Langdon, M. Kawasaki, *Adv. Eng. Mater.* **2018**, *20*, 1800642.

[41] M. Kawasaki, J.-K. Han, D-.H. Lee, J.-i. Jang, T.G. Langdon, *J. Mater. Res.* **2018**, *33*, 2700.

[42] V.N. Danilenko, S.N. Sergeev, J.A. Baimova, G.F. Korznikova, K.S. Nazarov, R.Kh. Khisamov, A.M. Glezer, R.R. Mulyukov, *Mater. Lett*. **2019**, *236*, 51.

[43] Y. Qi, A. Kosinova, A.R. Kilmametov, B.B. Straumal, E.Rabkin, *Mater. Charact.* **2018**, *145*, 389.

[44] D. Hernández-Escobar, Z.U. Raman, H. Yilmazer, M. Kawasaki, C.J. Boehlert, *Phil. Mag*. **2019**, *99*, 557.

[45] S.O. Rogachev, R.V. Sundeev, V.M. Khatkevich, *Mater. Lett.* **2016**, *173*, 123.

[46] S.O. Rogachev, S.A. Nikulin, A.B. Rozhnov, V.M. Khatkevich, T.A. Nechaykina, M.V. Gorshenkov, R.V. Sundeev, *Metall. Mater. Trans. A* **2017**, *48A*, 6091.

[47] S.O. Rogachev, R.V. Sundeev, N.Yu. Tabachkova, *Mater. Lett.* **2019**, *234*, 220.

[48] N. Ibrahim, M. Peterlechner, F. Emeis, M. Wegner, S.V. Divinski, G. Wilde, *Mater. Sci. Eng. A* **2017**, *685*, 19.

[49] K. Edalati, Z. Horita, *Mater. Trans.* **2010**, *51*, 1051.

[50] Z. Lee, S.R. Nutt, R. Rodriguez, R.W. Hayes, E.J. Lavernia, *Metall. Mater. Trans. A* **2003**, *34A*, 1473.

[51] K. Edalati, S. Toh, H. Iwaoka, M. Watanabe, Z. Horita, D. Kashioka, K. Kishida, H. Inui, *Scr. Mater.* **2012**, *67*, 814.

[52] W. Kelvin, *Philos. Mag.* **1871**, *42*, 362.

[53] H.L.F. von Helmholtz, *Philos. Mag.* **1868**, *36*, 337.

[54] D.A. Rigney, S. Karthikeyan, *Tribol. Lett.* **2010**, *39*, 3.

[55] M. Kawasaki, S.-H. Jung, J.-M. Park, J. Lee, J.-i. Jang, J.-K. Han, *Adv. Eng. Mater.* **2019** (in press). DOI: 10.1002/adem.201900483

[56] M. Kawasaki, S.N. Alhajeri, C. Xu, T.G. Langdon, *Mater. Sci. Eng. A* **2011**, *529*, 345.

[57] H.-J. Lee, S.K. Lee, K.H. Jung, G.A. Lee, B. Ahn, M. Kawasaki, T.G. Langdon, *Mater. Sci. Eng. A* **2015**, *630*, 90.

[58] L. Lutterotti, *Nucl. Instrum. Methods Phys. Res. Sect. B* **2010**, *268*, 334.

[59] S.V. Divinski,, J. Ribbe, D. Baither, G. Schmitz, G. Reglitz, H. Rösner, K. Sato, Y. Estrin, G. Wilde, *Acta Mater.* **2009**, *57*, 5706.
[60] S.V. Divinski, G. Reglitz, H. Rösner, Y. Estrin, G. Wilde, *Acta Mater.* **2011**, *59*, 1974.

[61] X. Sauvage, G. Wilde, S.V. Divinski, Z. Horita, R.Z. Valiev, *Mater. Sci. Eng. A* **2012**, *540*, 1.

