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Multiscale Hierarchical Structure and Laminated Strengthening and Toughening Mechanisms

Baoxi Liu, Lujun Huang, Lin Geng and Fuxing Yin

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Abstract

Metal matrix composites with multiscale hierarchical structure and laminated structure have been developed to provide a novel route to achieve high strength, toughness and ductility. In this chapter, a lot of scientific research has been carried out in the preparation, processing, properties and application of metal matrix composite. Many toughening mechanisms and fracture behavior of composites with multiscale hierarchical structure and laminated structure are overviewed. It is revealed that elastic property and yield strength of laminated composites follow the “rule of average.” However, the estimation of fracture elongation and fracture toughness is complex, which is inconsistent with the “rule of average.” The fracture elongation of laminated composites is related to the layer thickness size, interface, gradient structure, strain hardening exponent, strain rate parameter and tunnel crack, which are accompanied with crack deflection, crack blunting, crack bridging, stress redistribution, local stress deformation, interfacial delamination crack and so on. The concept of laminated composites can be extended by applying different combination of individual layer, and provides theoretical as well as experimental fundamentals on strengthening and toughening of metal matrix composites.

Keywords: metal matrix composites, multiscale hierarchical structure, laminated structure, rule of average, tunnel crack

1. Introduction

Since the 1980s in the last century, as a result of the significant progress in the material science and technology, many metal matrix composites (MMCs) reinforced with fiber,

whisker and particle reinforcements, especially titanium matrix composites (TMCs), have been developed in many applications in order to improve special stiffness, special strength and thermal stability, such as automobile, nuclear, aerospace, armor and sectors and so on [1]. However, MMCs usually exhibit low fracture elongation and toughness in comparison with monolithic metal and alloys due to the addition of reinforcements, which seriously limits their applications [2]. Herein, control of the microstructure and interface characteristics are the important keys to the development of high strength and toughness MMCs and have been achieved by thermomechanical, heat treatment, alloy composition, reinforcement selection and reinforcement surface modification methods [3]. These favorite methods contain many strengthening effects, such as grain refinement, dispersion, solution, work hardening and phase transition strengthening mechanisms. However, grain refinement strengthening is the only method to improve the strength without decreasing fracture elongation, but it is usually no longer suitable when the grain sizes fall below 1 μm [4]. Therefore, it is believed that innovative design of metal matrix composites should make a guide to improve the comprehensive mechanical properties of superior strength, toughness and fracture elongation [2].

Recently, more and more scientists have realized that the nature has provided numerous smart structures for desired mechanical and functional properties after many billion years of stringent evolution [2]. A good idea was inspired from bio-inspired microstructures in nature. Nature materials always reveal multiscale hierarchical structures, such as nacre, which contains laminated, brick, protrusions, bridge and network structures from macroscale, meso-scale, microscale, nanoscale and even the atom-scale as shown in **Figure 1**. They can perfectly delicate themselves at different levels and play a synergistic strengthening and toughening effect during the deformation and fracture process, which results in a superior combination of performances, such as the 3000 times higher toughness of mollusk shells than their individual constituents [5].

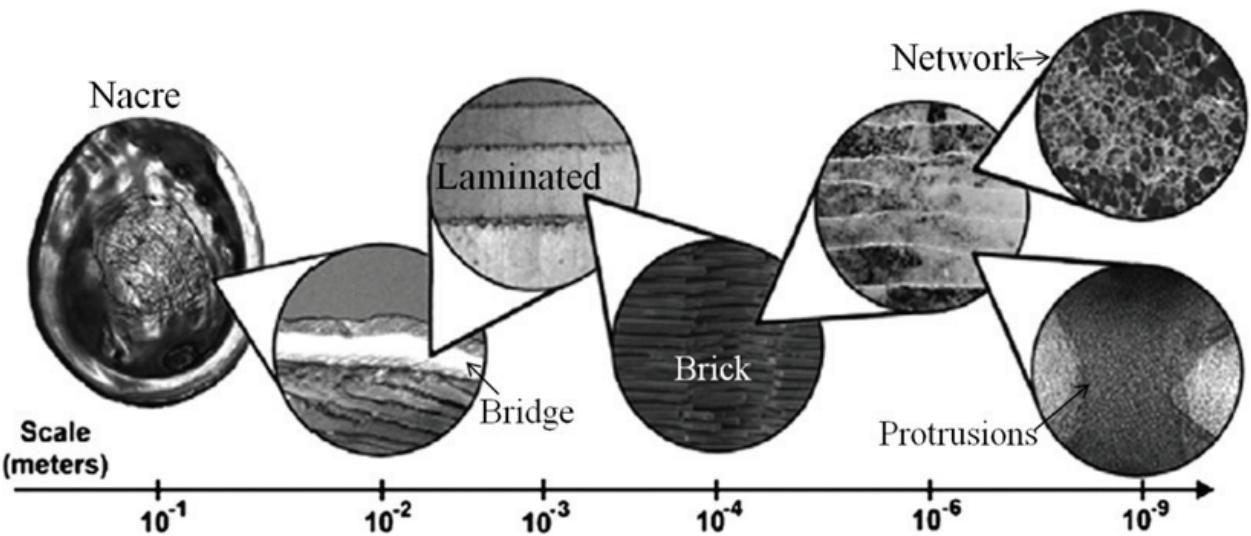


Figure 1. The multiscale hierarchical structures of nacre at several length scales [2, 5].

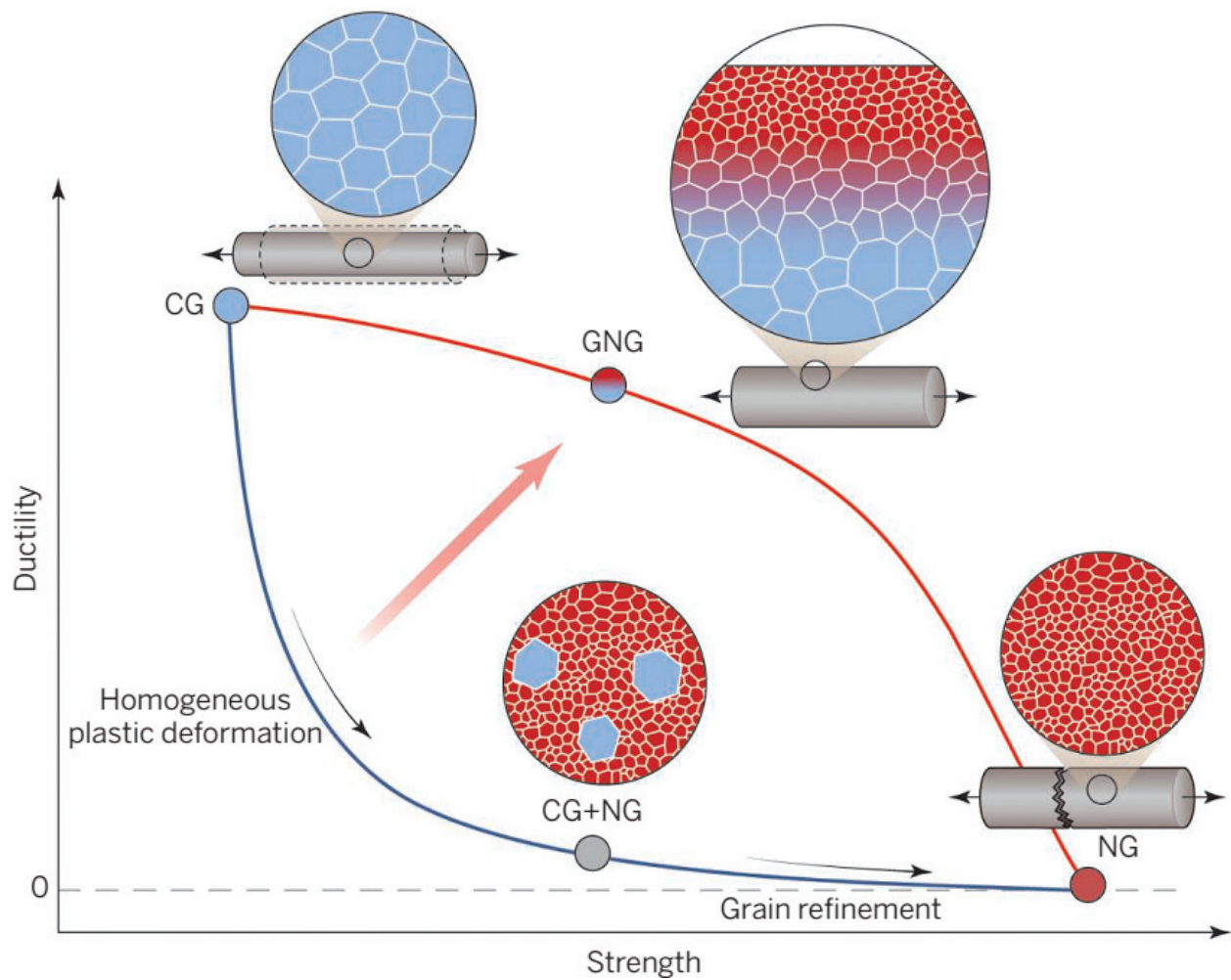


Figure 2. Strength-ductility synergy of metals with gradient grained structure [4].

