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Ultrasonic welding of metallic glasses: a review

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ABSTRACT **Metallic glasses (MGs) are recognized for their unique combination of high strength, high elasticity, and excellent corrosion resistance, making them promising for advanced engineering applications. However, the practical utilization of MGs in component fabrication is often hindered by constraints such as limited dimension and lack of plasticity at room temperature. To overcome these challenges, ultrasonic welding (UW), due to its solid-state connection nature, has emerged as an effective method for the joining and processing of MGs. This review highlights the advantageous role of UW in enhancing the manufacturability of MGs. Specifically, UW offers low thermal impact and fast processing times, while also exhibiting suitability for welding in extreme environments, including deep-sea, polar, and aerospace settings. Furthermore, the review provides a detailed comparison and summary of the joining mechanisms underlying UW of MGs, while also emphasizing the potential challenges and applications of UW.**

Keywords: metallic glasses, ultrasonic welding, ultrasonic vibration, extreme manufacturing

INTRODUCTION

Various metal artifacts, such as bronze, iron, and silverware, have significantly contributed to technological progress and productivity throughout human civilization [1]. Throughout the long history of metal utilization, the production of metallic materials has been predominantly crystalline due to the limitations in manufacturing technology. With the advancement of solidification technologies, a novel class of advanced engineering materials known as metallic glasses (MGs) has gradually entered the horizon of the scientific community [2], which requires the development of more suitable and efficient molding techniques. Since the critical breakthrough by Duwez *et al.* [3] in the 1960s using vacuum melt spinning to create amorphous ribbons, MGs have emerged as a novel class of engineering materials with unique properties. These materials have an amorphous structure similar to that of traditional glasses, with their major components being metallic elements. To form these materials, the metallic melt must be cooled extremely rapidly, faster than its specific critical cooling rate, which initially limited fabrication to very thin dimensions such as thin films.

Subsequent innovations in rapid solidification techniques enabled the production of bulkier and chemically diverse MGs. In recent decades, MGs, particularly those based on Zr and Fe, have garnered substantial attention from both research and industry sectors due to their unique properties as metastable alloys. These alloys are devoid of a lattice structure, presenting an amorphous atomic arrangement that is densely packed yet devoid of long-range order. The absence of structural imperfections such as grain boundaries and dislocations, which are prevalent in conventional crystalline metals, results in enhanced properties and performance of MGs [4,5]. In general, MGs exhibit impressive yield stress, high elastic strain limit, superior wear resistance, and excellent biocompatibility. These characteristics have facilitated their integration into a variety of applications, including medical devices [6], micro gears [7], and flexible gears [8]. Additionally, their exceptional strength and elastic limits enable them more prone to generating resonance effects, making them valuable in acoustic equipment where they amplify sound and enhance the auditory experience. In particular, Fe-based MGs have demonstrated the potential to reduce energy loss by more than 70% when employed as cores, replacing traditional crystalline Fe-Si alloys (silicon steel sheets), owing to their superior soft magnetic properties and low coercivity [9]. Furthermore, the potential applications of bulk MGs (BMGs) in aerospace and environmental protection sectors are equally promising, underscoring their significant potential for future development and innovation [10].

However, ensuring that the unique atomic structure and properties of MGs are preserved while achieving high-quality shaping remains a critical issue in their application and manufacturing. Firstly, the requirement for extremely rapid cooling rates (~10^6 K/s) to prepare BMGs restricts their size (~cm), making the fabrication of larger-size BMGs difficult. Secondly, the absence of defects such as dislocations in BMGs can lead to catastrophic failure at room temperature during yielding, which severely hinders their structural applications [11]. To overcome these challenges, researchers have focused on designing alloy compositions to enhance the glass-forming ability (GFA) of BMGs. This approach typically involves the extensive refinement of compositions and precise control of the fabrication process. As a result, an alloy composition-independent approach, additive manufacturing techniques, has emerged to produce larger BMGs with inherently low GFA. These techniques include spark

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plasma sintering [12,13], selective laser melting [14–16], friction welding [17,18], explosive welding [19,20], electron beam welding [21–23], ultrasonic welding (UW) [24,25], liquid-solid joining [26], resistance upset welding [27,28], fused filament fabrication [29], and thermoplastic welding [28].

So far, these additive manufacturing and welding techniques for MGs can be categorized into two primary approaches [30]. The first is liquid-phase welding, which includes selective laser melting, explosive welding, electron beam welding, laser welding, and liquid-solid joining. These methods require heating the material above its liquid temperature for joining, but they risk surpassing the crystallization temperature (T_x), which can trigger crystallization and the properties of MG. To avoid crystallization, these methods, known for their high energy density and small processing region, rapidly melt the microregion in an instant (10–100 ms) and quickly return into an amorphous state under a fast cooling rate [19]. Accordingly, this approach requires MG with a wide supercooled liquid region (SLR, temperatures between T_g and T_x) and equipment with high power and precision to ensure an adequate processing time window and prevent crystallization.

The second approach to welding MGs is solid-phase welding, which includes a variety of techniques such as spark plasma sintering, friction welding, resistance upset welding, thermoplastic welding, fused filament fabrication, interface design approach, and UW. This solid-phase welding for MG involves maintaining the temperature within the SLR and precisely controlling the timing to harness the Newtonian viscous flow of the thermodynamically stable supercooled liquid state, thereby facilitating effective welding [31]. While most solid-phase welding technologies are effective in preventing crystallization, they require precise control over processing temperature and time to ensure thermodynamic stability. Notably, one technique that stands out is UW, which can achieve welding of MGs at temperatures below T_g and will be discussed in detail in this paper.

UW, a process that utilizes high-frequency vibration energy for material joining, has a history dating back to the early 20th century [32]. It is characterized by extremely short processing times, low temperature, pollution-free cleanliness, and energy efficiency. Initially, ultrasonic vibration equipment was mainly used in laboratory research with limited applications and gradually has been utilized in various fields such as healthcare, manufacturing, and communications [33]. With advancements in electronic technology and vibration theory by the mid-20th century, UW, one of the newest ultrasonic vibration techniques, became more mature and was applied to processes like foil welding and copper tube sealing [32]. Today, UW is widely utilized in diverse industries such as automotive, electronics, medical, and packaging, recognized for its efficiency, eco-friendliness, and reliability [34]. This progress has not only refined current manufacturing processes but also broadened the scope of materials that can be effectively joined, stimulating innovation and efficiency across various sectors [35]. UW excels in versatility, energy efficiency, and the ability to join a wide range of materials, including metals, polymers, and composites, with minimal thermal distortion and environmental impact. Compared to other advanced joining techniques like laser welding, friction stir welding, and adhesive bonding, UW offers a balanced combination of efficiency, material integrity, and sustainability, making it a valuable tool with broad industrial applications [36]. UW can be categorized into two different types based on the direction of vibration [36]: ultrasonic plastic welding (UPW) and ultrasonic metal welding (UMW). UPW, where the vibration is parallel to the interface, is primarily used for plastics and fabrics, as shown in Fig. 1a. UMW, where the

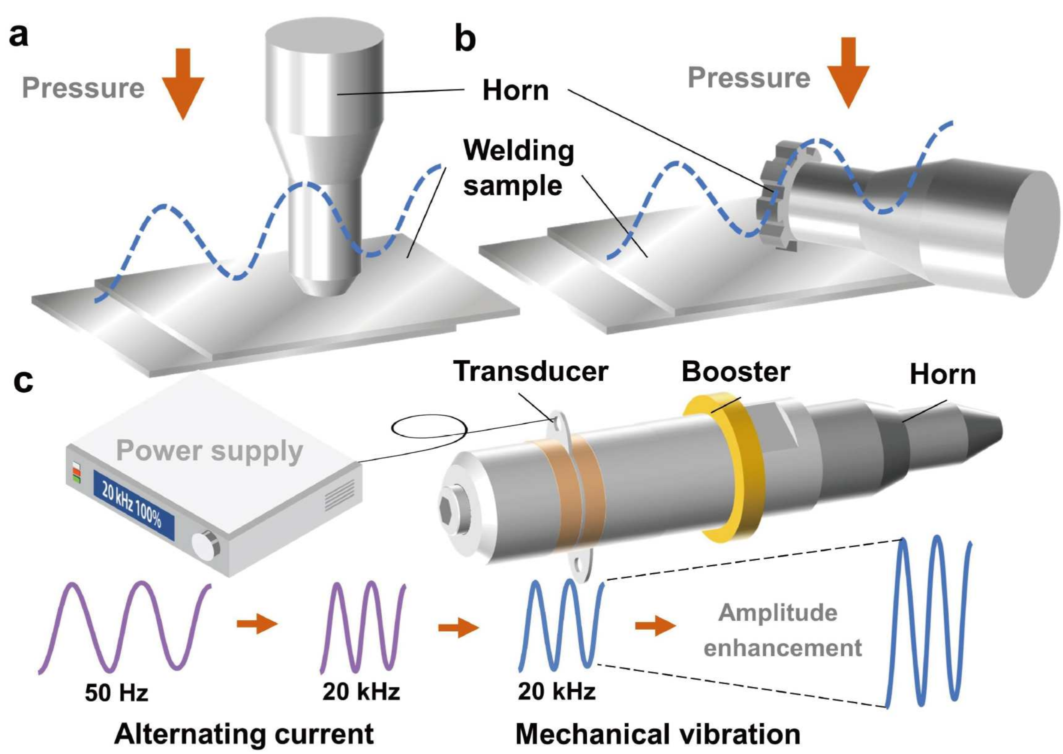

Figure 1 Schematic diagram of (a) ultrasonic plastic welding (UPW) and (b) ultrasonic metal welding (UMW). (c) The fundamental principle of UW.

vibration is perpendicular to the welding interface, is mainly applied for metal connections, as depicted in Fig. 1b. The fundamental principle of UW involves an ultrasonic generator converting low-frequency electrical current into high-frequency electrical current. This current is then transformed into mechanical vibrations of the same frequency by a transducer, which transmits and amplifies the vibration through a booster and horn to the welding tip. The tip applies vibrational energy to the joint area of the workpieces, promoting the reconstruction and rapid transformation of materials, thus enabling their interconnection and facilitating a robust union, as illustrated in Fig. 1c. Recognizing its commercial potential, both industry and academia have increased efforts to develop advanced UW equipment, aiming to expand its application to join plastics, metals, and other materials [37]. The two distinct UW technologies have been successfully applied to the welding of MGs, resulting in the formation of high-quality welded joints [38,39].