[62] J.-K. Han, J.-i. Jang, T.G. Langdon, M. Kawasaki, *Mater. Trans.* **2019**, *60*, 1131.

[63] K. Lu, *Science* **2010**, *328*, 319.

[64] E.O. Hall, *Proc. Phys. Soc. B* **1951**, *64*, 747.

[65] N.J. Petch, *J. Iron Steel Inst.* **1953**, *174*, 25.

[66] S.M. Uddin, T. Mahmud, C. Wolf, C. Glanz, I. Kolaric, C. Volkmer, H. Höller, U. Wienecke, S. Roth, H. Fecht, *Comp. Sci. Technol.* **2010**, *55*, 42.

[67] R. Casati, M. Vedani, *Metals* **2014**, *4*, 65.

[68] A. Sanaty-Zadeh, *Mater. Sci. Eng. A* **2012**, *531*, 112.

[69] R.J. Arsenault, *Mater. Sci. Eng.* **1986**, *81*, 175.

[70] R.J. Arsenault, L. Wang, C.R. Feng, *Acta Metall. Mater.* **1991**, *39*, 47.

[71] W.S. Miller, F.J. Humphreys, *Scripta Metall. Mater.* **1991**, *25*, 33.

[72] Z. Zhang, D.L. Chen, *Scripta Mater.* **2006**, *54*, 1321.

[73] Z. Zhang, D.L. Chen, *Mater. Sci. Eng. A* **2008**, *483–484*, 148.

[74] Q.B. Nguyen, M. Gupta, *J. Alloys Compd*. **2010**, *490*, 382.

[75] M. Kawasaki, B. Ahn, P. Kumar, J.-i. Jang, T.G. Langdon, *Adv. Eng. Mater*. **2017**, *19*, 1600578.

[76] C.A. Schuh, *Mater. Today* **2006**, *9*, 32.

[77] M. Kawasaki, J.-i. Jang, *Materials* **2017**, *10(6),* 596.

[78] N.Q. Chinh, P. Szommer, T. Csanádi, T.G. Langdon, *Mater. Sci. Eng. A*, **2006**, *434*, 326.

[79] H. Miyamoto, K. Ota, T. Mimaki, *Scripta Mater*. **2006**, *54*, 1721.

[80] H.W. Höppel, J. May, M. Göken, *Adv. Eng. Mater*. **2004**, *6*, 781.

[81] J. May, H.W. Höppel, M. Göken, *Scripta Mater*. **2005**, *53*, 189.

[82] H.W. Höppel, J. May, P. Eisenlohr, M. Göken, *Z. Metallkd.*, **2005**, *96*, 566.

[83] J. Mueller, K. Durst, D. Amberger, M. Göken, *Mater. Sci. Forum* **2006**, *503-504*, 31.
[84] A. Böhner, V. Maier, K. Durst, H.W. Höppel, M. Göken, *Adv. Eng. Mater*. **2011**, *13*, 251.

[85] V. Maier, K. Durst, J. Mueller, B. Backes, H.W. Höppel, M. Göken, *J. Mater. Res*. **2011**, *26*, 1421.

[86] V. Maier, B. Merle, M. Göken, K. Durst, *J. Mater. Res*. **2013**, *28*, 1177.

[87] J.M. Wheeler, V. Maier, K. Durst, M. Göken, J. Michler, *Mater. Sci. Eng. A*, **2013**, *585*, 108.

[88] M. Ruppert, W. Böhm, H. Nguyen, H.W. Höppel, M. Merklein, M. Göken, *J. Mater. Sci*. **2013**, *48*, 8377.

[89] P. Feldner, B. Merle, M. Göken, *J. Mater. Res*. **2017**, *32*, 1466.

[90] M. Wang, A. Shan, *J. Alloys Compd*. **2008**, *455*, L10.

[91] M.S. Mohebbi, A. Akbarzadeh, B.H. Kim, S.-K. Kim, *Metall. Mater. Trans. A*, **2014**, *45A*, 5442.