In 2010, Lu [6] proposed that the metal matrix composites can obtain superior tensile strength and fracture elongation by designing multiscale hierarchical structures. For example, surface mechanical grinding treatment (SMGT) can induce gradient grain distribution and laminated structure in Cu and Ni matrix, respectively. The inspired hierarchical structure is useful to the enhanced fracture toughness of metal matrix composites apart from the monolithic MMCs as shown in **Figure 2** [4, 7, 8]. Palkovic et al. [9] reported that slurry can be designed by the ideal of multiscale hierarchical structures, which can be expected to replace the silicate cement with huge energy consumption. Zheng et al. [10] fabricated multiscale hierarchical metallic metamaterials by three-dimensional (3D) architectures as shown in **Figure 3**, obtaining superior tensile elasticity of 20%.

On the basis of ideal that metal matrix composites with multiscale hierarchical structures can obtain superior mechanical properties, this proposal aims to synthesize laminated Ti-(TiBw/Ti) composites by using powder metallurgy, which contains laminated, network and needle-like structures as shown in **Figure 4**, in order to achieve reinforcing, toughening and plasticizing effect, and to clarify the influence rule of fabrication parameters, laminated, interfacial and network structure parameters on microstructural evolution and mechanical behavior of

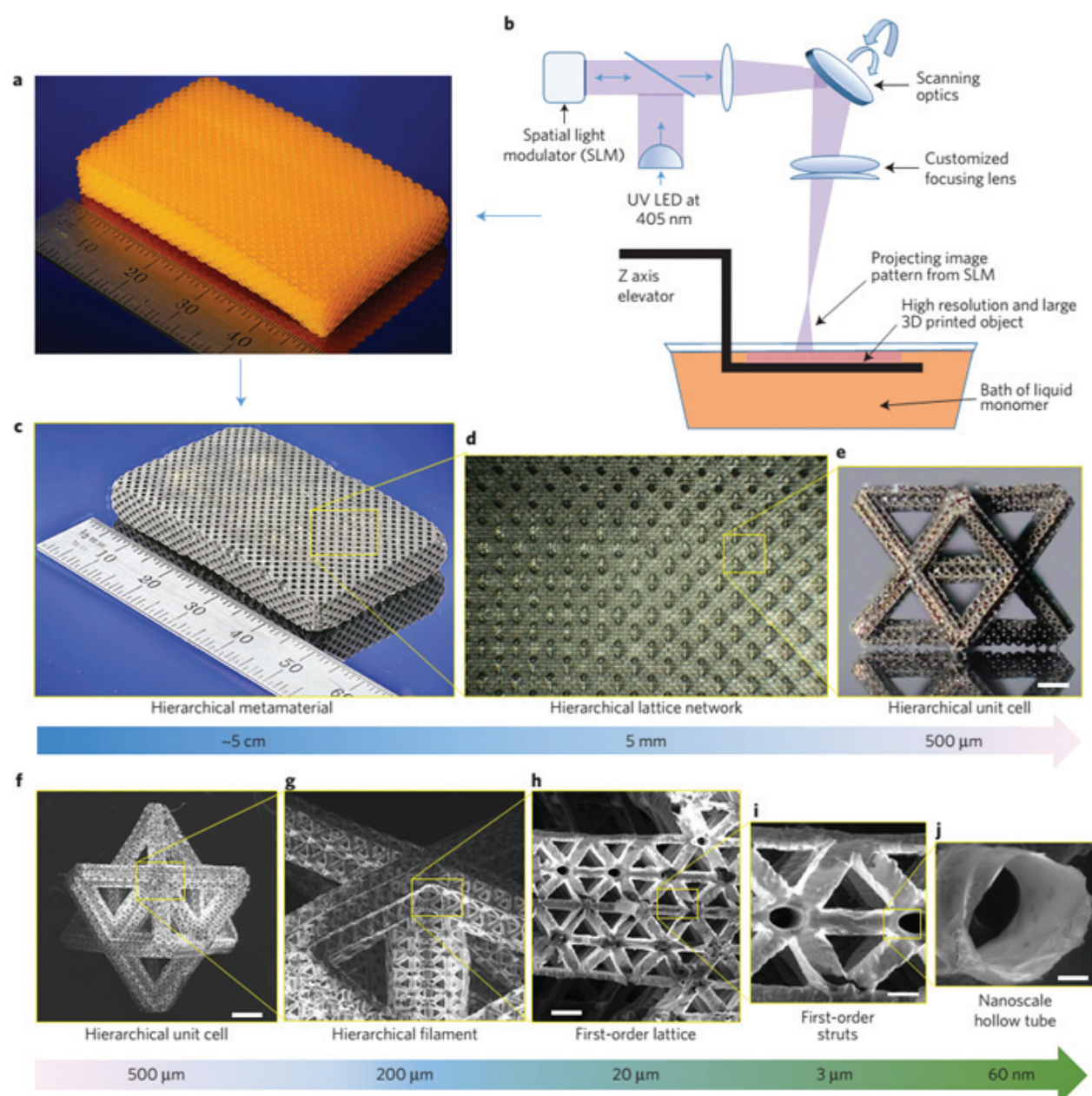


Figure 3. Multiscale hierarchical metallic metamaterials (a-f) [10].

titanium matrix composites. By using tensile and bending tests, the influence regular pattern of sintering temperature, layer thickness, layer thickness ratio and reinforcement volume fraction on mechanical properties should be investigated. It may solve the inverted shortcoming between strength and toughness. Through design and optimization of titanium matrix composites with two-scale hierarchical structures, this work will provide theoretical as well as experimental fundamentals on strengthening and toughening of metal matrix composites.

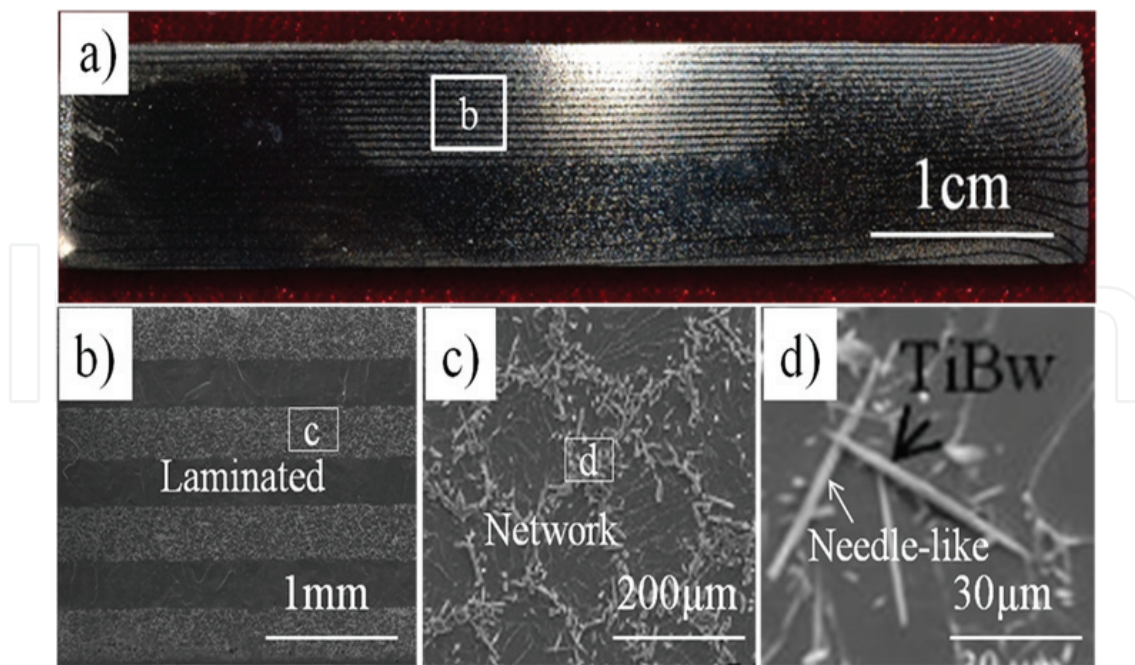


Figure 4. The multiscale hierarchical Ti-(TiBw/Ti) composites. (a) Macrograph, (b) laminated structure, (c) network structure and (d) needle-like structure [2].