After nearly six decades of rigorous research, a variety of welding techniques for MGs have been developed, with their processing temperature and time requirements detailed in Fig. 2. The majority of these techniques involve extended exposure to temperatures above T_g, which increases the risk of crystallization and adversely affects the mechanical and thermal properties of MGs [40,41]. In 2020, Ma *et al.* [42] introduced an innovative ultrasonic vibration-induced plasticity (UVIP) technology for MGs, which operates at temperatures below T_g to significantly reduce material stiffness and minimize the risks of crystallization. This phenomenon is harnessed to soften the interfaces of MGs, enabling them to undergo plastic-like deformation and coalesce into a unified structure, a process known as UW, as previously reported. Due to the unique and superior properties of MGs and their significant potential, this joining method has garnered widespread attention from both industry and academia for its ability to effectively join and expand the critical size of MGs without compromising their performance [24,32,43]. In view of the rapid advancements in this field, it is essential to provide a thorough and contemporary synthesis of the technological milestones and the current understanding of the processes and phenomena involved.

This review article provides an in-depth analysis of the UW techniques applied to MG composites and BMGs, underscoring their adaptability in extreme environments such as deep-sea, polar, and aerospace conditions. It delves into the welding mechanisms of MGs and discusses potential issues and future development directions in UW, including the advent of ultrasonic additive manufacturing (UAM). The review emphasizes the environmental friendliness and efficiency of UW, which requires no additional binders and consumes low energy. Furthermore, it highlights the solid-state connection nature of UW, which is beneficial for controlling the formation of transitional phases and is suitable for joining dissimilar metal materials. By integrating the latest research progress and technological advancements, this review positions UW as a key technology in the future of material processing and manufacturing.

MECHANISM OF UW IN MGs

In recent years, UW has played a significant role in improving the room-temperature brittleness and critical dimensions of MGs. The mechanisms of UW have been further developed and validated through continuous research and exploration [24,25,44]. The unique bonding mechanism of MGs under UW differs from traditional welding mechanisms, which rely on atomic diffusion, plastic, and creep deformations. Early research by Maeda *et al.* [45,46] indicated that to achieve effective joining, the interatomic distance must be reduced to sufficient proximity. However, two key challenges need to be addressed that enable the constituent atoms on the bonding surfaces to draw closer to each other, i.e., the removal of the oxide layer and contaminants on the surface, and the requirement of surface roughness at the atomic scale to facilitate rapid bonding before re-oxidation or recontamination occurs. Additionally, it has been pointed out the bonding mechanism in MG is different from other metallic materials and that plastic deformation, creep, and atomic diffusion do not predominantly contribute to the joining of MGs.

Subsequently, Kim *et al.* [47,48] also acknowledged that ultrasonic vibration aids in the removal of surface oxide films and organic contaminants. However, they have proposed a unique insight, suggesting that mechanical wear and elastic

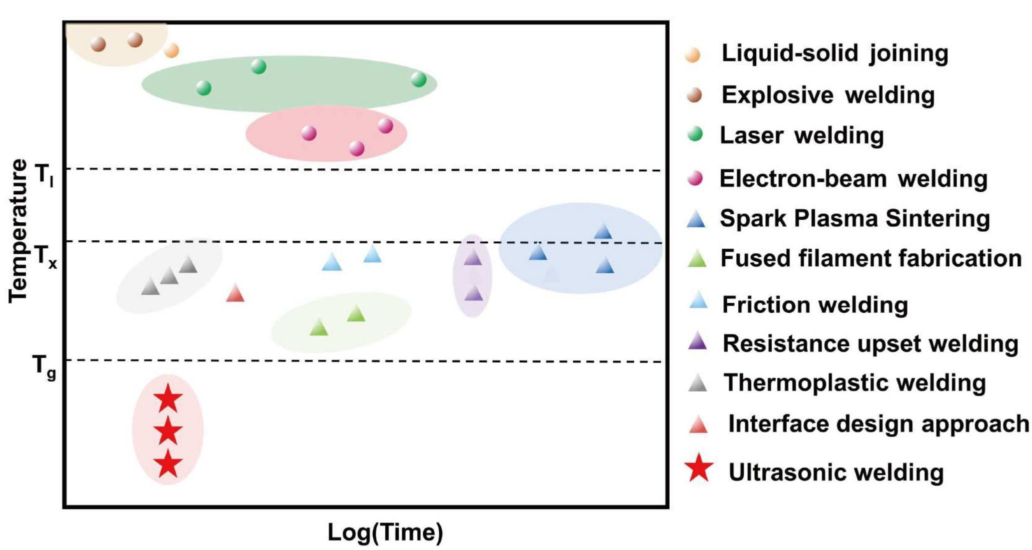

Figure 2 The processing temperature and time requirements diagram for various types of MG welding [12–29].

deformation occur in MGs when the energy input during welding is insufficient. They argued that the junction between MGs is due to the rapid increase in flash temperature at the interface roughness with sufficient welding energy, leading to softening, plastic deformation, and atomic diffusion. These processes allow the materials to come into close contact and form a joint. Despite these constructive speculations, the lack of specific experimental data means that the mechanisms of UW are still to be fully explored.

Ma *et al*. have initiated a series of studies on UW of MGs and have continued to delve into the joining mechanism. In 2019, they proposed a welding mechanism for MG ribbons under ultrasonic vibration, suggesting that MG ribbons can rapidly activate surface atoms at temperatures well below T_g, thereby promoting interatomic bonding [49]. Using molecular dynamics simulations, they confirmed that under high-frequency vibrations, the interfacial atoms exist in a higher energy state due to rapid surface dynamics, exhibiting viscous flow and diffusion and thus achieving mutual adhesion. This provided a simulated explanation for the mechanism. On this basis, they introduced a BMGs joining theory, emphasizing that joining is closely related to rapid surface dynamics, which induce viscous flow and adhesion of the interface. It was noted that under low-stress loading, MGs behave elastically without energy dissipation, similar to the findings of Kim *et al*. When subjected to ultrasonic vibration, MGs exhibit energy dissipation similar to that of rubber, indicating a different mechanical response to vibrations at various frequencies.

In 2020, Ma *et al*. [42] developed a novel ultrasonic vibration technique that allows for fast and low-temperature forming and welding of BMGs. This technique, based on a unique plasticity of MGs known as UVIP, enables the production of MGs with complex shapes while minimizing the risks of crystallization and oxidation. The unique plasticity of MGs under ultrasonic vibration, as demonstrated by dynamic modulus mapping (Fig. 3a), is a key factor in this process. Compared to the as-cast BMG, the local elastic moduli of the Pd-based BMG significantly decreased after UVIP but increased after thermal plasticity. These results indicate that the structure of the BMG samples was rejuvenated after UVIP but relaxed after thermal plasticity. According to the dynamic heterogeneity of MGs, this represents an increase in the volume fraction of liquid-like regions (flow units) within the BMG [50]. This phenomenon can be attributed to the high strain experienced by these liquid-like regions under high-frequency vibrations, which occur without sufficient time for stress release, thereby leading to the accumulation of stress and subsequent expansion of the liquid-like regions. Subsequently, the atomic structure changes and exhibits unique

