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Synthesis of Hybrid Nanocrystalline Alloys by Mechanical Bonding through High-Pressure Torsion

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An overview of the mechanical bonding of dissimilar bulk engineering metals through high-pressure torsion (HPT) processing at room temperature is described in this Review. 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 macroscale microstructural evolution involves the concept of making tribomaterials and, for some dissimilar metal combinations, microscale microstructural changes demonstrate the synthesis of metal matrix nanocomposites (MMNCs) through the nucleation of nanoscale intermetallic compounds within the nanostructured metal matrix. Further straining by HPT during mechanical bonding provides an opportunity to introduce limited amorphous phases and a bulk metastable state. The mechanically bonded nanostructured hybrid alloys exhibit an 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

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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 the 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 applications requiring high electric

In the Materials Science community, one of the promising developments over the past three decades is microstructural refinement and nanostructuring 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 (BNMs) 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 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 after SPD processing.

Among numerous examples of the 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, $\varepsilon_{\rm eq}$, is given by the relationship of the form^[7]

$$\varepsilon_{\rm eq} = \frac{2\pi Nr}{h\sqrt{3}} \tag{1}$$

where *N* is the number of torsional turns, *r* and *h* are the radius and thickness of the disk sample, respectively, and the resultant shear strain, γ , is given by $\gamma = \sqrt{3}\varepsilon_{eq}$.^[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



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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 to avoid severe damage on the HPT anvils.

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In the past 5 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 lightweight 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 the sample set-up were tried in a series of preliminary studies of the processing approach, and recently a fixed procedure was established by demonstrating unique microstructural and structural formation in several different metal combinations.

Accordingly, this article is designed to review the procedures of the reported mechanical bonding of dissimilar metals by HPT and summarize the developments in the 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 last section before conclusions describes the critical parameters needed to promote mechanical bonding by HPT and the feasibility of the synthesis of metastable bulk nanostructured alloys by HPT-induced mechanical bonding.

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 multilayered plate from dissimilar metals but with micrometer-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 for 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 summary displays 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 face-centered cubic (FCC) metals are well studied and often easier to cold bond as 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



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bonding to provide direct contact between the metals, thereby utilizing 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 this Review, a much more powerful mechanical bonding of dissimilar metals is now feasible for the synthesis of hybrid metal systems by 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 semicircle 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 2b, and the processed disk demonstrated the mixture of the nanoscale Al- and Cu-rich phases involving two



Figure 1. Map showing a series of metal combinations for successful cold bonding by rolling and/or ARB. Reproduced with permission.^[30] Copyright 2008, NIMS.



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Figure 2. a) Schematic illustration of the solid-state reaction of Al and Cu semicircular samples by HPT and b) appearance of Al–Cu sample before and after HPT for 100 turns. Reproduced with permission.^[31] Copyright 2013, Elsevier.

intermetallic compounds of Al₂Cu and Al₄Cu₉. 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**a shows a schematic drawing of the initial quarter disks, and Figure 3b shows the actual 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 3c, and the calculated equivalent strain in the processed disk was estimated in the spiral hybrid texture, as shown in Figure 3d.

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 multilayered 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



Figure 3. a) A schematic drawing, 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 2012, Wiley.





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Figure 4. General set-up for HPT processing showing a stack of three disks between the anvils. Reproduced with permission.^[35] Copyright 2015, Elsevier.

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/Fe/Al,^[41] Al/Ti/Al,^[41] Cu/Al/Cu,^[42] Cu/ZnO/Cu,^[43] Zn/Mg/Zn,^[44] Fe/V/Fe,^[45,46] and V–10Ti–5Cr/Zr–2.5Nb/V–10Ti–5Cr.^[47]

It should be noted that some of these listed combinations of dissimilar metals for mechanical bonding by HPT use different sample volumes 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 is 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.



Figure 5. a) A schematic sample set-up for the mechanical bonding by HPT of alternately stacked 19 Cu and 18 Ta thin foils, b) placing the stack between HPT anvils. Reproduced with permission.^[48] Copyright 2017, Elsevier.

3. Structural Evolution and the Formation of MMNCs

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 the 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 6a,^[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 6b 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 Cu and ZnO.