2. Multiscale hierarchical structure

Yin [11] concluded that the one-scale hierarchical distribution structure of reinforcements contains dual matrix, network, laminated, gradient structural and other bi-continuous types. **Figure 5** shows the four structures in the titanium matrix composites reinforced with TiBw. Morsi and Patel et al. [12–14] fabricated a so-called “dual matrix” Ti-TiB composites as shown in **Figure 5(a)**, which contains isolated reinforcement-rich phase and continuous Ti matrix phase, exhibiting superior fracture toughness. Huang et al. [2, 15, 16] fabricated one-scale network hierarchical TiBw/Ti composites and two-scale network hierarchical TiBw + Ti₅Si₃/Ti composites as shown in **Figure 5(b)**. The elastic modulus, passion ratio and yield strength reach to the upper limit of Hashin-Shtrikman theory, as well as high oxidation resistance, superior fracture elongation, toughness and tensile strength. Han et al. [17] fabricated the laminated Ti-(TiB + La₂O₃)/Ti composites by powder metallurgy and hot rolling process as shown in **Figure 5(c)**, which results in an improvement of 30% in elongation compared with monolithic composites with same volume fraction of reinforcements. In 2003, Panda et al. [18–21] fabricated TiB/Ti and TiB₂/Ti functional gradient composites by powder metallurgy as shown in **Figure 5(d)**, which effectively weakens the thermal residual stress and inhibited the formation of cracks.

Figure 6 shows the fracture characteristics of the “dual matrix” Al₂O₃/Cu composites. The crack propagated along the Cu matrix and Al₂O₃ ceramic interface or inside the Cu matrix as

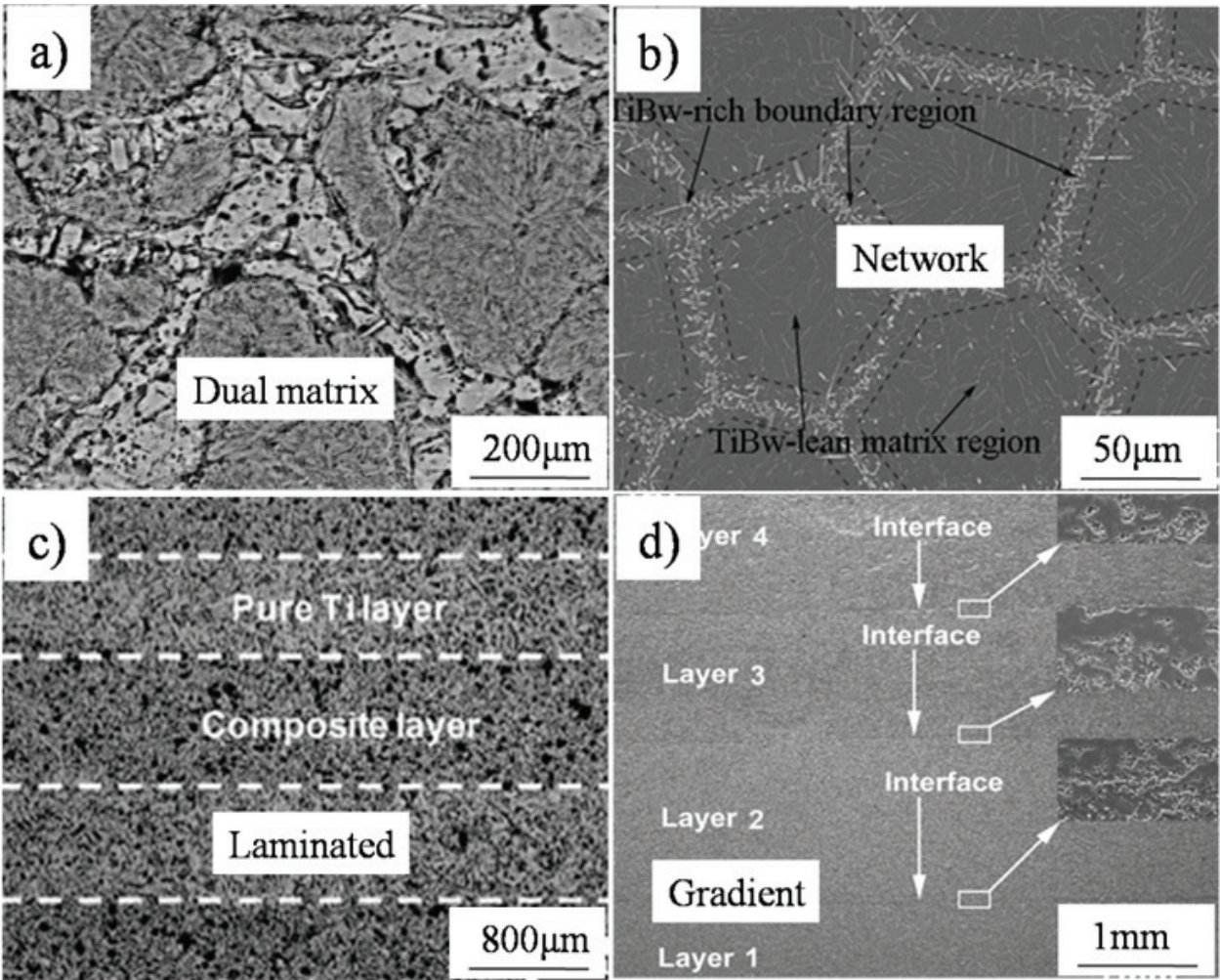


Figure 5. Four structural types of TiBw/Ti composites. (a) Dual matrix [12], (b) network [2], (c) laminated [17] and (d) gradient structure [18].

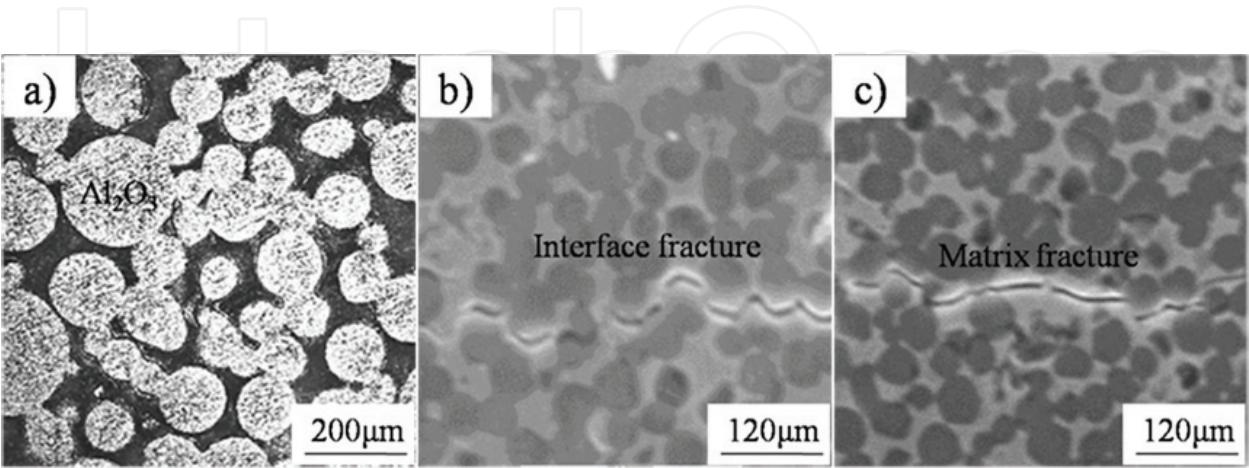


Figure 6. The fracture morphology of Al₂O₃/Cu composites [22]. (a) Microstructure, (b) interface fracture and (c) matrix fracture.

shown in **Figure 6(b, c)**. The two tortuous propagation paths absorbed a lot of fracture energy and plastic deformation work, respectively, which are benefit to the high fracture toughness compared to the popular belief that crack propagation occurs in the brittle ceramic phase. Herein, the interface fracture is attributed to the high thermal tensile stress at the interface developed during the fabrication process [22].

Huang et al. [2, 23, 24] concluded that the titanium matrix composites with network structure correspond to the upper limit of Hashin-Shtrikman (H-S) bound. Based on H-S bounds proposed by Hashin and Shtrikman in 1963 [25, 26], the reinforcement with network structure can be viewed as “stiffer” phase, which is continuous and encapsulates the titanium matrix as shell structure. The elastic modulus of this structure can be predicted by the H-S upper bounds. However, the titanium matrix composites with homogeneous distribution of reinforcement can be taken as that the soft phase encapsulates the stiffer phase, whose elastic modulus is near to the lower limit of H-S bound, and the H-S bounds can be expressed as follows [2]:

$$E_{HS-upper} = \frac{E_w[E_w V_w + E_m(2 - V_w)]}{E_m V_w + E_w(2 - V_w)} \quad (1)$$

$$E_{HS-lower} = \frac{E_m[E_m(1 - V_w) + E_w(1 + V_w)]}{E_w(1 - V_w) + E_m(2 - V_w)} \quad (2)$$

where E_w and E_m are the elastic modulus of whisker reinforcement and matrix, respectively, V_w and V_m are the volume fractions of the whisker and matrix, respectively. The network structure can enhance the strengthening and toughening effect at room temperature, which is similar with the grain-boundary strengthening mechanism. At high temperature, it can also overcome the grain-boundary weakening behavior. Therefore, TiBw/Ti composites with network structure obtain superior room and high-temperature properties as shown in **Figure 7**.