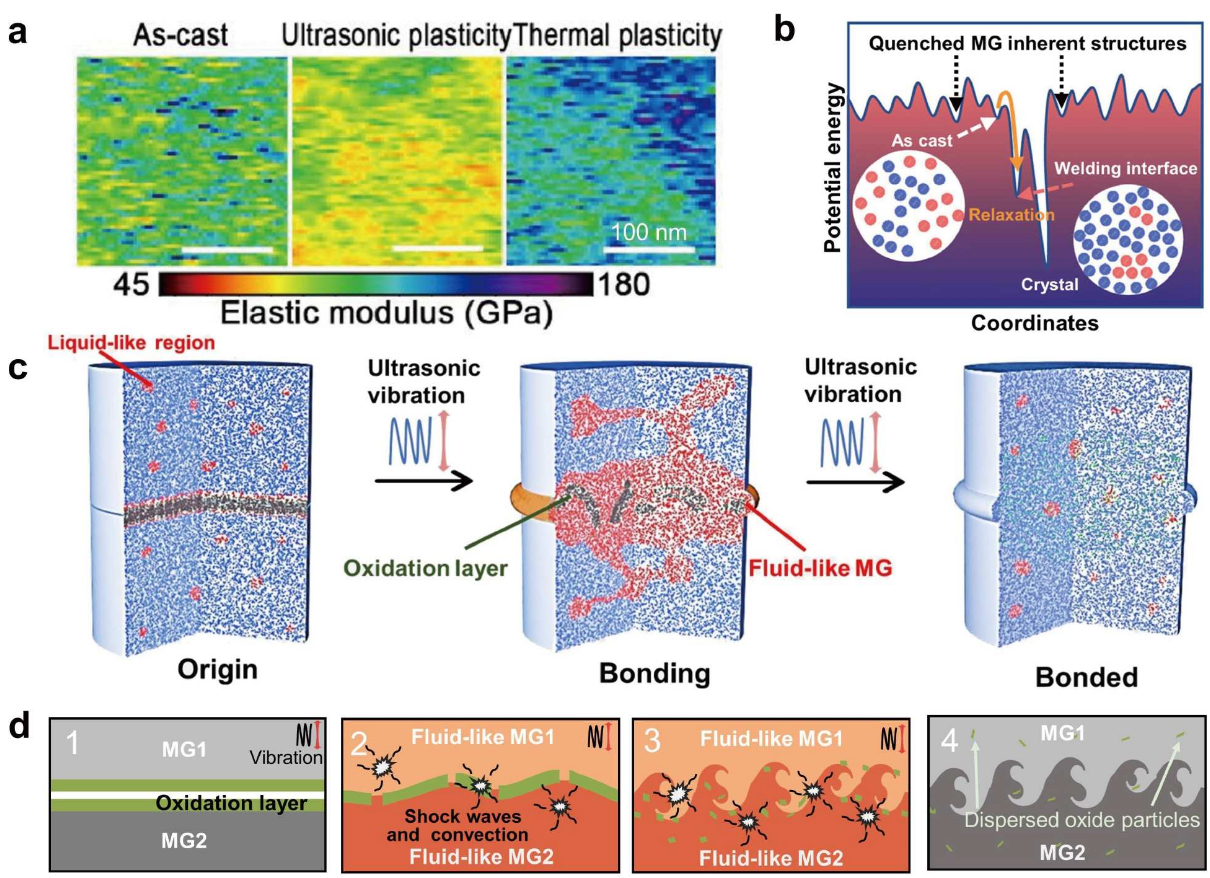

Figure 3 Schematic representation of the UW mechanism in MGs, showing dynamic modulus mapping and structural evolution. (a) Dynamic modulus mapping on the surface of Pd-based samples. Reprinted with permission from Ref. [42]. Copyright 2020, Elsevier. (b) Schematic representation of the structural evolution and stabilization processes in a potential energy landscape. The as-cast MG is quenched at a relatively high energy, and it will cross an energy barrier and stabilize to a higher-density atomic stacking state after processing. (c) Schematic diagram of UVIP and joining of the interface of MGs (atomic scale). (d) Schematic diagram of the mechanism for UW with irregular interface morphology. Reprinted with permission from Ref. [24]. Copyright 2023, Springer Nature.

rheological softening behavior due to the dynamic heterogeneity of BMGs and the expansion of atomic-sized liquid-like regions. In summary, UW achieves the rejuvenation and eventual collapse of BMGs, leading to overall flow softening behavior (UVIP).

Based on the UVIP, Ma *et al.* [24] provided an advanced UW mechanism for MGs. Under ultrasonic vibration, liquid-like regions can be rapidly activated, particularly at the interface between the two MGs, which accelerates the relaxation process and leads to a more intensive arrangement of these regions. As illustrated in the schematic of energy landscapes (Fig. 3b), the activated configurations jump out of the saddle point into neighboring basins with lower potential energy. As illustrated in Fig. 3c, this special rheological softening behavior causes the liquid-like regions to expand continuously at the joining interface and inside two BMGs. The rigid-like regions are destroyed under high-frequency vibration and pressure, and the liquid-like regions gradually connect into a single entity. Additionally, as shown in Fig. 3d (Stage 1), the cavitation effect of ultrasonic vibration produces ultra-high-intensity impacts and convection at the MG interface. This promotes the rupture and dispersion of the brittle oxide layer (Stage 2) and also leads to intense mutual mixing of the liquid-like regions on both sides of the interface (Stage 3). Ultimately, this results in an irregular connection interface, and when the vibration stops, the liquid-like regions at the interface transform back into rigid-like regions (Stage 4), forming a stable welding joint of MGs. In summary, the UW of MGs is due to the unique amorphous structure of MGs, which, under the excitation of ultrasonic vibration, promotes the breaking and dispersion of the brittle oxide layer and exhibits rapid surface dynamics and atomic-scale liquid-like region expansion, softening the interface and enabling rapid connection.

UW OF MGs AT AMBIENT CONDITIONS

UW of MGs with crystalline metal

In the early days of MG research, which began in 1978, Kreye *et al.* [51] successfully joined ~50-μm thick Ni-based MG to copper strips using 30 kHz UMW equipment. The researchers conducted shear tests and analyzed the electron diffraction patterns obtained from transmission electron microscopy (TEM) to characterize the joint, as shown in Fig. 4a. They successfully achieved a specific level of shear strength in the Ni-based MG without crystallization. However, for several decades following these initial findings, the UW of MGs faced a period of prolonged stagnation, primarily due to limitations in vibration technology and materials science. The revival of the UW technique in the early 2000s was driven by advancements in ultrasonic vibration equipment and the emergence of large-size BMGs [52]. These advancements enabled the UW of MGs to crystalline metals with renewed vigor.

Fe-based MG foils, renowned for their superior soft magnetic properties, are integral components in amorphous motor technology. However, a formidable challenge arises during the assembly of these MG stators with Al housings: achieving a robust and durable bond between the two materials. In 2015, Ji *et al.* [39] explored the UPW (27 kHz) of 100 μm-thick Fe-based MG foils to Al alloy and self-welding to each other, employing a Sn-based solder with heating to about 247–347°C. The findings revealed that both the solder/Fe-based MG interface (Fig. 4b) and the solder/Al interface achieved metallurgical bonding facilitated by the Sn-based solder. Notably, when the ultrasonic vibration power and welding time were sufficiently high, pronounced interfacial wetting and the microstructure refinement were observed on the Al alloy surface as shown in Fig. 4c. The researchers pointed out that the robustness of the joints and the observed phenomena were likely attributed to the synergistic effects of ultrasonic energy, including acoustic cavitation, micro-explosions, shock waves, and microstreaming. Furthermore, additional research has demonstrated that ultrasonic treatment facilitates the wetting of the MG and solder interface during UW with heating, thereby enabling effective bonding between the solder and the substrate [53,54].

In addition, researchers have developed a UW process that employs MG as filler. In 2019, Wu *et al.* [55] utilized a 40 μm -thick Ni-based MG as a filler to join 100 μm-thick Al and Cu ribbons at room temperature, utilizing 35 kHz UPW equipment, as shown in Fig. 4d. The researchers noted that the interface of this MG composite did not form any intermetallic compounds (IMCs), and the MG part remained in an amorphous state. The hardness and modulus of the materials at the junction were intermediate between those of the original ribbons and Ni-based MG. They proposed that this method, integrated with conventional mechanical machining or laser cutting techniques, enables additive manufacturing through a layer-by-layer deposition process, thus expediting the advent of 3D printing.

However, the utilization of solder in welding has its drawbacks, such as the formation of brittle IMCs at the interface, which can compromise the strength and reliability of the joint. In 2022, Becker *et al.* [56] advanced UMW techniques by employing a 400 μm-thick Zr-based MG (AMZ4) ribbon and a 1 mm-thick Al alloy (AA5754) sheet at room temperature without any solder, resulting in a welded joint with a shear strength exceeding 175 MPa, as shown in Fig. 4e. They also utilized infrared thermography to verify that the temperature during the welding process did not surpass the crystallization temperature required, and X-ray diffraction (XRD) analysis confirmed the amorphous state of the materials, indicating no crystallization occurred.

As previously reported, while MG ribbons have demonstrated successful joining with crystallized alloys, the UW of larger BMGs has remained elusive, presenting a notable challenge in the realm of BMG fabrication. In 2021, Zhao *et al.* [57] proposed a novel two-stage welding method using Zn-6Al-3Cu-20Sn alloy as solder to bond a 4 mm-thick Zr-based BMG to 1 mm-thick Ti-6Al-4V alloy sheet. The process begins with heating the Ti alloy and the solder together to achieve melting and joining at 550°C. Subsequently, the temperature is decreased to approximately 400°C, and UPW is applied to join them with Zr-based BMG. The resulting welded joint exhibited a shear strength of about 50 MPa. The researchers posited that the formation of pits and grooves on the joint surface not only enhanced the chemical bonding but also increased the mechanical interlocking strength through the anchoring effect, as shown in Fig. 4f. This method not only overcomes the challenges of joining Ti alloy and BMG, which are typically difficult with conventional welding techniques but also exemplifies a versatile multi-step welding approach that enhances the joining capabilities of dissimilar materials. However, UW of BMGs typically requires heating the material to temperatures approaching the T_g, which significantly amplifies the risk of crystallization.