[92] M. Hockauf, L.W. Meyer, *J. Mater. Sci*., **2010**, *45*, 4778.

[93] R.Z. Valiev, N.A. Enikeev, M.Y. Murashkin, V.U. Kazykhanov, X. Sauvage, *Scr. Mater*. **2010**, *63*, 949.

[94] T.G. Karnavskaya, E.V. Avtokratova, A.M. Bragov, M.V. Markushev, O.S. Sitdikov, V.N. Perevezentsev, M.Y. Shcherban, *Tech. Phys. Lett.* **2012**, *38*, 620.

[95] L.W. Meyer, M. Hockauf, L. Krüger, I. Schneider, *Int. J. Mat. Res*. **2007**, *98*, 191.

[96] I. Sabirov, Y. Estrin, M.R. Barnett, I. Timokhina, P.D. Hodgson, *Scr. Mater*. **2008**, *58*, 163.

[97] I. Sabirov, M.R. Barnett, Y. Estrin, P.D. Hodgson, *Scr. Mater*. **2009**, *61*, 181.

[98] I. Sabirov, M.R. Barnett, Y. Estrin, I. Timokhina, P.D. Hodgson, *Int. J. Mat. Res*. **2009**, *100*, 1679.

[99] G.T. Gray III, T.C. Lowe, C.M. Cady, R.Z. Valiev, I.V. Aleksandrov, *Nanostruct. Mater*. **1997**, *9*, 477.

[100] R.Z. Valiev, I.V. Alexandrov, Y.T. Zhu, T.C. Lowe, *J. Mater. Res*. **2002**, *17*, 5.

[101] Y.M. Wang, E. Ma, *Applied Phys. lett.* **2003**, *83*, 3165.

[102] Y.M. Wang, E. Ma, *Acta Mater.* **2004**, *52*, 1699.

[103] Y.J. Li, X.H. Zeng, W. Blum, *Acta Mater.* **2004**, *52*, 5009.

[104] Y.M. Wang, E. Ma, *Mater. Sci. Eng. A* **2004**, *375–377*, 46.
[105] Q. Wei, S. Cheng, K.T. Ramesh, E. Ma, *Mater. Sci. Eng. A.* **2004**, *381*, 71.

[106] F.H. Dalla Torre, E.V. Pereloma, C.H.J. Davies, *Scr. Mater.* **2004**, *51*, 367.

[107] Y.J. Li, W. Blum, *Phys. Status Solidi. A*, **2005**, *202*, R119.

[108] F.H. Dalla Torre, E.V. Pereloma, C.H.J. Davies, *Acta Mater*. **2006**, *54*, 1135.

[109] M. Dao, L. Lu, Y.F. Shen, S. Suresh, *Acta Mater*. **2006**, *54*, 5421.

[110] J. Chen, L. Lu, K. Lu, *Scr. Mater*. **2006**, *54*, 1913.

[111] Y.-H. Zhao, J.F. Bingert, X.-Z. Liao, B.-Z. Cui, K. Han, A.V. Sergueeva, A.K. Mukherjee, R.Z. Valiev, T.G. Langdon, Y.T. Zhu, *Adv. Mater*. **2006**, *18*, 2949.

[112] A. Mishra, M. Martin, N.N. Thadhani, B.K. Kad, E.A. Kenik, M.A. Meyers, *Acta Mater*. **2008**, *56*, 2770.

[113] Y. Zhao, Y. Li, T.D. Topping, X. Liao, Y. Zhu, R.Z. Valiev, E.J. Lavernia, *Int. J. Mat. Res*. **2009**, *100*, 1647.

[114] T. Kunimine, T. Fujii, S. Onaka, N. Tsuji, M. Kato, *J. Mater. Sci.* **2011**, *46*, 4290.