Figure 8 shows the illustration diagram of oxidation and fracture resistant mechanisms of titanium matrix composites with network structure [2]. After oxidation process of 140 h at 973K, the surface of titanium matrix composites with network structure is still smooth without any spallation and cracks. The in situ reacted TiC particle forms a dense continuous wall as shown in **Figure 8(b)**, which can effectively inhibit the oxidation velocity as shown in **Figure 8(c)** [2, 27].

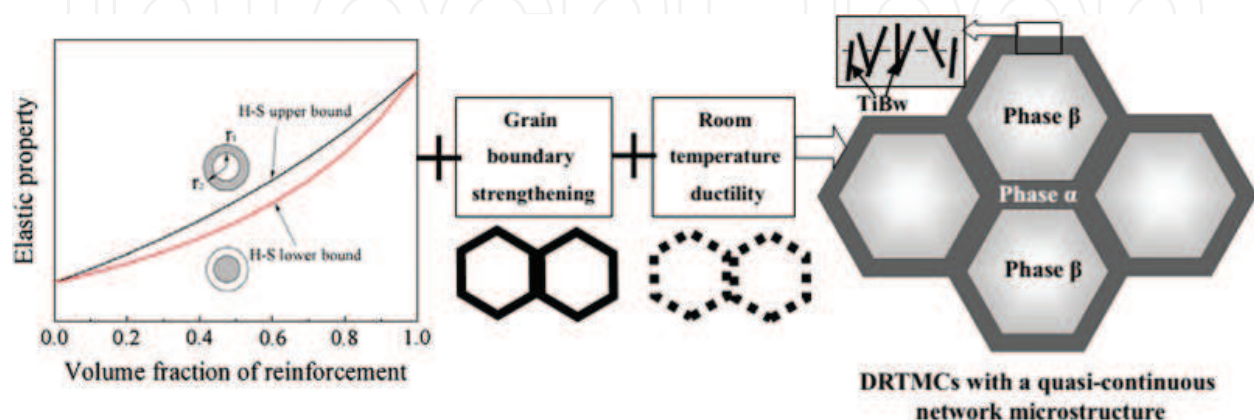


Figure 7. It shows the strengthening and toughening mechanisms of TiBw/Ti composites.

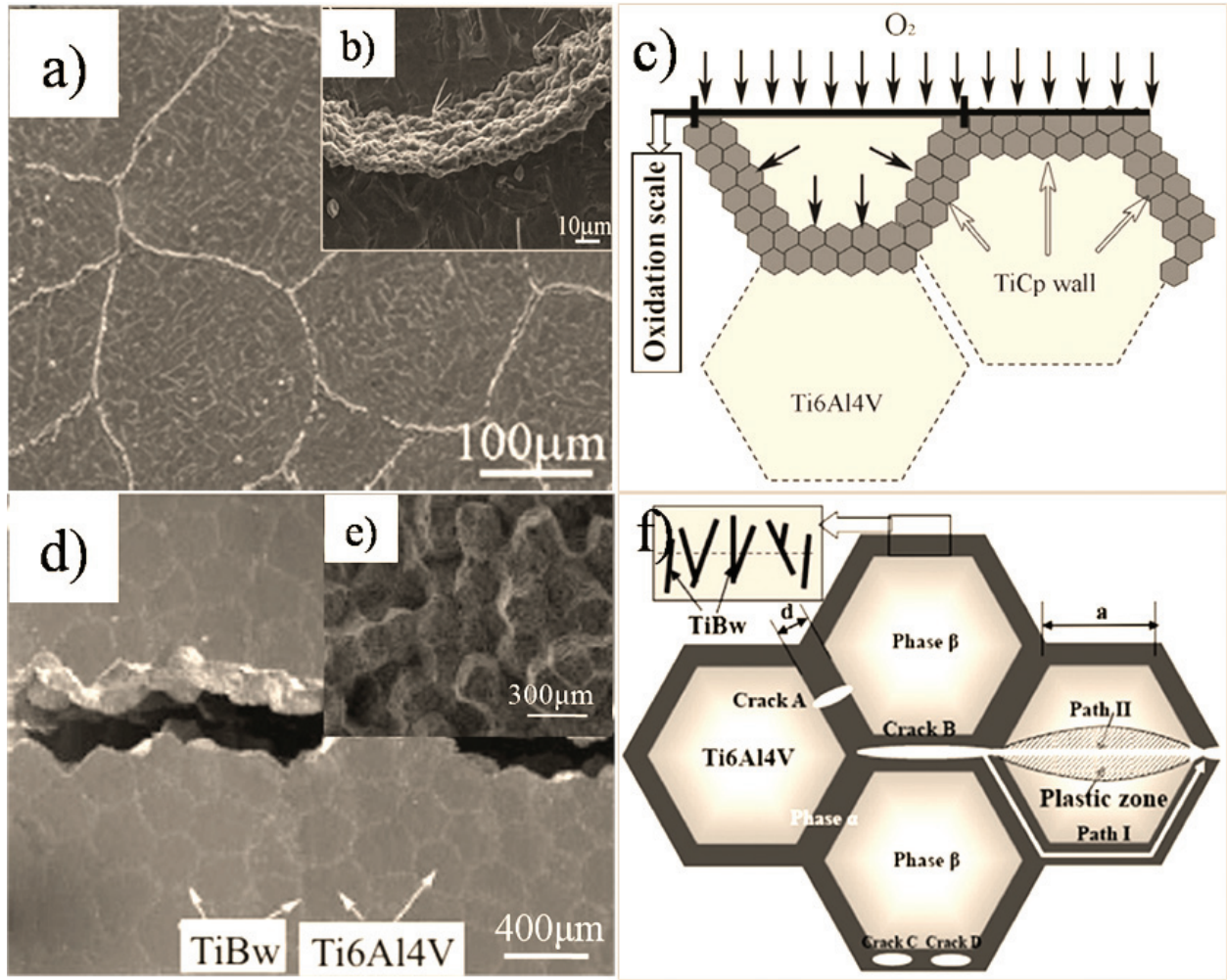


Figure 8. Schematic illustration showing the oxidation and fracture resistant mechanisms of titanium matrix composites with network structure [2]. (a–c) Oxidation and (d–f) fracture resistance.

During the fracture process, the TiBw/Ti6Al4V composites are mainly fractured at the network boundary as shown in **Figure 8(d, e)**, and the matrix tearing is also the fracture characteristics with low volume fraction of reinforcement. **Figure 8(f)** illustrates the crack propagation paths and toughening behaviors of TiBw/Ti6Al4V composites with low volume fraction [2]. Crack A shows that the crack perpendicular to the network boundary cannot propagate through the softer matrix phase because of low crack size, which can play strengthening and toughening effect in the composites. However, the different propagation paths along path I or path II mainly depend on their absorbing energy. The effect reinforcement strength and absorbing energy can be expressed as follows [2, 28]:

$$\sigma = \sqrt{\frac{\pi EG}{2(1 - \nu^2)d}} \quad (3)$$

$$Q = \sigma \cdot S \cdot L = AL \sqrt{\frac{\pi EG}{2(1 - \nu^2)d}} \cdot d^2 = AL \sqrt{\frac{\pi EG}{2(1 - \nu^2)}} \cdot d^{\frac{3}{2}} \quad (4)$$

where σ is the effective reinforcement strength and G is the critical strain energy release rate for the dynamic propagation of a crack. ν is the Poisson's ratio, d is the reinforcement size along the crack direction, S is the crack surface area, L is the open crack length and A is the parameter related to the surface and length.

The absorbing energy of crack propagation along path I (Q_I) and II (Q_{II}) can be deduced as Eqs. (5) and (6), respectively [2].

$$Q_I = AL \cdot B \sqrt{\frac{\pi E_I G_I}{2(1 - \nu_I^2)}} \cdot (3a)^{\frac{3}{2}} \quad (5)$$

$$Q_{II} = AL \cdot B \sqrt{\frac{\pi E_{II} G_{II}}{2(1 - \nu_{II}^2)}} \cdot (2a)^{\frac{3}{2}} \quad (6)$$

Therefore, the crack can propagate along path I when $Q_I \leq Q_{II}$.