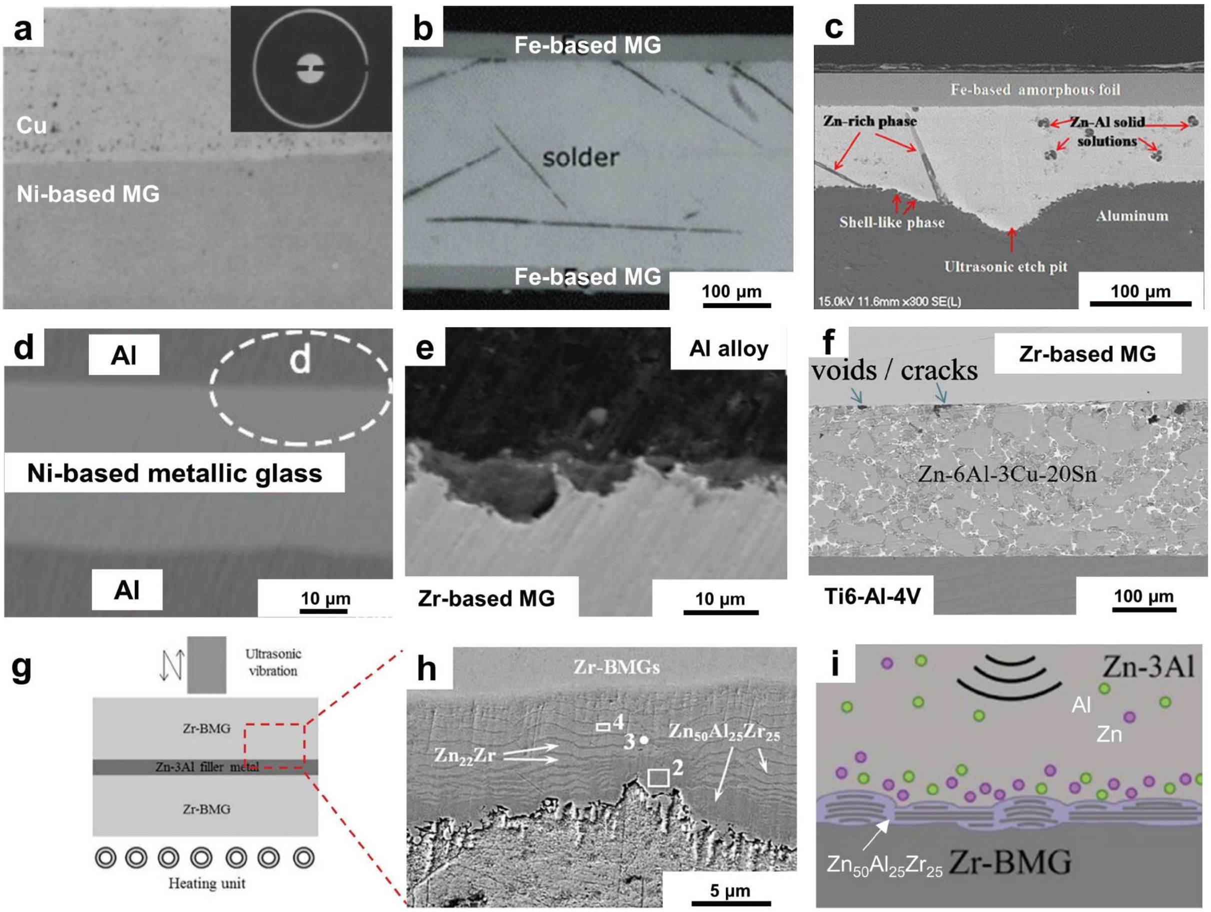

Figure 4 The UW joint of MG and crystalline metal/filler. (a) Optical micrograph of Ni-based MG/Cu joint fabricated by UMW and inset showing corresponding electron diffraction pattern. Reprinted with permission from Ref. [51]. Copyright 1978, Elsevier. Scanning electron microscopy (SEM) images of (b) the interface of the joints in Fe-based MG by UPW and (c) Fe-based MG ribbon/Al joint interface with Zn-Al solder and a heating temperature of 350°C. Reprinted with permission from Ref. [39]. Copyright 2015, Springer Nature. (d) The cross-sectional SEM image of Al joint fabrication by UPW with Ni-based MG filler. Reprinted with permission from Ref. [55]. Copyright 2019, MDPI. (e) SEM images of cross-sectional morphologies of Zr-based BMG/Al alloy interfaces after UMW. Reprinted with permission from Ref. [56]. Copyright 2022, MDPI. (f) SEM images of cross-sectional microstructure of Zr-based MG/Ti-6Al-4V UW joint with a heating temperature of 390°C and Zn-6Al-3Cu-20Sn solder. Reprinted with permission from Ref. [57]. Copyright 2021, Springer Nature. (g) Schematic picture of the UPW of Zr-based BMG with Zn-3Al solder. (h) SEM image of Zr-based MG joint interface by UPW with Zn-3Al solder and a heating temperature of 415°C. Reprinted with permission from Ref. [58]. Copyright 2018, Elsevier. (i) Schematic diagram of the reaction mechanism of Zr-BMG/Zn-3Al interface. Reprinted with permission from Ref. [59]. Copyright 2022, Elsevier.

Moreover, in 2018, Xiao *et al.* [58] proposed using 30 kHz UPW equipment to facilitate the liquid/solid interface reaction between Zn-3Al alloy and 1.3 mm-thick Zr-based BMG with heating to 415°C, as shown in Fig. 4g, resulting in a welded joint with a shear strength of 100.3 MPa. They then utilized the same approach to weld Zr-based BMG with 1060 Al alloy, achieving the highest shear strength of 67.86 MPa in 2022 [59]. Due to cavitation effects and metallurgical reactions that enhance atomic diffusion, they completely broke down the original passive film, forming an IMC layer ($Zn_{22}Zr$ and $Zn_{50}Al_{25}Zr_{25}$), and enabling a thorough interfacial reaction, as shown in Fig. 4h. Fig. 4i presents a schematic diagram illustrating the reaction mechanism, where enhanced cavitation effects intensify metallurgical reactions, facilitating the rapid formation of a continuous IMC layer at the interface, ultimately resulting in an effective welding joint.

In addition, some researchers have developed a rotational UW device to replace the horn with a roller, based on UMW equipment, to achieve the welding of MG to crystals, as illustrated in Fig. 5a. The roller applies transverse vibration and radial downforce to the foil, welding the entire interface and expanding the welded joint area. In 2018, Wang *et al.* [60] successfully welded cross-stacked multilayer Fe-based ribbons and Al1080 foils using rotational UW to create laminated composites at 190°C heating. In 2019, Misra *et al.* [61] used the same approach to fabricate Fe-based MG and Al1060 laminated composites with substrate preheating, as shown in Fig. 5b, without obvious crack initiation at the interface. Tensile tests showed that the composite material exhibited a maximum tensile strength of about 165 MPa, higher than the tensile strength of the Al layer, with the fracture mode characterized by a combination of brittle fracture in the MG layer and ductile fracture in the Al layer.

This chapter encapsulates the advancements and challenges in UW of MGs with crystalline metals and fillers. It highlights the historical progress, with early successes in joining MGs to copper strips, and the subsequent stagnation and revival due to technological limitations and advancements. The chapter