[115] D. Zhou, H. Geng, D. Zhang, W. Zeng, C. Kong, P. Munroe, *Mater. Sci. Eng. A* **2017**, *688*, 164.

[116] A.P. Zhilyaev, S. Lee, G.V. Nurislamova, R.Z. Valiev, T.G. Langdon, *Scr. Mater.* **2001**, *44*, 2753.

[117] H. Jiang, Y.T. Zhu, D.P. Butt, I.V. Alexandrov, T.C. Lowe, *Mater. Sci. Eng. A* **2000**, *290*, 128.

[118] A.P. Zhilyaev, K. Oh-ishi, T.G. Langdon, T.R. McNelley, *Mater. Sci. Eng. A* **2005**, *277*, 410.

[119] A. Hohenwarter, R. Pippan, *Metall. Mater. Trans. A* **2019**, *50A*, 601.

[120] J.-K. Han, J.-M. Park, W. Ruan, K.T. Carpenter, A. Tabei, J.-i. Jang, M. Kawasaki, *Adv. Eng. Mater.* (in press).

[121] K. Edalati, R. Uehiro, K. Fujiwara, Y. Ikeda, H.-W. Li, X. Sauvage, R.Z. Valiev, E. Akiba, I. Tanaka, Z. Horita, *Mater. Sci. Eng. A* **2017**, *701*, 158.

[122] J.-K. Han, K.-D. Liss, T.G. Langdon, M. Kawasaki, *Scientific Reports* (under review)

[123] B. Wan, W. Chen, T. Lu, F. Liu, Z. Jiang, M. Mao, *Resour. Conserv. Recy*. **2017**, *125*, 37.

Figure captions

Figure. 1 Map showing a series of metal combinations for successful cold bonding by rolling and/or ARB. Reproduced with permission.[30] Copyright Year, Publisher.

Figure. 2 (a) Schematic illustration of solid-state reaction of Al and Cu semi-circular samples by HPT and (b) appearance of Al-Cu sample before and after HPT for 100 turns. Reproduced with permission.[31] Copyright Year, Publisher.

Figure. 3 (a) A schematic drawing and (b) the actual sample set-up shown with four quarter-circular disks of Al-6061 alloy and Cu, (c) the processed disk in computational modeling and (d) the estimated equivalent strain in the processed spiral hybrid texture. Reproduced with permission.[32] Copyright Year, Publisher.

Figure. 4 General set-up for HPT processing showing a stack of three disks between the anvils. Reproduced with permission.[35] Copyright Year, Publisher.

Figure. 5 A schematic sample set-up for the mechanical bonding by HPT of alternately stacked 19 Cu and 18 Ta thin foils. Reproduced with permission.[48] Copyright Year, Publisher.

Figure. 6 (a) A set of the compressed Cu/ZnO/Cu disks under 5 GPa without any torsional turns, and the microstructure at (b) overall and (b1)-(b4) some focused regions in the Cu-ZnO system after HPT for 5 turns under 5 GPa. Reproduced with permission.[43] Copyright Year, Publisher.

Figure. 7 Cross-sections and Vickers hardness variation (left) and XRD profiles (right) for (a) an Al-Fe system for 20 turns at 1.0 GPa and (b) an Al-Ti system after 50 turns at 6.0 GPa. Reproduced with permission.[41] Copyright Year, Publisher.

Figure. 8 A schematic display of the formation of tribomaterials. Reproduced with permission.[54] Copyright Year, Publisher.

Figure. 9 (a) A series of cross-sectional micrographs and (b) hardness variations at the disk diameter for the mechanically bonded Al-Mg system after HPT for 1-60 turns and (c) an XRD peak profile for the Al-Mg system after 20 HPT turns. Reproduced with permission.[35,38,55] Copyright Year, Publisher.

Figure. 10 (a) A series of cross-sectional micrographs and (b) hardness variations at the disk diameter for the mechanically bonded Al-Cu system after HPT for 10-60 turns and (c) XRD peak profiles for the Al-Cu system after 20 and 60 HPT turns. Reproduced with permission.[40] Copyright Year, Publisher.