The concept of functionally gradient Ti-TiB composites arose as a result of efforts to prevent the cracking in TiB-Ti joints induced by the coefficient of thermal expansion (CTE) mismatch during high-temperature processing. Functionally gradient materials (FGM) effectively decrease the

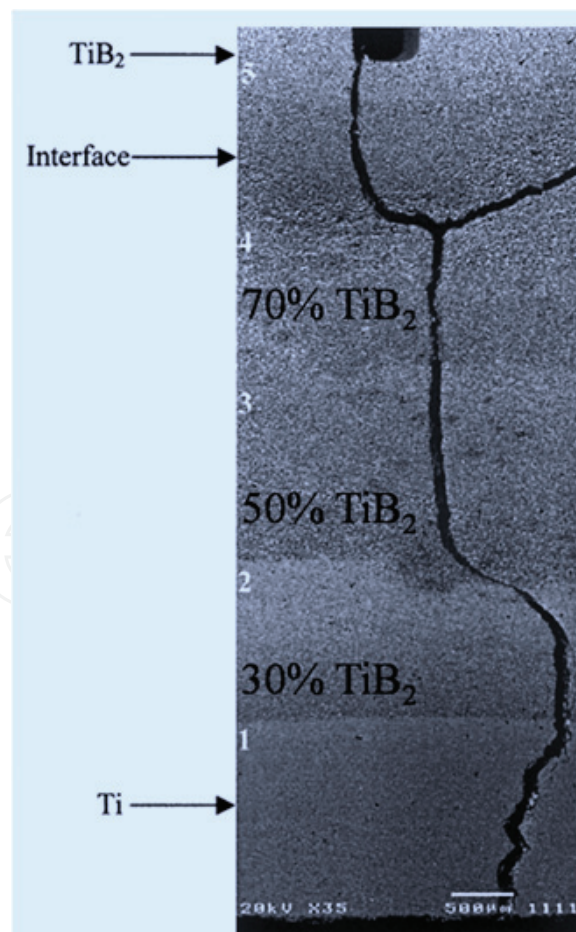


Figure 9. The crack propagation of functionally gradient TiB-Ti composites [29].

thermal stresses and delay the plastic yielding and the fracture failure of the metallic layer by the load shearing of the ceramic [18–21]. Meanwhile, it is a better overall use of the available materials by distributing them in a suitable spatial direction. Ma et al. concluded that the main crack deflected to the layer interface, and it is interesting to note that crack bifurcation was initiated at the interface between layers 5 and 4, resulting into the stepwise fracture process rather than catastrophic failure as shown in **Figure 9**. Moreover, it has good ballistic properties that make it popular armor application [21, 29].

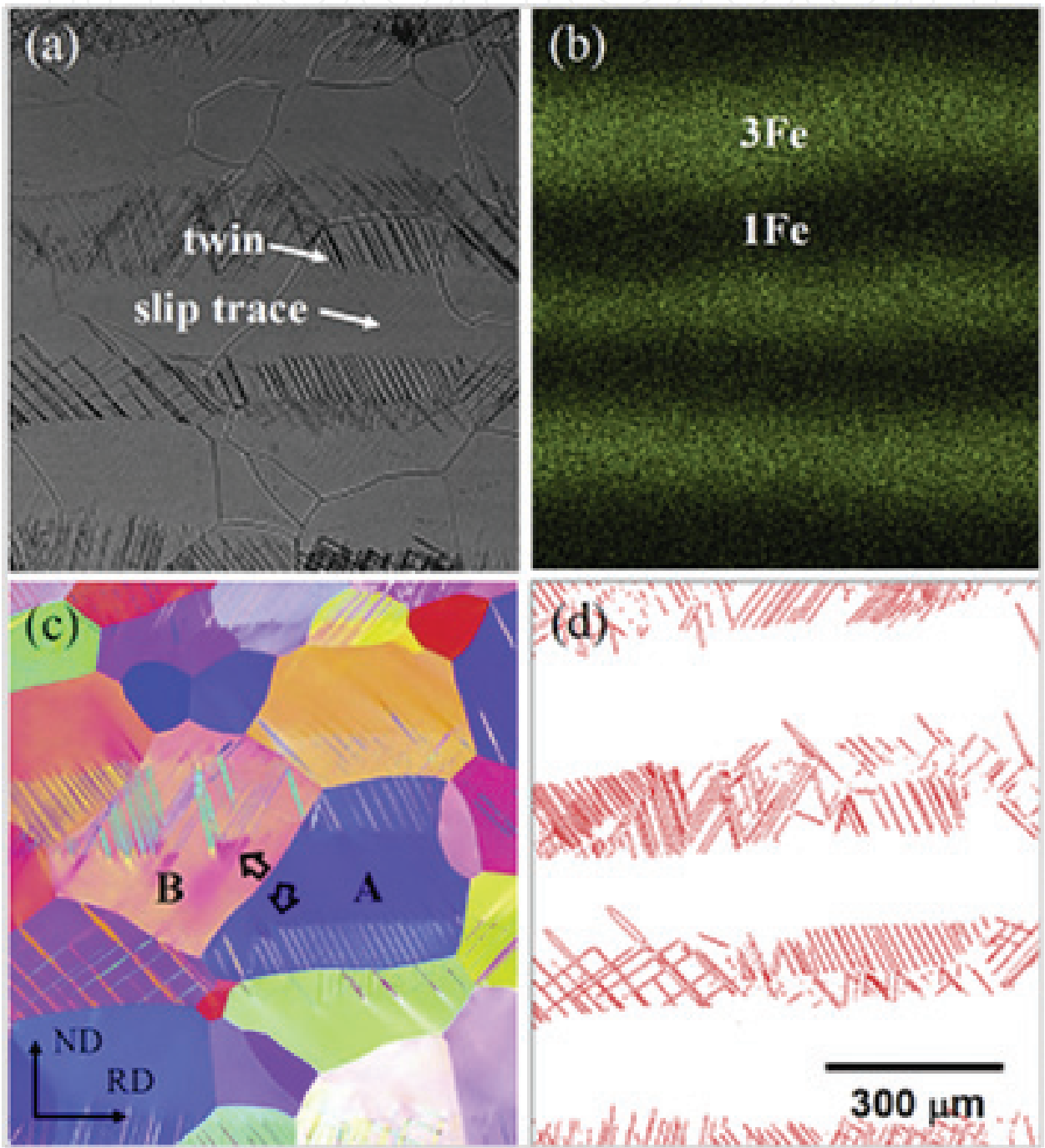


Figure 10. (a–d) $[332]\langle 113 \rangle$ Twinning and dislocation deformation of laminated Ti composites [30].

Min et al. [30, 31] fabricated a multilayered Ti-10Mo-1Fe/Ti-10Mo-3Fe composites by hot roll bonding and subsequent heat treatment as shown in **Figure 10(a)**. The layer interface can be detected by Fe concentration as shown in **Figure 10(b)**. It is interesting to note that Ti-10Mo-1Fe layer was deformed by twinning, whereas deformation characteristic of Ti-10Mo-3Fe is dislocation slip as shown in **Figure 10(c, d)**. The artificially multilayered twinning/dislocation slip deformation characteristics can provide a high strength-ductility combination over a wider range to meet industrially applicable strength and formability requirements.

3. Laminated structure

Actually, composite materials with laminated structures have been a design ideal to improve the mechanical and functional properties from antiquity. “The Iliad” of homer has described that Achilles’s shield has five layers, and the sequence of laminates is bronze/tin/gold/tin/bronze. The Aeneas’s spear penetrated the first two layers but trapped by gold layer during combat, revealing superior mechanical properties. In 1837, an iron-laminated composite dating as far back as 2750 BC was found in the Great Pyramid in Gizeh, Egypt, and many layers of wrought iron have been inexpertly welded tougher by hammering as shown in **Figure 11** [32, 33]. Lamination architect ideal may be attributed to the following three reasons: (1) only thin sheets could be carburized in the earliest form of wrought iron, so lamination was a good way to create bulk composite materials; (2) it is no expensive to sandwich hard rare steel between more cheap common materials and (3) optimizing the combination of strength, toughness and sharpness is the basis for lamination from the mechanical view of materials. Therefore, many weapons were made by lamination design from ancient, such as Adze blade, Japanese sword, Merovingian blade and Chinese blade [33].

Since 1988, American scientists took the sandwich materials into the aircraft door C-17, and the investigations of laminated and functional gradient metal composites enter into the period of rapid development [34]. Jiang et al. [35] fabricated laminated Ti-Al₃Ti composites using reaction vacuum sintering in 2003. During the impact testing process, the laminated composites can absorb lots of impact works through forming delamination cracks, and it is expected to be applied in the armor equipment and ballistic system. In 2006, Japan government sponsored a research project named “the layer integrated steel and metals,” which aims to fabricate multi-layer steel with high strength (>1200 MPa) and superior tensile elongation (>20%) through the composite design of high strength steels and ductile steel layers [36]. In 2008, Inoue et al. [37] fabricated multilayered SS420/SS304 steel with high toughness and reported that fracture elongation is gradually increasing with the decrease of layer thickness, which is attributed to the decreasing size of tunnel cracks. Nambu et al. [38] reported that multilayered steel with strong interfacial bonding strength exhibits superior longitudinal fracture elongation compared with the laminated composites with weak interfaces. Moreover, they found that the laminated structures can change the deformation mode of martensite layer, transiting from poor work hardening to improved hardening [39].