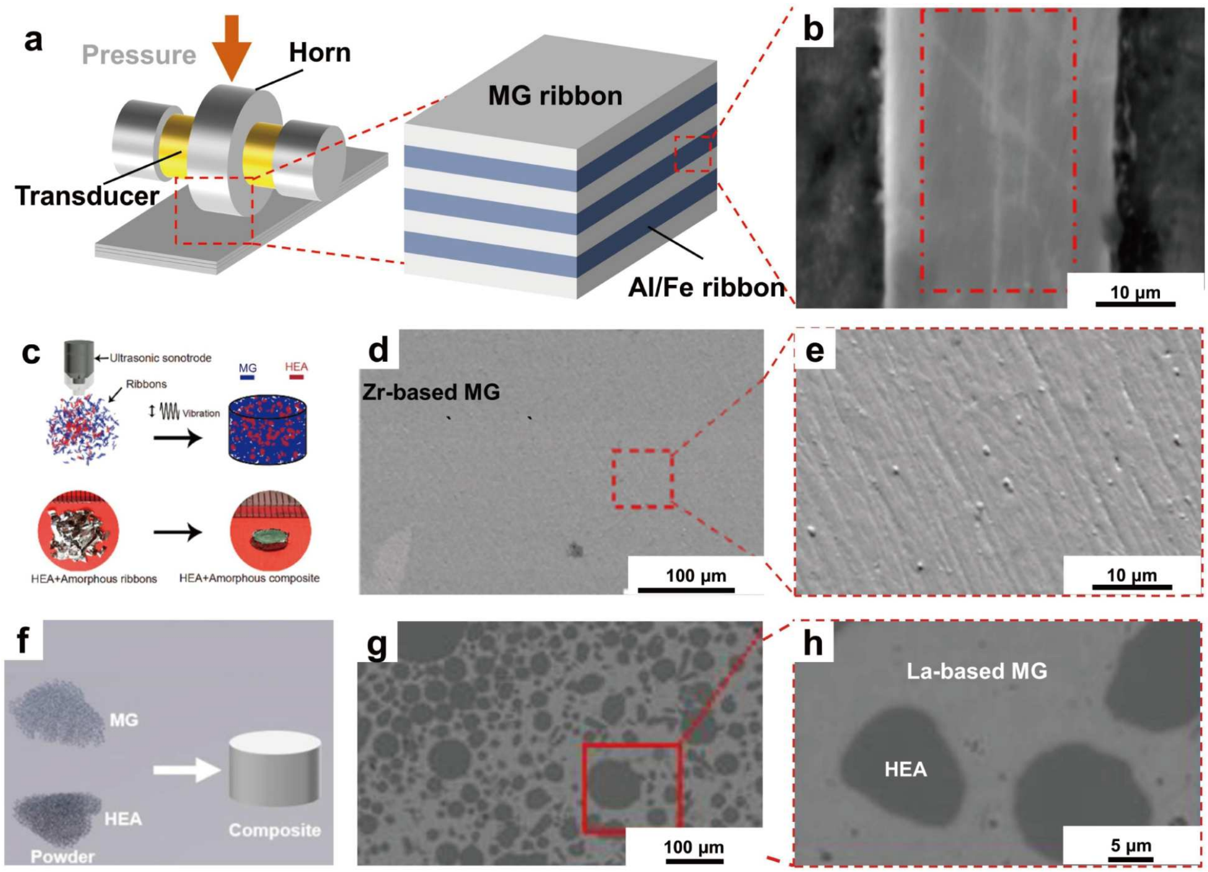

Figure 5 The ultrasonic additive consolidation process and the UW joint of MGs and HEAs. (a) Schematic diagram of ultrasonic additive consolidation process with Fe-based MG and Al alloy, and (b) the corresponding interface morphology of laminated composites. Reprinted with permission from Ref. [61]. Copyright 2019, Elsevier. (c) A schematic diagram for fabricating the HEA and Zr-based MG composite, (d) its interface SEM image, and (e) the corresponding enlarged image. Reprinted with permission from Ref. [66]. Copyright 2020, Springer Nature. (f) A schematic diagram for fabricating the HEA and La-based MG composite, (g) its interface SEM image, and (h) the corresponding enlarged image. Reprinted with permission from Ref. [67]. Copyright 2022, Elsevier.

underscores the benefits of UW, such as avoiding brittle IMCs by not using solder and the potential for additive manufacturing with MGs. It also discusses the innovative use of MG as a filler, enabling room-temperature welding without compromising the amorphous state. Despite these advantages, challenges remain in joining larger BMGs without heating to temperatures that risk crystallization. The development of rotational UW devices offers a promising approach to creating laminated composites. The mechanism of UW in composites is not entirely the same as in MGs. While the enhanced cavitation effects from ultrasonic vibrations also break and disperse the oxide layer, they also intensify the metallurgical reactions at the interface, promoting the rapid formation of a continuous IMC layer and ultimately resulting in an effective weld joint. In summary, while UW has shown significant potential in joining dissimilar materials, further research is needed to address the challenges of welding larger BMGs and to refine techniques for achieving robust, durable bonds without solder and heating.

UW of MGs with high-entropy alloy

Following extensive research on the welding of MGs with conventional crystalline metals, high-entropy alloys (HEAs) have emerged as a novel material with superior strength, toughness, and corrosion resistance [62–65]. In 2020, Ma *et al.* [66] successfully bonded $Al_{80}Li_5Mg_5Zn_5Cu_5$ HEA ribbons and Zr-based MG ribbons into a 1 mm thick, 5 mm diameter cylindrical block using a 20 kHz UPW equipment at room temperature, as illustrated in Fig. 5c. This composite material, characterized by its unique interlaced structure, combines soft MG and rigid HEA phases without any crack or porosity, as shown in Fig. 5d, e. The composite leverages the advantageous properties of both materials, significantly enhancing the mechanical properties compared to those of a single material. The bonding quality was further characterized using computed tomography (CT), which revealed no internal porosity or cracks. Subsequent their research in 2022 explored the utilization of La-based MG with CoCrFeNiMn HEA ribbons to fabricate a 3 mm thick, 5 mm diameter cylindrical block [67]. By tuning the mass ratios of MG and HEA in the composites, the regulation of the compressive strength and plasticity of the composites is achieved. These studies confirm the versatility of UW in joining MGs with HEA, enabling the artificial design and manufacture of novel multiphase and multicomponent materials.

In summary, UW has been successfully applied to a diverse range of material combinations, including Fe-based MG foils with Al alloys, Zr-based MGs with Al alloys, Ni-based MGs with Al and Cu ribbons, and HEA with Zr-based MGs, demonstrating its broad material compatibility. This advanced welding technique not only capitalizes on the exceptional strength and high elasticity of MGs but also integrates the ductility and formability of conventional metals. The resulting composite materials exhibit a synergistic combination of these desirable properties. The bonding mechanism in UW is primarily due to severe plastic deformation at the interface, induced by ultrasonic vibration and substrate preheating. This process effectively breaks down surface oxide layers, realizes metallurgical bonding, and facilitates atomic diffusion across the MG interfaces. The unique capabilities of UW in joining innovative materials, such as HEAs and MGs, underscore its pivotal role in materials science. As the demand for lightweight, high-strength materials continues to grow, UW is poised to broaden its application in sectors such as automotive, aerospace, and energy.

UW of MGs

The UW of MG with traditional crystalline metals or HEAs has opened up new avenues for structural applications of MG. This integration not only broadens the range of possible material combinations but also addresses some of the inherent challenges in MG fabrication, such as improving ductility and thermal stability [61]. However, the limited applications of MGs are primarily due to the inherent limitations of their GFA, which requires cooling rates typically in the range of 10 to 10^3 K/s to prevent crystallization and form amorphous structures [15,68,69]. Traditional casting methods often result in crystallization and a loss of the desired amorphous characteristics when attempting to increase the size of MGs [41]. Now, joining technologies, particularly UW, has become a significant avenue for developing larger MGs [70,71]. UW has made substantial contributions to expanding the size limit of BMGs [25], allowing researchers to fully harness the potential of larger-size BMGs and paving the way for the development of high-performance materials.

In 2008, Maeda *et al*. [46] pioneered the UMW of Zr-based MGs without the need for any solder, successfully achieving the bonding of pure MGs. Their study utilized a 75 kHz vibration frequency and a vibration duration spanning from 5 to 3000 ms and investigated the feasibility of UMW for joining $Zr_{55}Cu_{30}Ni_5Al_{10}$ MG strips. Notably, they discovered that effective joining could be accomplished without external heating, albeit with a limited bonding area and "weak" strength, as depicted in Fig. 6a. However, by introducing external heating at 110°C, the researchers significantly expanded the bonding area and enhanced the strength of the joint. In 2009, they conducted a more in-depth study on the microstructure and thermal stability of UW joints of 50 µm-thick $Zr_{55}Cu_{30}Ni_5Al_{10}$ MG strips [45]. Directly measuring the interface temperature using a thermocouple, they confirmed that the maximum temperature remained below T_g, suggesting that UW has the potential advantage of maintaining the amorphous structure of MGs. These results marked a pioneering advancement in the MG fabrication field, providing significant technical insights for future manufacturing methods. In 2014, Kim [47] successfully achieved the UW of a 1 mm-thick Cu-based MG plate using 20 kHz UW equipment in just 1 s, with no signs of crystallization at the weld joint in the XRD pattern. As shown in Fig. 6b, the SEM image of the joint analysis confirmed the disappearance of the interface, and the differential scanning calorimetry analysis indicated that the thermal properties of the MG plate remained unchanged after welding. This study highlights the robust capability and potential of UW for joining larger-size BMGs, thereby broadening their industrial application prospects.

Subsequently, UW of MGs gained increasing attention from researchers. In 2015, Zhu *et al*. [72] utilized 20 kHz UMW equipment to successfully join five layers of 25-µm-thick Fe-based MG ribbons. The microstructure and amorphous structure of the weld joints were characterized by SEM, as shown in Fig. 6c, and XRD. No visible interface was observed, indicating

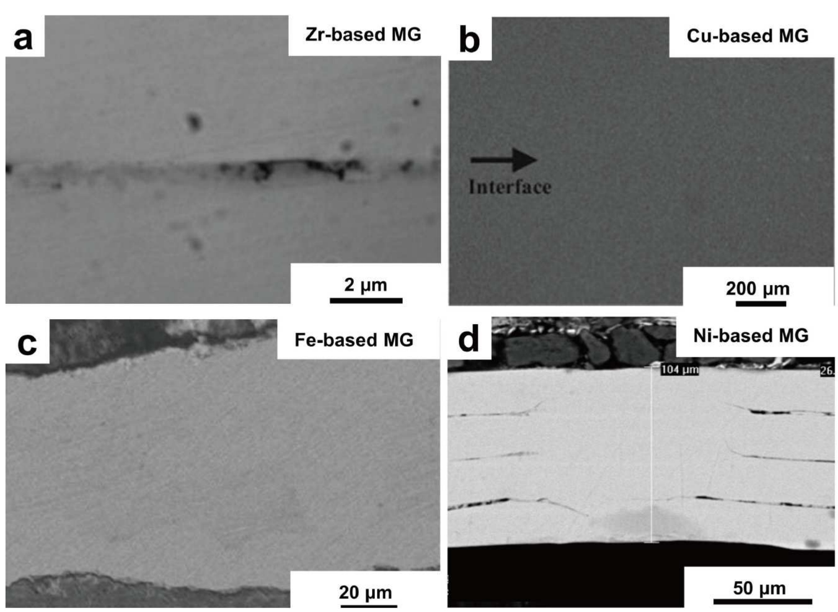

Figure 6 SEM images of Zr-based, Cu-based, Fe-based, Ni-based MG UW joint. (a) Interfacial microstructure of Zr-based MG ribbons joined by UMW without external heating. Reprinted with permission from Ref. [46]. Copyright 2008, Elsevier. (b) Cross-sectional morphology of the Cu-based MG plate joining interfaces by UW. Reprinted with permission from Ref. [47]. Copyright 2014, Elsevier. (c) Cross-sectional morphology of the Fe-based multilayer MG interface by UMW. Reprinted with permission from Ref. [48]. Copyright 2015, Trans Tech Publications. (d) Cross-sectional morphology of Ni-based multilayer BMG by UMW. Reprinted with permission from Ref. [74]. Copyright 2020, Trans Tech Publications.

the successful joining of the MG foils without any defects or pores, and the MG remained amorphous. In 2019, Li *et al.* [73] successfully implemented multilayer additive welding of 40 μm -thick Ni-based MG ribbons using 35 kHz UMW equipment, implying the potential for layer-by-layer fabrication of complex MG structures. In 2020, Nicolaeiscu *et al.* [74] also utilized 20 kHz UMW equipment to achieve multilayer additive manufacturing of 20 μm-thick Ni-based MG, as shown in Fig. 6d. This study reaffirmed the applicability of UW technology for the additive manufacturing of Ni-based MG strips. These researches have laid the groundwork for a future where UW may be seamlessly integrated with traditional machining and, optimistically, laser cutting technologies. Such integration promises to facilitate the rapid additive manufacturing of large and complex MG components, simultaneously addressing the size limitations and processing challenges associated with MGs.