The observation of phase fragmentation without 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 7a,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 (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, whereas the hardness values are higher as $Hv \approx 330$ and ≈ 350 in the Al–Fe and Al-Ti systems, respectively, at 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 presence of separate Al, Fe, and Ti phases without any compositional mixture, thereby nucleating new intermetallic compounds in these metal systems after HPT. The mechanical alloying and the consolidation process of Al and Ti powders by hot isostatic pressing should be noted^[50] and HPT^[51] demonstrated the formation of nanoscale 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. That 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





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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 2018, Elsevier.



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 2018, Cambridge University Press.

phase mixture. 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 MMNCs

Depending on the metal selections, several studies demonstrated the successful nucleation of nanoscale 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** shows the structural and hardness evolution of 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 9a,^[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 is shown in Figure 9b,^[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 9c at the disk edge of the Al–Mg system after 20 HPT turns.^[38]

It is apparent after 1 HPT turn that a multilayered structure remains throughout the disk diameter with the fragmented Mg layers having thicknesses of $\approx 200 \,\mu\text{m}$, and with no visible segregation at the Al–Mg interfaces. After increasing the numbers of HPT turns to 5–20, a similar microstructure consisting of multilayers of the Al and Mg phases was observed at the central regions at $r < 2.5 \,\text{mm}$ of the disk after 5 turns, and this was reduced to $r \approx 1.0 \,\text{mm}$ after 10 and 20 turns. In contrast, the disk





Figure 8. A schematic display of the formation of tribomaterials. Reproduced with permission.^[54] Copyright 2009, Springer Nature.

edge at r > 2.5 mm after 5 turns contains homogeneous distributions of very fine Mg phases with thicknesses of \approx 5–10 µm to even a true nanoscale of \approx 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 \approx 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 multilayered microstructure remained in small regions at the disk centers even after 40 and 60 HPT turns.

The hardness variation in Figure 9b shows the achievement of extreme hardness of Hv \approx 330 at the disk edge after 20 turns and the hardness saturates and is reasonably constant up to 60 turns. The low hardness values which are equivalent to the reference values for 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 \approx 115 and \approx 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 a small contribution from the formation of hard intermetallic compounds of Al₃Mg₂ and Al₁₂Mg₁₇ through the diffusion reaction shown in Figure 9c, where the XRD analysis followed by materials analysis using diffraction (MAUD)^[58] analysis computed the presence of Al₁₂Mg₁₇ 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 HPT mechanical bonding was conducted on the disk stacks of Al/Cu/Al. The cross-sectional micrographs and the hardness variation along the disk diameters are shown in **Figure 10**a,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



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.^[38] Copyright 2017,Elsevier^[55]; Copyright 2019, Wiley.



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 2018, Wiley.



are shown in Figure 10c.^[40] In the micrographs, the Al-rich phase is shown in the bright color, whereas the gray region at the disk center corresponds 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 multilayered microstructure over a wide region from the disk center is present at $r \ge 3-4$ mm and a mixture of very fine Cu-rich phases is at the disk edge after 10 HPT turns. Increasing the HPT processing to 60 turns significantly reduces the multilayered 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 10b 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 compound of Al₂Cu was estimated by XRD and associated MAUD analysis, as shown in Figure 10c. 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 the 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 XRD peaks indicate the presence of two intermetallic phases of Mg₂Zn₁₁ and MgZn₂ 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, attributing to an intensive mixture of the metallic phases and the formation of intermetallic compounds. Specifically, studies demonstrating the formation of nanoscale intermetallic phases defined the synthesized hybrid alloy systems as MMNCs.

A critical factor for the formation of MMNCs during 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 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 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 semicircle Al and Cu disks, as shown in Figure 2.^[31] Specifically, it was estimated that the diffusion coefficients were 10¹²–10²² times higher than lattice diffusion for the Al–Cu system during nanostructuring as well as the formation of intermetallic phases through HPT, and the estimated values are comparable with surface diffusion of the alloy system. **Figure 12** shows the change of the diffusion coefficient against 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



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 2019, Taylor & Francis.





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 2013, Elsevier.

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 mixture at RT.