Strength and toughness are the most important performance in traditional structural materials [1–3, 40]. Recently, with the development of national defense and civil sectors, such as aerospace, nuclear, shipbuilding, automobile and so on, a high demand of strength and

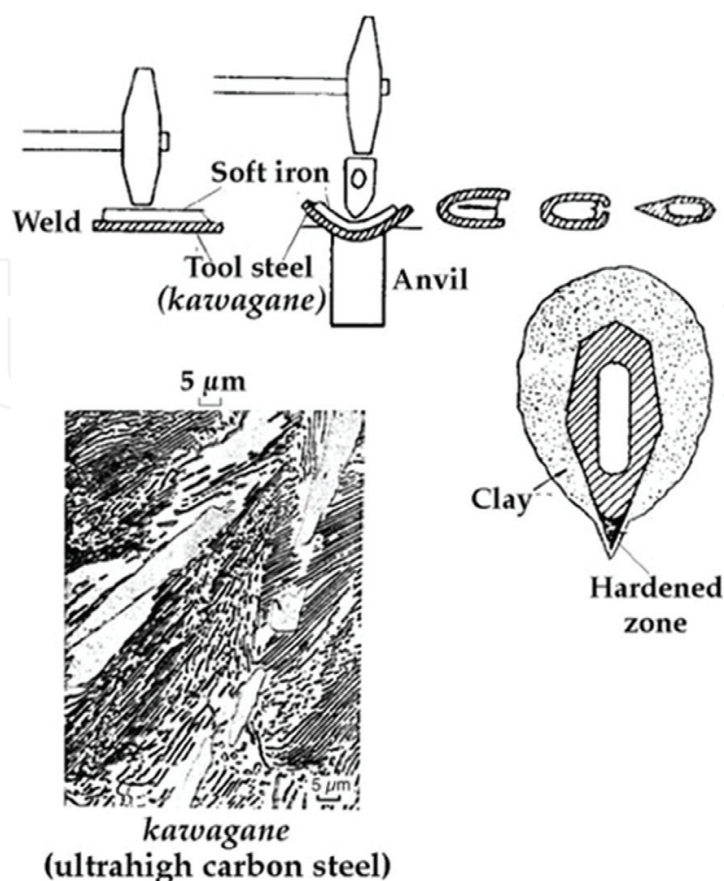


Figure 11. The fabrication process of slip blacksmiths [32, 33].

toughness is necessary for structural materials to adapt the increasingly stringent environment and application conditions [4, 6, 41]. For centuries, scientists have been conducting investigations to develop composite materials with laminated structures to improve strength and toughness [32–34, 42]. Typical laminated composites, such as ceramic-ceramic [43], metal-metallic glass [44], metal-ceramic [45], metal-metal [46], metal-metal composites [47] and metal-ceramic-intermetallic [48] systems have shown desirable structural properties as a result of many layers and interfaces, and these laminated composites allow for the possibility of combining the high ductility and toughness of soft layers with the high strength of hard layers.

Though the design concept, strengthening and toughening mechanisms of laminated composites are ancient and attempted for a long time, the relationship between microstructure and mechanical properties are still unclear and ambiguous. Lesuer et al. [32] concluded that the elastic property and yield strength of laminated composites follow the “rule of average.” However, the estimation of fracture elongation and fracture strength is complex, which is inconsistent with the “rule of average” as shown in **Figure 12** [40]. Therefore, the further investigation of the fabrication and structure factors of laminated composites may prove useful to understand the relationship between structure and performance. The results could be used as guide to design microstructures and improve mechanical properties by multilayer structure, which require a completely different approach from the investigations of conventional monolithic composite material.

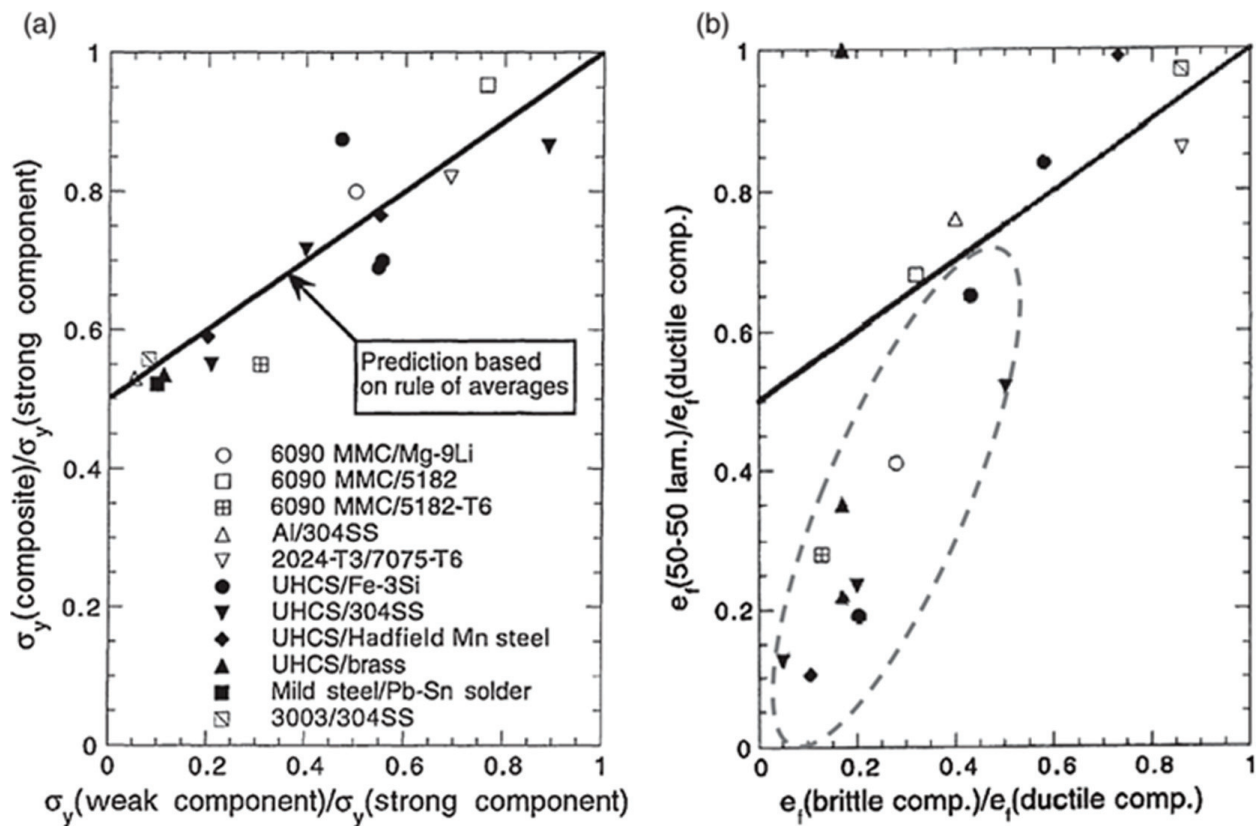


Figure 12. (a) Yield strength and (b) elongation of different metals consisting of 50% strong (of brittle) component and 50% weak (or ductile) component [40].

4. Strengthening and toughening mechanisms of laminated structure

Typical laminated composites have shown desirable mechanical properties, such as tensile strength, fracture toughness, compact toughness, formability, bending properties and fracture elongation. A number of papers have been published on the reinforcing and toughening of laminated composites and illustrated that laminated metal composites are especially effective in improving fracture-related properties by various intrinsic and extrinsic mechanisms [32, 49].

Recently, the strengthening and toughening mechanisms of laminated composites mainly focus on the investigation of plastic deformation, crack propagation and fracture morphology. Herein, Cetin and Hutchinson et al. [50–52] carried out the depth investigation on the tunnel cracks, plastic bifurcations and delamination cracks and found that the fracture elongation is related to the layer thickness, layer thickness ratio, yield strength, ultimate strength, interfacial bonding strength, work hardening index, strain rate sensitivity exponent and interfacial reaction phase of individual layer. Serror [53] reported that multilocalized necking of laminated composites is benefit to relieve the localized stress concentration and improve plastic deformation capability during maximum tensile testing process based on the plastic bifurcation theory and finite element method (FEM) as shown in **Figure 13**. Zhang et al. [54] fabricated multilayer Cu/Al sheets using rolling bonding method and found that laminated structure can effectively

delay the premature localized necking behavior of Cu sheet. They also investigate that nano-meter indentation behavior and found that size effect on the Cu/Cr, Cu/Au and Cu/Ni multi-layered composites by magnetron sputtering. The ultimate strength would obviously be enhanced with the decreasing layer thickness in the nanoscale. However, the ductility and fracture toughness of laminated composites are ambiguous due to the complex interaction between layer thickness and interfacial structure [55]. The contribution of this section is to draw the series of deformation and fracture mechanisms of laminated composites and to provide guide for the reinforcing and toughening effect research and design.