In 2019, Ma *et al.* [49] introduced an innovative method for joining different MGs using 20 kHz UPW equipment, successfully welding MG strips of various compositions into BMGs with a diameter of 5 mm and height of 3 mm. This approach achieved rapid atomic bonding between MG strips at temperatures significantly below T_g, thereby overcoming the limitations of traditional preparation methods in terms of dimension and composition. Meanwhile, the process has the advantage of swiftly bonding MGs with different physical properties without inducing crystallization. The researchers utilized La-, Zr-, and Pd-based MGs to fabricate both homogeneous (La-La, Zr-Zr, Pd-Pd) and heterogeneous (La-Zr, Zr-Pd, La-Pd) BMGs. As depicted in Fig. 7a, the TEM image of the Pd-La welded sample interface revealed a distinct boundary. The corresponding high-resolution TEM (HRTEM) image and three different regions of selected area diffraction patterns (SADP), as shown in Fig. 7b, indicated clear halo rings, confirming the amorphous state of the multiphase BMG. Energy-dispersive X-ray spectroscopy (EDS) results confirmed the interdiffusion of elements across the different MG regions, highlighting the presence of Pd and P in the La-based amorphous phase, as well as La, Al, and Co in the Pd-based amorphous phase. This study has successfully demonstrated a UW technique that is adept at synthesizing both single-phase and multiphase MGs. Consequently, this technique not only facilitates the enhancement of structural integrity and performance characteristics of multiphase MGs but also establishes a versatile framework for the design and fabrication of innovative MG compositions [58].

The following year, Ma *et al.* [25] further explored the UW of MGs, demonstrating a new approach for preparing BMGs with larger sizes. Using a 20 kHz UPW equipment, researchers joined two pieces of La-based BMG and formed a larger BMG. Four welding samples were prepared, varying the welding energy from 50 to 400 J. Their findings revealed a direct correlation

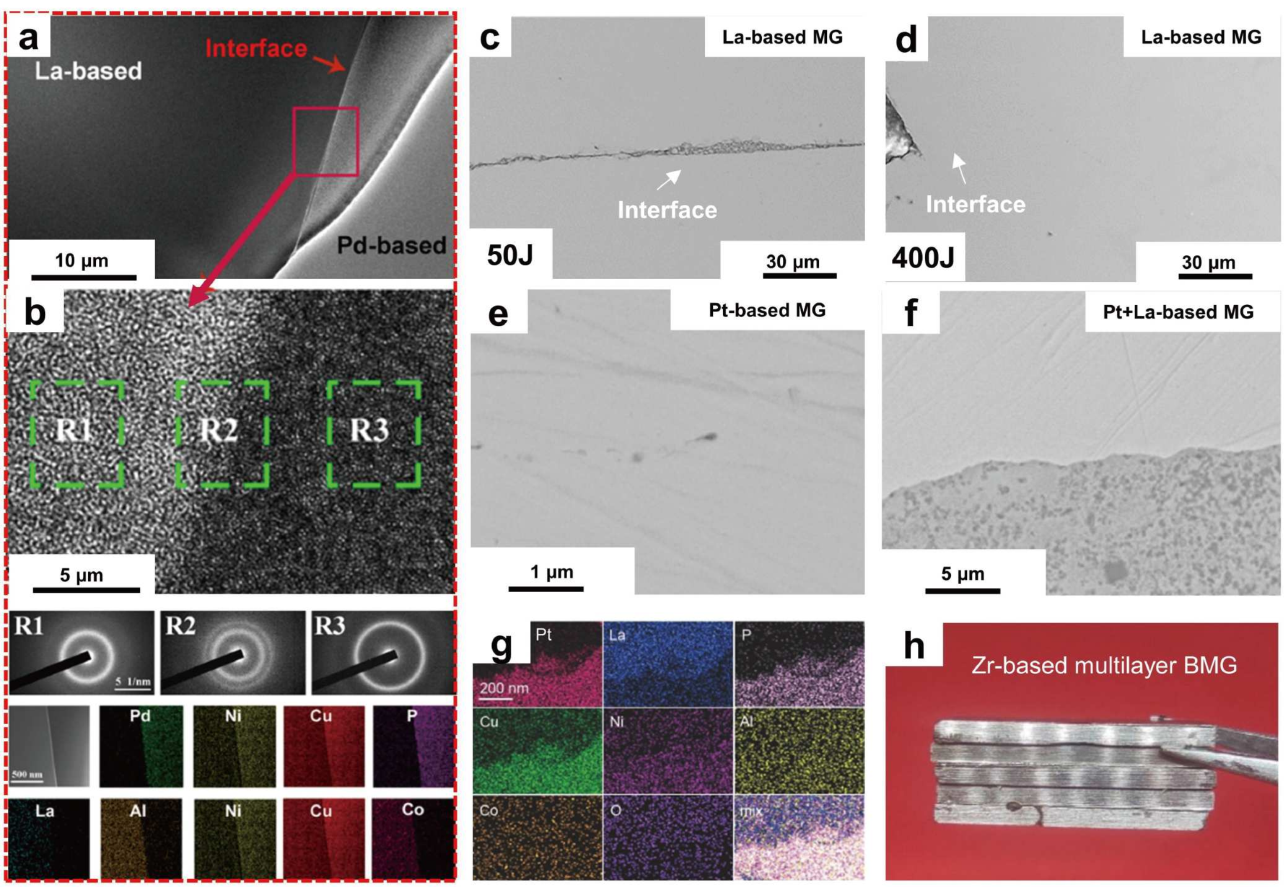

Figure 7 UW of La-based, Pd-based, Pt-based, and Zr-based MGs without additional heating. (a) SEM image of the La-Pd-based dual-phase BMGs and (b) its HRTEM image, corresponding SADP, and EDS patterns. Reprinted with permission from Ref. [49]. Copyright 2023, AAAS. Cross-sectional SEM images of La-based MG welding samples with welding energy at (c) 50 and (d) 400 J. Reprinted with permission from Ref. [25]. Copyright 2020, Elsevier. Cross-sectional SEM images of (e) Pt-based MG and (f) Pt-La-based MG fabrication by UW. (g) EDS maps of the Pt-La-based MG. (h) Optical image of multilayer Zr-based MGs fabrication by UW. Reprinted with permission from Ref. [38]. Copyright 2021, Springer Nature.

between welding energy and the strength of the MG, with the optimal welding strength achieved at 400 J, matching the hardness of the cast material. SEM images (as shown in Fig. 7c, d) revealed a phenomenon consistent with the trend of hardness changes, with the welding seam narrowing and bending as welding energy increased. This research marked a novel UW endeavor with rare-earth-based BMGs and offered a fresh perspective on the fabrication of BMGs of virtually unlimited size and damage repair.

In 2021, Ma *et al.* [38] further substantiated the versatility of UW for MGs with varying compositions by expanding the selection of MG substrates, including commonly used Zr-based MGs and precious metal Pt-based MGs. This research adopted a multi-amorphous phase design approach, achieving heterogeneous welding such as Pt-La, Pt-Zr based MG, as well as homogeneous welding like Zr-Zr, Pt-Pt based MG (as shown in Fig. 7e). As shown in Fig. 7f, the ultrasonic heterogeneous welding sample of La-based and Pt-based MG displayed different contrasts and a distinct bending interface, resulting in a strong welded joint. The corresponding EDS spectrum, as shown in Fig. 7g, indicated the interdiffusion of elements from both amorphous phases, confirming the interconnectedness of the welding materials. Additionally, the researchers demonstrated the multilayer welding of Zr-based BMG into a single entity, as shown in Fig. 7h, verifying that the UW is suitable for additive manufacturing to create larger-size BMGs, thus surpassing the GFA limitations of MGs.

The collective research efforts in the field of UW of MGs have revolutionized the approach to joining and fabricating MG components. These investigations consistently show that UW can effectively bond a variety of MG compositions, including Zr-based, Pt-based, pd-based, and rare earth La-based MGs, into larger and more complex structures. This advancement offers a flexible and efficient method for the large-scale fabrication and damage repair of MGs, overcoming previous limitations in size and composition. These findings also demonstrate the potential for integrating UW with other manufacturing technologies, such as laser cutting, which could significantly expand the application areas of MGs in various industries. In summary, the innovative UW techniques for MGs have opened new avenues for the design, fabrication, and application of these unique materials, promising a future with broader industrial utilization and more complex structural components.