Figure. 11 The XRD line profile for the mechanically-bonded Zn-Mg system after HPT for 1, 15 and 30 turns. Reproduced with permission.[44] Copyright Year, Publisher.

Figure. 12 A plot describing the change of diffusion coefficient against the Cu concentration in Al. The estimated diffusion coefficients for the mechanically bonded Al-Cu system by HPT are shown at red points 1-7. Reproduced with permission.[31] Copyright Year, Publisher.

Figure. 13 A relationship between r_b and the estimated equivalent strain and the shear strain for mechanically-bonded Al-Mg and Al-Cu systems after HPT at 6.0 GPa. Reproduced with permission.[55] Copyright Year, Publisher.

Figure. 14 Estimated Vickers microhardness values with increasing Mg content in Al solid solutions in the disk after HPT for 10 turns; the estimated total hardness is denoted by the red squares with error bars, and the hardnesses of the three different strengthening mechanisms of solid solution strengthening, HP strengthening and precipitation strengthening are denoted by the blue upright triangles, green circles and pink inverted triangles with error bars, respectively. Reproduced with permission.[35] Copyright Year, Publisher.

Figure. 15 A logarithmic plot of nanoindentation hardness against strain rate for the disk edges of the HPT-induced mechanically-bonded Al-Cu alloy. Reference data of pure Al and Cu after HPT for 10 turns are included in the plot. Reproduced with permission.[55] Copyright Year, Publisher.

Figure. 16 Variation of the strain rate sensitivity with grain size for various UFG Al[78-98] and UFG Cu[89,99-115] samples processed though different SPD processing procedures and for the HPT-synthesized hybrid Al-Cu system[55]. The encircling ovals provide a visual representation of the trends for Al, for Cu and for the Al-Cu system.

Figure. 17 (a) Schematic illustrations of the different sample set-ups and (b) a comparison of the macro-scale microstructures of the two samples with five disk stacking and three disk stacking for an Al-Mg system after HPT under 6.0 GPa for 10 and 20 turns, respectively. Reproduced with permission.[41] Copyright Year, Publisher.

Figure. 18 (a) Schematic illustration of the sample stacking in the order of Al/Mg/Cu/Fe/Ti/Al and (b) a comparison of the macro-scale microstructures of two samples after HPT under 2.0 GPa for 1 and 33 turns, respectively.

Figure. 19 Micrographs taken at the vertical cross-sections and the corresponding hardness variations for the scale-up Al-Mg disks having 25 mm diameter after HPT for 10 turns (upper) and 20 turns (lower). Reproduced with permission.[120] Copyright Year, Publisher.

Figure. 20 A high-resolution TEM micrograph showing the amorphous phase in the Zr alloy phase in the mechanically-bonded vanadium and Zr alloys by HPT for 5 turns. Reproduced with permission.[47] Copyright Year, Publisher.

Figure. 21 Cross-sectional microstructure, TEM micrographs taken at the disk edge and center and the corresponding hardness distribution at the cross-section of the synthesized Al-Mg alloy disk after 100 HPT turns, and (b) the XRD profiles of the mechanically-bonded Al-Mg system after HPT for 100 turns and HPT processing followed by natural aging for 60 turns with the reference XRD profiles of an as-received Al and Al after HPT for 5 turns. Reproduced with permission.[122] Copyright Year, Publisher.

Table caption

Table 1. The measured density, the maximum Vickers hardness value, and the estimated strength-to-weight ratio for the Al-Mg system[38] after HPT for 5-20 turns, Al-Cu system[40] after 20 and 60 HPT turns and Al-Fe system[62] after HPT for 20 turns with the reference materials of Al-1050 and ZK60 alloys after HPT for 5 turns and CP Cu and CP Fe after HPT for 10 turns.