4.1. High fracture toughness of laminated composites

The typical natural laminated composite is nacre material with the block-wall structures as shown in **Figure 1**. The nacre contains mineral layer (CaCO_3) with the volume fraction of 99% and protein with the volume fraction of 1%. Nacre reveals high toughening mechanisms as follows: (1) the layer boundary with many projections reveals obvious bridging reinforcing effect; (2) interfacial delamination can play toughening effect when loaded on the nacre; (3) the staggered block-wall microstructures with a high sheet lap length and length/diameter ratio can decrease the relative displacement of fiber sheets during fracture process; (4) the mineral layer can be bonded by protein with three-dimensional network structure and (5) the nacre with convex-curved macroscopic morphology can state biaxial stress state, and the convex-curved plane only affords average compression stress, seldom of tensile stress and bending stress, which is a benefit to the ceramic matrix composites [5].

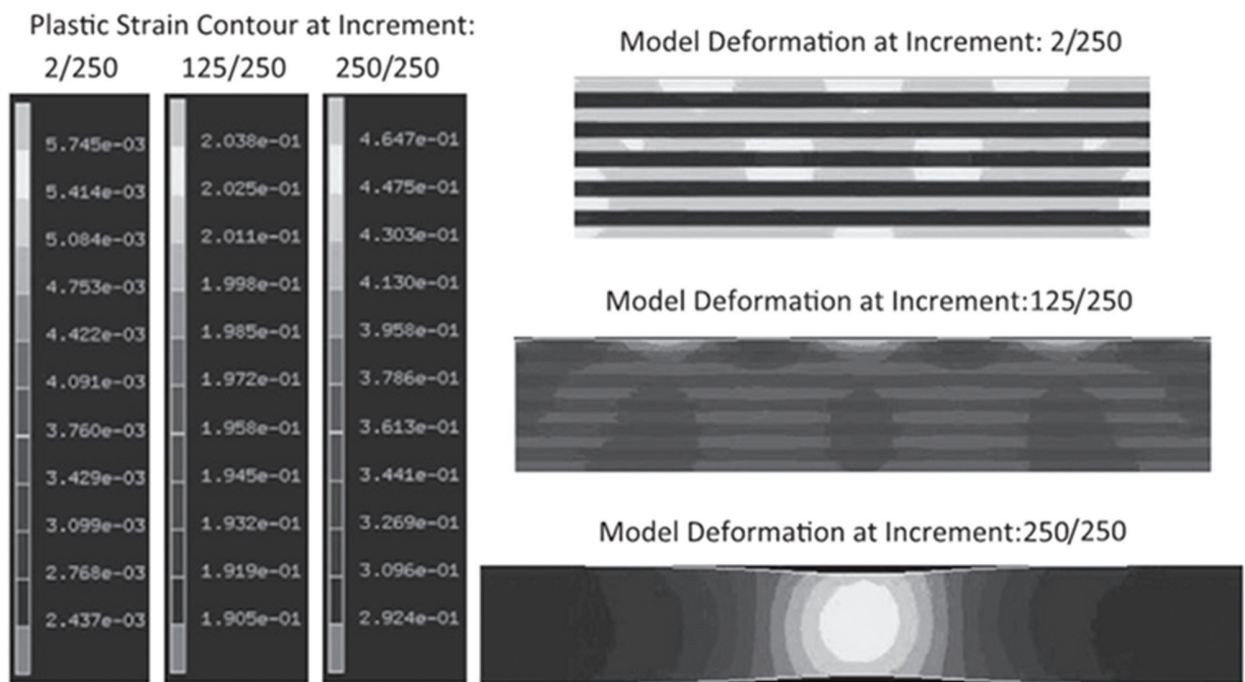


Figure 13. Three snapshots of the deformation and corresponding plastic strain contours of laminated composites under plane strain uniaxial tension [53].

Figure 14 shows the mechanical properties and toughening mechanisms of laminated ceramics with different interfacial status and constitute layers [56, 57]. Two kinds of laminated ceramic composites, the laminated composites with strong interface and weak interface, can be divided according to interfacial bonding strength. When the laminated ceramic composites afford bending force, the two kinds of laminated composites always improve the bending properties and fracture toughness through different toughening types. The laminated ceramic composites with strong interfacial strength possess high residual stress during fabrication process, and the main crack may appear to deflect or bifurcation on the strong interface during the fracture process induced by residual compression stress. Therefore, crack propagation path can be extended and the stress concentration at the crack tip can be mitigated, so the laminated ceramic composites with strong interfacial bonding strength can obtain high fracture toughness. The laminated ceramic composites with weak interfacial strength can also obtain high fracture toughness by delamination crack. When the main crack propagates into the weak interfaces forward, it also deflects or forms interfacial crack along the weak interface due to large amount of defects. The stress concentration can be relieved and the stress state can be changed into biaxial stress from complex triaxial stress, which resulted into the shielding phenomenon of main crack tip [58].

The improvement in fracture toughness of laminated ceramic composites can be mainly attributed to two categories: intrinsic and extrinsic mechanisms as shown in **Figure 15** [59, 60]. Intrinsic toughening mechanism implies in situ resistance of the microstructure (precipitates, particle spacing, grain size, morphology and so on) on crack propagation. For example, toughness can be enhanced by changing the nature, distribution and/or interface properties of second phase particles to suppress damage in the form of microcracking or microvoid

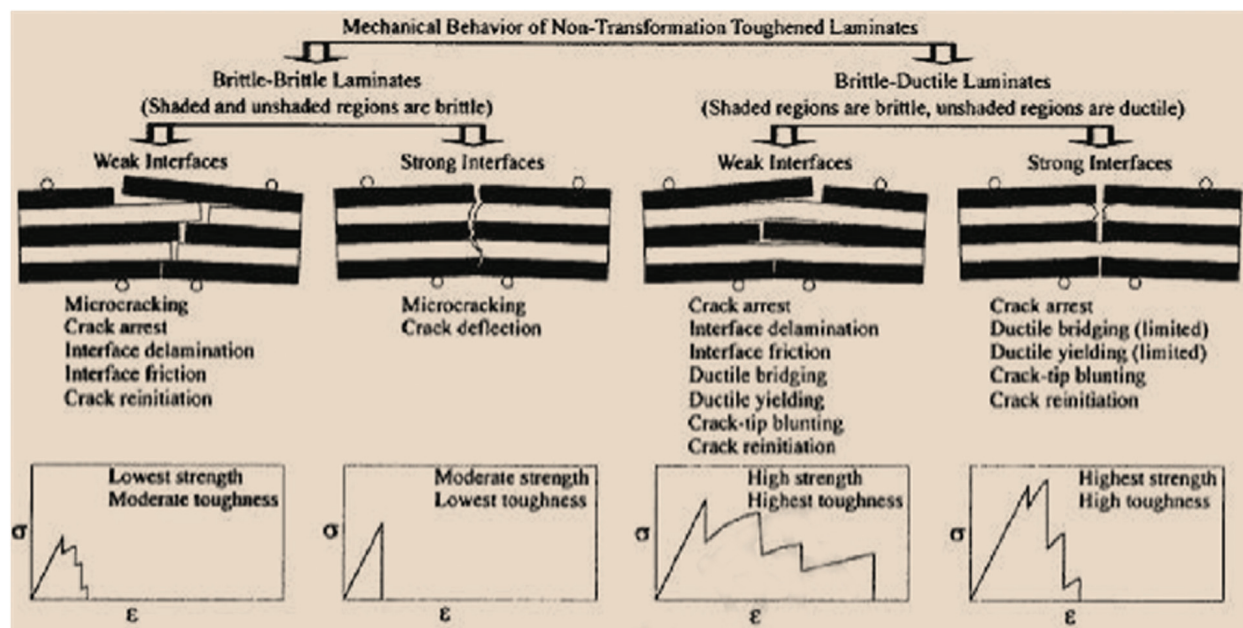


Figure 14. The mechanical properties and toughening mechanisms of laminated ceramics with different interfacial status and constitute layers [57].