UW OF MGs IN EXTREME ENVIRONMENTS

With the ongoing progress of industrial technology and the expansion of exploration fields into more challenging terrains, human activities have extended to various extreme environments, such as the deep sea, space, explosive and flammable environments, polar regions, and high-radiation areas. In these conditions, the reliability and safety of equipment are of crucial importance. Welding, a key technology for jointing and repairing materials, has become increasingly indispensable for applications in extreme environments. Moreover, in extreme environments, special requirements are imposed on the integrity of materials and structures. In these environments, equipment damage and failure can lead to catastrophic consequences. Therefore, on-site welding and repair capabilities in extreme environments have become important, such as offshore wind turbine repairs, high-altitude bridge repairs, and space exploration [75]. Among these extreme environments, underwater welding stands out as a representative challenge. Underwater welding is broadly categorized into wet and dry welding.

The dry welding process requires the employ of a gas chamber to enclose the welding area for welding, which is too difficult to operate and requires special equipment and technical support, thus, limiting its widespread application. In contrast, wet welding is performed directly underwater using arc welding, offering portability and ease of operation, and is currently the primary method used.

However, wet underwater welding also faces significant challenges. The welding temperature can range from 500 to 800°C, coupled with extremely high cooling rates, which may lead to the degradation of mechanical properties in the welding area. Additionally, the unique characteristics of the underwater environment often result in the formation of pores caused by molecular hydrogen, carbon monoxide, or water vapor. The welding process also demands substantial currents (several hundred amperes), posing high demands on welding equipment and increasing the risk of electric shock to welders or divers. Furthermore, increased water depth can also affect the stability [75,76].

To address these challenges, the development of effective wet joining technologies is particularly crucial, especially in fields like national defense, offshore mining, energy storage, and aerospace exploration. The advancement of these technologies is vital for meeting the stringent requirements for joining processes in extreme environments and overcoming the limitations of existing techniques, thereby enhancing the safety and reliability of welding operations. Ma *et al.* [24] have conducted in-depth research and discovered the potential of MGs as an advanced material in joining technologies for extreme environments. MGs possess excellent mechanical properties, such as high strength, high elasticity, superior wear resistance, and corrosion resistance, making them an ideal material for many applications. Particularly in liquid environments, the application prospects of MGs are particularly broad. For instance, in marine engineering and the chemical industry, MGs are crucial for the reliability and safety of equipment.

Ma *et al.* [24,44] have delved into the application of UW for the joining of MGs in extreme liquid environments, such as freshwater, seawater, alcohol, and liquid nitrogen. This technique utilizes mechanical vibrational energy to achieve material bonding, offering significant advantages such as low-temperature demands, swift welding, simple operation, strong environmental adaptability, and high safety. Fig. 8a presents the SEM morphology of the interfacial microstructure of Zr-based MG under UPW without external heating in freshwater, demonstrating the achievement of a seamless MG joint by modulating welding energy. Fig. 8b displays the SEM images of heterogeneous Zr-Ti-based MG welding joints, highlighting the irregular boundary morphology at the interface, with researchers noting the presence of mixed amorphous phases at a more microscopic level in Fig. 8c. As shown in Fig. 8d, a magnified image reveals the interpenetration of bands with different contrasts, indicating the presence of discrete oxide layers near the Zr-based MG side. Furthermore, researchers have explored various homogenous and heterogeneous welding of MGs, including Zr-based and Ti-based MGs. CT scanning was employed to preliminarily assess the bonding quality of the joint, showcasing the relative density maps at multiple positions within the welded joint. Fig. 8e, f illustrate the CT scans of

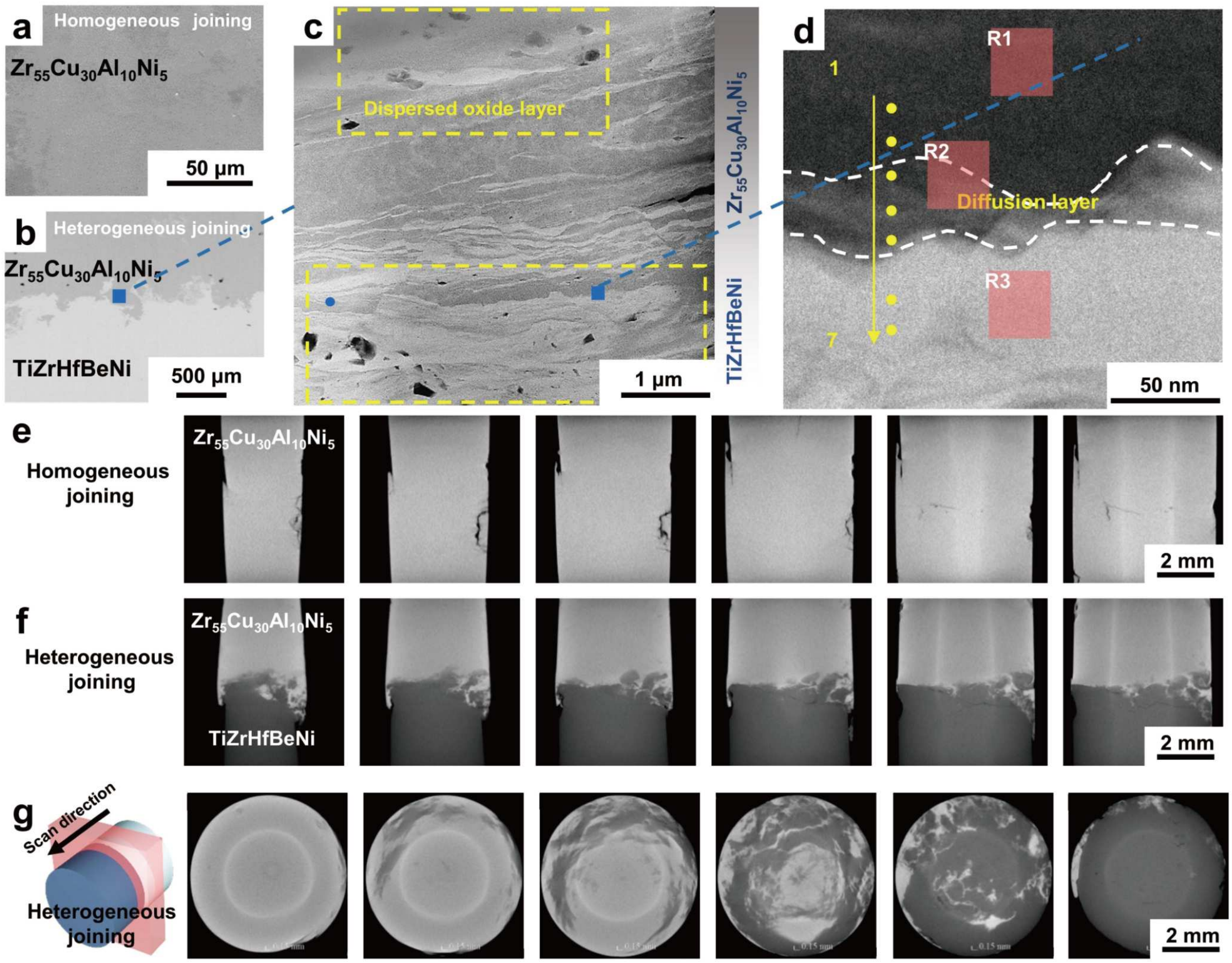

Figure 8 Microstructure of MG welding samples. (a) SEM morphology of the Zr-based homogeneous samples after UW. (b) SEM morphology of the Zr-Ti-based heterogeneous sample. (c) High-angle annular dark field (HAADF) image at the interface of the heterogeneous joined sample. (d) HAADF at the diffusion layer of the heterogeneous joined sample. CT scan patterns of (e) homogeneous and (f) heterogeneous joined samples. (g) CT scan patterns of heterogeneous joined samples. The inset shows the scanning direction (cross-section). Reprinted with permission from Ref. [24]. Copyright 2023, Springer Nature.

homogenous Zr-based MG joint and heterogeneous Zr-Ti-based MG joint separately. The uniform relative density distribution in the homogenous welding CT scans, without any seams or voids, provided evidence that samples have good welding quality. In contrast, Fig. 8f shows a contrast disparity on the sample surface due to the density differences between the two MGs. The axial CT scan in Fig. 8g reveals a distinct contrast transition at the junction of heterogeneous materials, indicating mutual diffusion at the interface to form a welding joint.

To further investigate the ultrasonic bonding strength of MGs, researchers employed various characterization methods, including mechanical tensile testing, bending testing, micro-hardness, and nanoindentation. The results show that the tensile strength of the welded samples reached ~1512 MPa, which is ~94.2% of the cast sample (~1615 MPa). This value is higher than that achieved by other welding technologies, such as laser welding, which typically reaches about 93% of the cast sample's strength. These findings are illustrated in Fig. 9a. The bending strength in the three-point bending test reached ~2930 MPa, nearly matching ~3109 MPa of the cast sample, as shown in Fig. 9b. Fig. 9c shows the Vickers hardness distribution in the section of the Zr-based joined sample. The hardness has a slight increase with the increase of joining energy in the joined seam, with 700 J samples achieving the same hardness as the as-cast MG. These findings suggest that under ultrasonic vibration, the oxide layer at the interface of heterogeneous MGs gradually disappears, exposing fresh interfaces that integrate as the energy increases. The welded joints exhibit excellent mechanical properties, providing a viable strategy for connections in extreme environments such as ocean engineering, aerospace engineering, and polar exploration.