Such a severe mixture of the dissimilar metal phases during mechanical bonding by HPT is visualized in macroscale micrographs, as shown in Figure 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 shown in the Al-Mg and Al-Cu systems in Figure 9 and 10, respectively. Taking a boundary radius, $r_{\rm b}$, which is the distance from the disk center dividing the multilayered structure from the severe mixture of metal phases at the disk edges, Figure 13 shows a relationship between $r_{\rm b}$ and the estimated equivalent strain and the shear strain computed using Equation (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 $r_{\rm b}$ and strain for both alloy systems, whereas the rate of change in $n_{\rm b}$ depends on the selection of dissimilar metals. This analysis implies, therefore, that there is a possibility to expand the regions with a severe mixture of dissimilar metal phases to achieve a homogeneous microstructure within the entire metal volume when applying a much higher strain 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 are that the processed bulk alloys are not the same as the

Shear strain 0 5 800 200 400 600 HPT: 6.0 GPa, RT, 1 rpm dividing from severe mixture, r_b (mm) Al-Mg system Distance from disk center Al-Cu system 3 2 0 0 100 200 300 400 500 600 Equivalent strain

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 2019, Wiley.

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 mixture 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 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 Al, Mg, and Cu alloys after HPT for the high numbers of turns. It should be noted that the density measurements for the mechanically bonded alloys were carried out at the disk edges where the measured volumes and weights are for the severely mixed alloy systems and the forming MMNCs for the Al–Mg and Al–Cu systems.

It is shown 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 \approx 455 MPa cm³ g⁻¹ for Al–Mg after 20 HPT turns, \approx 390 MPa cm³ g⁻¹ for Al–Fe after 20 HPT turns.



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.

Materials	Density [g cm ⁻³]	Maximum hardness [Hv]	Strength-to-weight ratio [MPa $cm^3 g^{-1}$]
Al/Mg/Al (HPT: 5 turns) ^[38]	2.50	130	170
Al/Mg/Al (HPT: 10 turns) ^[38]	2.48	270	350
Al/Mg/Al (HPT: 20 turns) ^[38]	2.34	330	455
Al/Cu/Al (HPT: 20 turns) ^[40]	4.47	400	290
Al/Cu/Al (HPT: 60 turns) ^[40]	4.32	500	390
Al/Fe/Al (HPT: 20 turns) ^[62]	3.81	320	296
CP AI (HPT: 5 turns) ^[38]	2.73	65	80
ZK60 (HPT: 5 turns) ^[38]	1.84	110	190
CP Cu (HPT: 10 turns) ^[40]	8.96	150	55
CP Fe (HPT: 10 turns) ^[62]	7.87	310	128

In practice, the computed specific strength increases with increasing HPT turns for the Al–Mg and Al–Cu systems. Moreover, the estimated specific strength for the HPT-induced alloy systems is significantly higher than the base materials of Al, Mg, Cu, and Fe which are strengthened by grain refinement through HPT processing. In fact, the specific strength of the mechanically bonded alloys is higher than many structural metals including steels which exhibit ≈ 150 MPa cm³ g⁻¹ and the engineering polymeric composites, ceramics, and carbon fibers demonstrating ≈ 200 MPa cm³ g⁻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 Figure 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 transmission electron microscopy (TEM) analysis and Mg contents and the phase fractions of intermetallic phases by XRD and MAUD analyses. These results are used to estimate each hardness contribution increase as separate and discrete effects, and Figure 14 shows the estimated total Vickers microhardness values from the three different strengthening mechanisms with increasing Mg content in an Al solid solution after



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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 blue upright triangles, green circles, and pink inverted triangles with error bars, respectively. Reproduced with permission.^[35] Copyright 2015, Elsevier.

HPT for 10 turns.^[35] In practice, the \approx 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 \approx 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 9b. Consequently, the evaluation confirmed the simultaneous occurrence of these strengthening mechanisms that are available due to SPD at low processing temperatures 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 complexity in 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 matrices on 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 load bearing in 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 Al₂O₃ or Y₂O₃ 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 (Δ CTE) between the matrix and reinforcement is also an important strengthening mechanism for composite materials as it leads to the generation of dislocations at the interfaces.^[69–71] The contribution of Δ CTE 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 Δ CTE. However, the intermetallic-based Al MMNCs synthesized by mechanical bonding through HPT provide significantly small Δ CTE 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 process is at RT, the Orowan strengthening has higher values than the CTE mismatch effect, whereas for those with a processing temperature higher than RT (as at \approx 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 where very fine (\approx 100 nm) insoluble particles are present in a metal matrix.^[68,72–74] Also, reductions in both diameter and the interparticulate spacing of the fine second-phase influence 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. Micromechanical 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 microstructure across their disk diameters, as shown in Figure 7, 9, and 10. Thus, the novel technique of nanoindentation provides a useful testing procedure^[75] for detecting and understanding the plastic yield at 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 micromechanical 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 the 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 postdeformation 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] whereas the hardness also increased at the disk edges after HPT through 60 turns, as shown in



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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 2019, Wiley.