Furthermore, Ma *et al.* [24] have explored the fabrication of more complex structures using UW techniques. As depicted in Fig. 9d, framework structures were crafted. Inspired by common industrial threaded connections, a method was designed to embed MG into threaded structures during the welding process, capitalizing on the softening characteristic of MG, as shown in Fig. 9e. Additionally, a riveting process was developed using MG as rivets to connect alloy plates, drawing on traditional riveting methods, as shown in Fig. 8f. The research team has also utilized the unique softening phenomenon of MG during ultrasonic vibration to create various special structures, such as grating

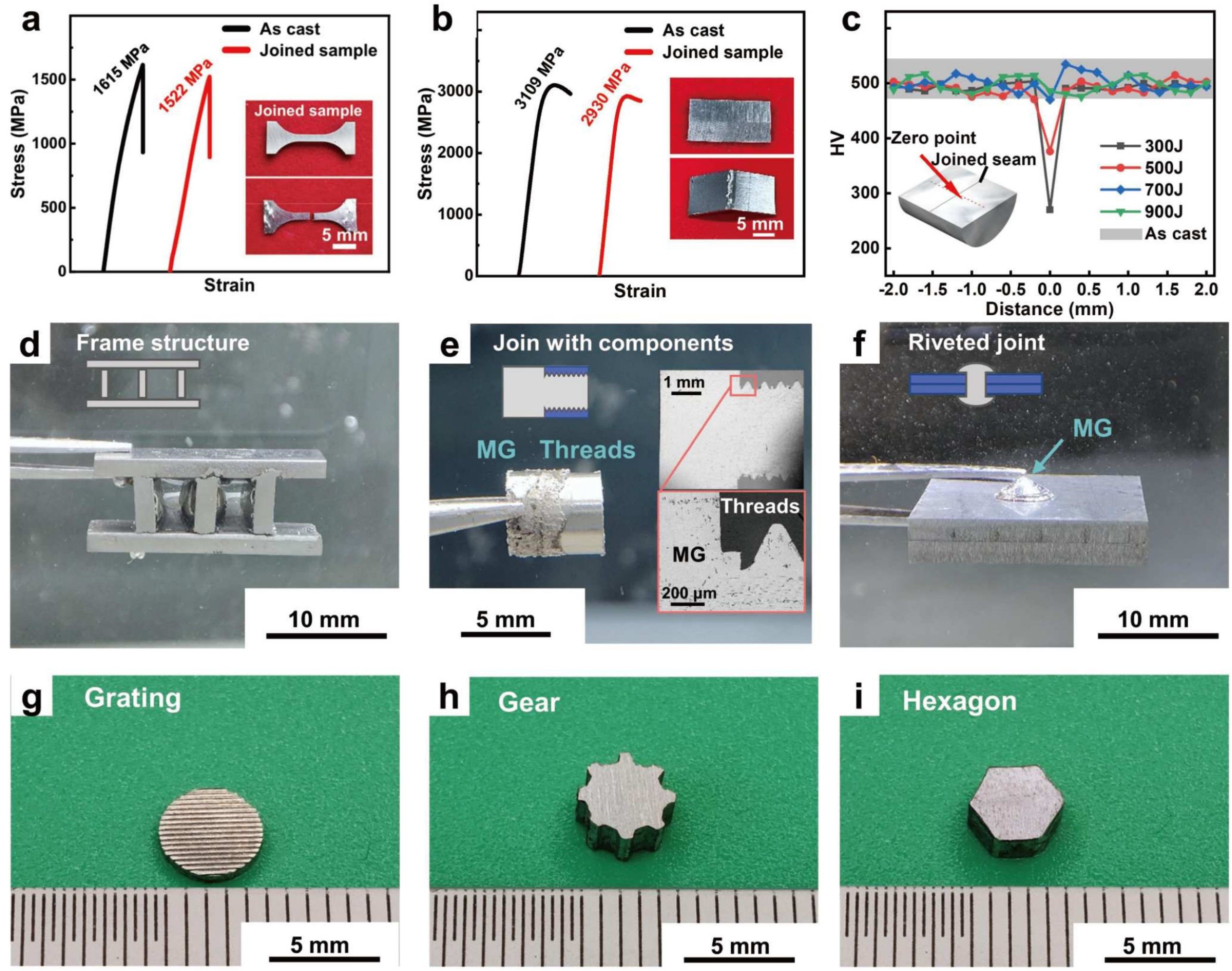

Figure 9 Mechanical property and images of UW and UVIP samples. (a) Tensile strength comparison for the as-cast Zr-based MG and joined sample under room temperature. (b) Bending strength comparison for the as-cast Zr-based MG and joined sample. (c) The micro-hardness test results of 20 dots in the longitudinal section of Zr-based joined samples under different energy. Different complex joining types under liquids: (d) frame structure, (e) joining with threads based on the flow deformation of MGs under ultrasonic vibration and its section images in the inset, and (f) riveting of steel parts based on the ultrasonic softening properties of MGs. Reprinted with permission from Ref. [24]. Copyright 2023, Springer Nature. Images of (g) grating, (h) gear, and (i) hexagonal by UVIP under seawater. Reprinted with permission from Ref. [44]. Copyright 2024, Elsevier.

structures, gears, and hexagons, as shown in Fig. 9g–i [44]. Due to the susceptibility of MG to shear band-induced softening, which often results in catastrophic failure during deformation, the unique plastic softening phenomenon observed under ultrasonic vibration, known as UVIP, is particularly significant. This phenomenon enables the UW of MG in a variety of liquid environments, including water, seawater, alcohol, and liquid nitrogen, demonstrating the broad applicability of ultrasonic vibration in extreme liquid conditions. The development and application of UW technology offer a potential solution for welding operations in extreme environments and are expected to play a pivotal role in future industrial applications.

CONCLUSIONS AND FUTURE OUTLOOK

The review highlights the promising development and application prospects of UW technology in the field of MG. UW stands out for its ability to join MGs without compromising their amorphous nature, offering a new solution for MG bonding. Utilizing the mechanical energy from high-frequency vibrations, UW can rapidly join MGs in a low-stress and heat-free manner, typically within one second. The advantages of technology are particularly evident in its application under extreme conditions, such as freshwater, seawater, alcohol, and liquid nitrogen, making it a promising strategy for manufacturing in offshore, polar, oil and gas, and space environments. UW is insensitive to vacuum and microgravity conditions, does not involve material melting, arc generation, or spatter, and does not require filler wires. It is not dependent on gravity-induced flow for material movement in the welding area, making it easily automatable and suitable for in-orbit aerospace welding applications. Moreover, this technology has been successfully applied to join various types of MGs, including Zr-, La-, Ni-, Ti-, Fe-, Pt-, Pd-, and Cu-based MGs, demonstrating its versatility and potential in designing new amorphous composite materials and developing new properties. Building on this foundation, UAM could be developed to prepare 3D shapes and form robust MG parts. As the technology advances, UAM may become an indispensable manufacturing technology for future space exploration and extreme environment applications.

However, the mechanical characterization of ultrasonically welded joints of MGs, particularly in terms of tensile and flexural strength, is an area that requires further exploration to fully elucidate the potential advantages of these joints. Currently, the scale of MGs that can be welded using ultrasonic methods is limited to the centimeter level. This limitation poses a significant challenge for the application of ultrasonic welding in large-scale MG components, as it restricts the size and complexity of structures that can be fabricated. Expanding this capability to decimeters or even meter sizes could significantly enhance the demonstration of the effectiveness of UW and broaden the application areas for MGs. Such scale-up would enable the realization of large-size structural applications of MGs in fields such as architectural and marine engineering. To address this challenge, future research should focus on developing advanced UW equipment with higher power capacities and improved energy distribution systems to handle larger components. Additionally, optimizing welding parameters and exploring hybrid techniques that combine UW with other advanced manufacturing methods could provide potential solutions for scaling up the welding process. Lastly, the mechanism underlying the UW of MGs, which is considered a unique phenomenon due to the distinctive structure of MGs, is an area that demands further research to develop a more comprehensive and universally applicable theory. Understanding this mechanism is crucial for optimizing the welding process and ensuring consistent and reliable joint formation across different MG systems.

In conclusion, UW technology offers significant potential for joining MGs without compromising their amorphous structure, making it a valuable tool for the fabrication and shaping of larger MG components, as well as for applications in extreme environments and space exploration. However, further research is necessary to enhance the mechanical properties of welded joints and scale up the welding capabilities to larger MG components. Additionally, a deeper understanding of the underlying welding mechanism is crucial for developing a more comprehensive theory applicable to various MG systems.

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金属玻璃的超声焊接研究进展

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摘要 金属玻璃(MGs)因其独特的高强度、卓越的耐腐蚀性和与众不同的物理性能组合而备受认可，使其在先进工程应用中具有巨大潜力. 然而，金属玻璃在部件制造中的实际应用常常受到诸如尺寸有限、室温下缺乏可塑性等限制. 为了克服这些挑战，超声波焊接(UW)凭借其固态连接特性，已成为连接和加工金属玻璃的有效方法. 本综述强调了UW在增强金属玻璃可制造性方面的优势作用. 具体来说，UW具有热影响低和加工时间快的特点，同时在极端环境(包括深海、极地和航空航天环境)中也显示出适用性. 此外，综述还详细比较和总结了UW连接金属玻璃的机制，并强调了UW的潜在挑战和应用.