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 acquires an improved *m* value of ≈ 0.08 with narrower error bars than after 20 turns. The results demonstrated 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.

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 *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 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 micromechanical 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 shown



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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 through 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, Cu, and the Al–Cu system.

by the arrow in Figure 16, for increasing HPT turns. Together with high hardness, as shown in Figure 10b, the observed excellent plastic response at the disk edges of the Al–Cu system leads to the conclusion that mechanical bonding and concurrent grain refinement through HPT is an excellent strategy for fabricating lightweight hybrid metal systems and forming a variety of MMNCs which show extraordinary physical and mechanical properties. It should be noted that there has been only one report demonstrating tensile testing on a mechanically bonded Al–Mg system through the bulk-state reaction by HPT^[116] but the test was conducted under limited testing conditions. To comprehensively understand the strength and ductility of the mechanically bonded alloy system, further developments in mechanical characterization procedures are required.

5. Feasibility of the HPT-Induced Mechanical Bonding

5.1. Numbers of Disks and Metal Varieties for Severe Phase Mixture

It is well defined in numerous fundamental studies on HPT processing that the critical processing parameters of HPT include compressive pressure^[117] and numbers of torsional turns.^[118,119] These parameters are also important for the HPT-induced mechanical bonding of dissimilar metals like the importance of torsional turn shown in Figure 13. In addition to these parameters, the number of stacking disks is a unique parameter, providing a difference in the mechanically bonded materials. In practice, mechanical bonding by HPT can be applied from upward of two disks of dissimilar metals such as Al/Mg.^[39]

A recent study showed a preliminary result of the influence of the number of stacked disks on the evolution of microstructure at the macroscale in the Al–Mg system. A total of five disks of Al and Mg, stacked in the order of Al/Mg/Al/Mg/Al, was processed by HPT under 6.0 GPa for 10 turns and **Figure 17**a shows the schematic illustrations of the different sample set-ups, and Figure 17b shows a comparison of the macroscale 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 $r_{\rm b} \approx 2.0$ mm, and there is a reasonable similarity in the microstructural evolution. Thus, this preliminary study provides information on the effect of large numbers of alternately 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 17b, 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 higher 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 five different commercial purity metals of Al, Mg, Cu, Fe, and Ti is shown in **Figure 18**, which is a schematic drawing of the sample stacking in the order of Al/Mg/Cu/Fe/Ti/Al, and in Figure 18b, which depicts the cross-sectional micrographs of the alloy system after HPT under





Figure 17. a) Schematic illustrations of the different sample set-ups and b) a comparison of the macroscale microstructures of the two samples with fivedisk 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 2018, Cambridge University Press.



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 macroscale microstructures of two samples after HPT under 2.0 GPa for 1 and 33 turns, respectively.

2.0 GPa for 1 and 33 turns. 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 macroscale after 33 HPT turns. Moreover, each separate metal phase necked during mechanical bonding, so 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 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 rotational speed during HPT should be reduced for large disk samples to minimize heat generation from friction with the anvils.^[120] 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 disks having 25 mm diameter after HPT for 10 turns (upper) and 20 turns (lower),^[121] 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 9a,b; the bright phase denotes the Al-rich phase and the dark phase denotes the Mg-rich phase in Figure 19.

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It is apparent that the final thickness of the processed alloy is \approx 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 the 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 centers in the 25 mm diameter disks 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 shown in Figure 9a. However, large Mg phases are apparent at the midradii 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 midradii positions, and the edges of the scale-up Al-Mg disks.^[121] Therefore, it is difficult to consider the complete dissolution of Mg atoms only at the disk center. A high hardness of $Hv \approx 250$ 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 \approx 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

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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.^[121] Copyright 2019, Wiley.

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 SPDprocessed samples is that the processing facility must have the capability to apply a sufficiently severe hydrostatic pressure, thereby creating a high compressive pressure in the case of HPT processing. The experiments discussed earlier 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 Alloy

This last section describes recent findings regarding the feasibility of using 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 multilayered 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-SPD of HPT for 1200 turns led to the formation of amorphous phases on several powder mixtures of Mg immiscible alloys,^[122] but the locations, sizes, and the volume fractions of the amorphous phases were not provided in these reports and structural homogeneity/heterogeneity is not clear.

In contrast, a recent report on a continuous HPT processing for an efficient amount of high shear strain demonstrated, for the first time, a bulk nanostructured metastable Al alloy in a



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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 2019, Elsevier.

supersaturated solid-solution state.^[123] 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 mechanical bonding synthesized a homogeneous



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(a) (b) HPT: 100 turns, 6.0 GPa, RT, 1 rpm Al-Ma HPT: 100 turns, 6.0 GPa, RT 111 ntensity 200 220 311 Al-Mg natural aging Al-Ma 100 turns AL5 turns Distance from id-section (mm) 0.3 as-recevied AI 360 300 240 180 0 2.5 3.0 3.5 4.0 5.0 4.5 -0.3 _4 -2 4 0 2 Scattering vector (A⁻¹) Distance from the center (mm)

Figure 21. a) 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.^[123] Copyright Year 2019, Springer Nature.

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 \approx 350–370, that is, by comparison with the alloy system with lower numbers of turns as shown in Figure 9b, a record high hardness achieved in the Al–Mg alloy system synthesized by HPT mechanical bonding.

Figure 21b 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.^[123] This evaluation provides an interesting finding that the Al-Mg system after 100 turns and additional annealing shows the FCC peaks for Al without any peaks for Mg or intermetallic phases, whereas 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 \approx 38.5 at% and an average of \approx 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^[124] and excellent contributions to current manufacturing techniques in diffusion bonding, welding, and mechanical joining.

6. Conclusions

1) Mechanical bonding of dissimilar engineering metals has been studied using conventional HPT processing for introducing unique alloy systems at RT. Currently, a variety of metal combinations are studied with 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 MMNCs by nucleating nanoscale 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 the 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 that there is 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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- [1] O. Vidal, F. Rostom, C. François, G. Giraud, Elements 2017, 13, 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, Y. 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. I. 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. Alloys 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.



- [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. K. 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, *Philos. 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. Y. 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 https://doi.org/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.



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- [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, Scr. Metall. Mater. 1991, 25, 33.
- [72] Z. Zhang, D. L. Chen, Scr. 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, 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, Scr. 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, Scr. 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. Mater. 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. Mater. Res. 2009, 100, 1679.
- [99] G. T. Gray, 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, Appl. 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. Mater. 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] M. M. Castro, S. Sabbaghianrad, P. H. R. Pereira, E. M. Mazzer, A. Isaac, T. G. Langdon, R. B. Figueiredo, J. Alloys Compd. 2019, 804, 421.
- [117] A. P. Zhilyaev, S. Lee, G. V. Nurislamova, R. Z. Valiev, T. G. Langdon, Scr. Mater. 2001, 44, 2753.
- [118] H. Jiang, Y. T. Zhu, D. P. Butt, I. V. Alexandrov, T. C. Lowe, *Mater. Sci. Eng. A* 2000, 290, 128.
- [119] A. P. Zhilyaev, K. Oh-ishi, T. G. Langdon, T. R. McNelley, *Mater. Sci. Eng. A* 2005, 277, 410.
- [120] A. Hohenwarter, R. Pippan, Metall. Mater. Trans. A 2019, 50A, 601.
- [121] J.-K. Han, J.-M. Park, W. Ruan, K. T. Carpenter, A. Tabei, J.-I. Jang, M. Kawasaki, *Adv. Eng. Mater.* **2019** https://doi.org/10.1002/adem. 201900971.
- [122] 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.
- [123] J.-K. Han, K.-D. Liss, T. G. Langdon, M. Kawasaki, Sci. Rep. 2019, 9, 17186.
- [124] B. Wan, W. Chen, T. Lu, F. Liu, Z. Jiang, M. Mao, Resour., Conserv. Recycl. 2017, 125, 37.