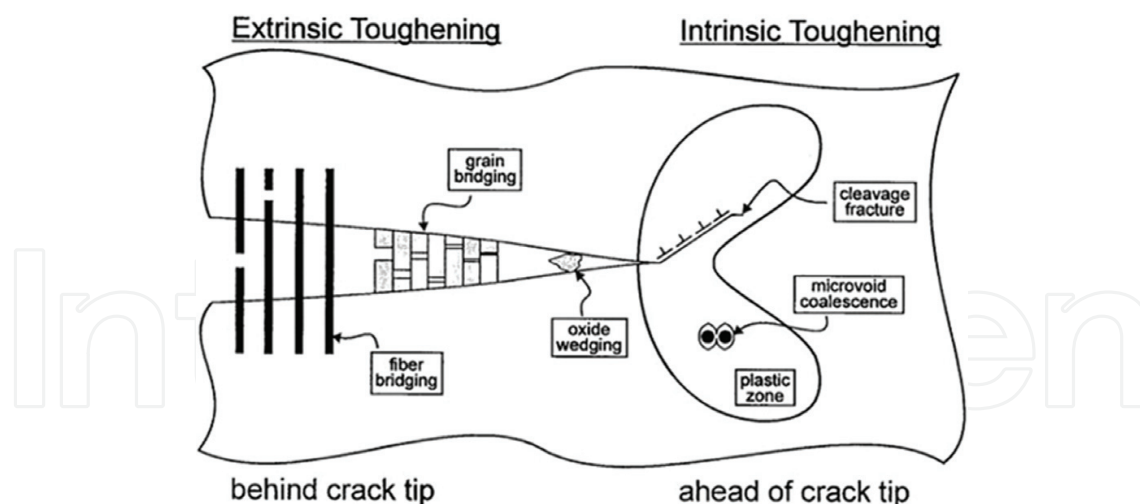


Figure 15. The intrinsic and extrinsic toughening mechanisms [60].

formation ahead of the crack tip. Extrinsic toughening mechanism indicates the microstructure behind of the crack tip to effectively decrease the crack driving force, which is initiated by the residual stress that reduces local stress intensity at the crack tip, such as delamination at weak interfaces [45], crack bridging [61], stress redistribution [32, 33], crack front convolution [32], local front convolution [33], crack trapping [48], ductile ligament [60], buckling [62], multiple cracking [63], crack bifurcation in the compressive stress layer [60], phase transformation under stress and compositional gradient [59] and crack deflection by mismatch of thermal expansion coefficients between different layers inevitably generates thermal residual stresses during subsequent cooling of laminated composites [64]. These behaviors need a significant amount of energy absorption and long crack propagation paths, resulting in an increase in fracture toughness as shown in **Table 1** [33]. Therefore, extrinsic mechanisms, on the other hand, improve fracture properties by providing alternative crack propagation routes or reducing the driving force for crack growth through various processes that shield the crack tip from the applied stress [65].

4.2. Gradient laminated distribution of microstructure

It is all known that metal with nanoscale grains can obtain superior compression properties but low tensile ductility, which is due to premature strain localization during the tensile processing [4, 6]. However, metal with coarse grains can obtain high tensile fracture elongation but low tensile strength, which is attributed low strain hardening capacity. What happens if different metals with nanometer grains and with coarse grain are combined? In 2011, Lu et al. [7] introduced nanosize layer on the surface of copper during the serious plastic deformation process by surface mechanical attrition technology. The grain size can be changed from nanoscale into microscale with the increase of depth gradually. The laminated copper with gradient grain size distribution can obtain superior tensile yield strength and high tensile ductility, and the tensile ductility can reach to 100% without crack initiation. In 2014, Liu et al. introduce high-rate shear deformation with strain gradients on bulk nickel, forming nanometer-scale laminated structure with 20 nm, which exhibits a superior hardness of 6.4 GPa [8]. Lu

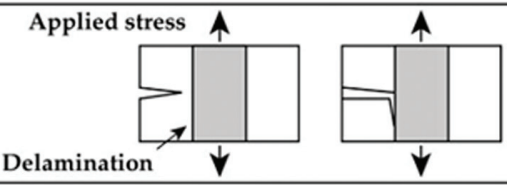
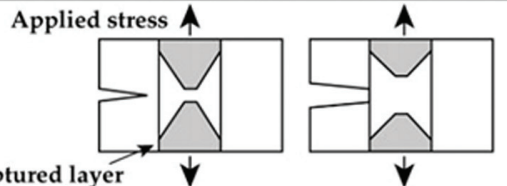
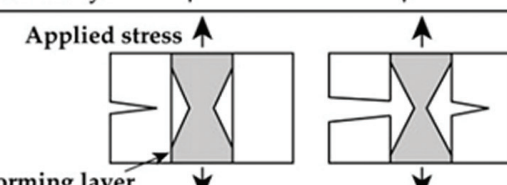
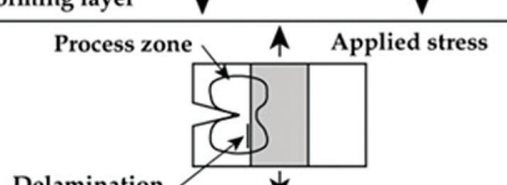
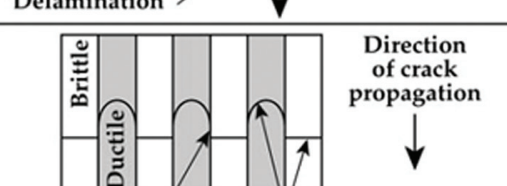
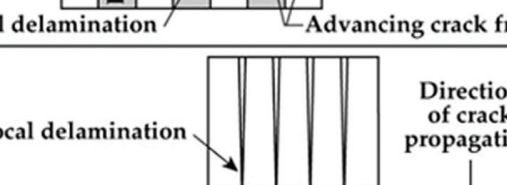
Mechanism (Testing orientation)	Volume fraction dependence	R-curve behavior possible	
Crack deflection (Crack arrester)	No	...	 Applied stress ↑ Delamination ↓
Crack blunting (Crack arrester)	No	...	 Applied stress ↑ Ruptured layer ↓
Crack bridging (Crack arrester)	Yes	Yes	 Applied stress ↑ Deforming layer ↓
Stress redistribution (Crack arrester, crack divider)	...	Yes	 Process zone ↑ Applied stress ↑ Delamination ↓
Crack front convolution (Crack divider)	Yes	Yes	 Brittle Ductile Local delamination Advancing crack front Direction of crack propagation ↓
Local plane stress deformation (Crack divider)	...	Yes	 Local delamination Advancing crack front Direction of crack propagation ↓

Table 1. Toughening mechanisms for laminated metal composites [33].

proposed that the gradient nanometer grained surface layer results in synergy effect of strength and ductility rather than the tradition trade-off between strength and ductility [4].

4.3. Size effect in the laminated composites

Gao et al. [66] reveal that the laminated composites become insensitive to flaw when the layer thickness is lower than the critical flaw size according to the research on nacre. The stress field becomes more and more uniform as the layer thickness decreases. The critical flaw size is expressed as follows:

$$L_{critical} = \frac{\alpha \Gamma E}{\sigma^2} \quad (7)$$

where α is crack propagation parameter, about 1.3–1.9. E is the Young's modulus, and Γ is the fracture energy of atomically sharp cracks. Kumar et al. proved that when the layer thickness is small enough compared to the critical flaw size, $L_{critical}$, no matter how high value of applied stress, even reach the ultimate strength of the material and the laminated composites will rupture wholly rather than classical damage mode, which experience nucleation, coalescence, growth and step-by-step crack propagation path. Once layer thickness is beyond the $L_{critical}$ and tunnel crack forms, the laminated composites will damage like domino, which does not need a lot of fracture energy. However, the wholly fracture process will spend more fracture energy. Therefore, laminated composites with small layer thickness can obtain more fracture strength and fracture toughness.

However, most experimental results reveal that the ductility of laminated composites is seriously limited with reducing layer thickness to a critical value (<100 nm), although fracture strength has been shifted. Zhang et al. [55] indicated that the fracture mode of the laminated Cu/Au and Cu/Cr composites changed from ductile shearing to brittle shearing with decreasing layer thickness as shown in **Figure 16**. Laminated composites with thick layer thickness exhibit high plastic deformation behavior, which is attributed to dislocation multiplication and gliding mechanism, until stress concentration at the interface due to the dislocation piling up, and the yield strength is consistent with the Hall-Patch equation in microscale. However, the interfacial bonding strength gradually decreases with decreasing layer thickness, and the dislocation can transmit across the layer interface to accommodate plastic deformation when the shear stress exceeds the interfacial bonding strength; the yield strength can be estimated by Hall-Patch equation. Therefore, the transition from ductile fracture to brittle fracture may be attributed to the layer thickness size, interfacial constraining effect and columnar grain orientation.

According to Hsia et al. [67] theoretical model as shown in **Figure 17**, the plastic deformation zone size (r_p) at the onset of crack tip is expressed as follows:

$$r_p = \frac{1}{2\pi} \left(\frac{K_{Ic}}{\sigma_s} \right)^2 \quad (8)$$

where K_{Ic} is the fracture toughness and σ_s is the yield strength of the metal. Therefore, there is no confinement exists, and the fracture energy is independent of layer thickness when the layer thickness (h) is higher than $2r_p$. However, the plastic deformation behavior becomes confined when $h \leq 2r_p$, resulting in a lower fracture energy. Meanwhile, the tensile stress at the crack tip increases as h decreases. When h approaches the micro level, the dislocation activities within the thin ductile layer can no longer be treated with continuum plasticity theory, and the confined ductile layer with thin thickness makes dislocations pile up at interface, and even the preexisting dislocations with low number are unlikely to blunt the tunnel crack tip, which results in the transition from ductile to brittle behavior

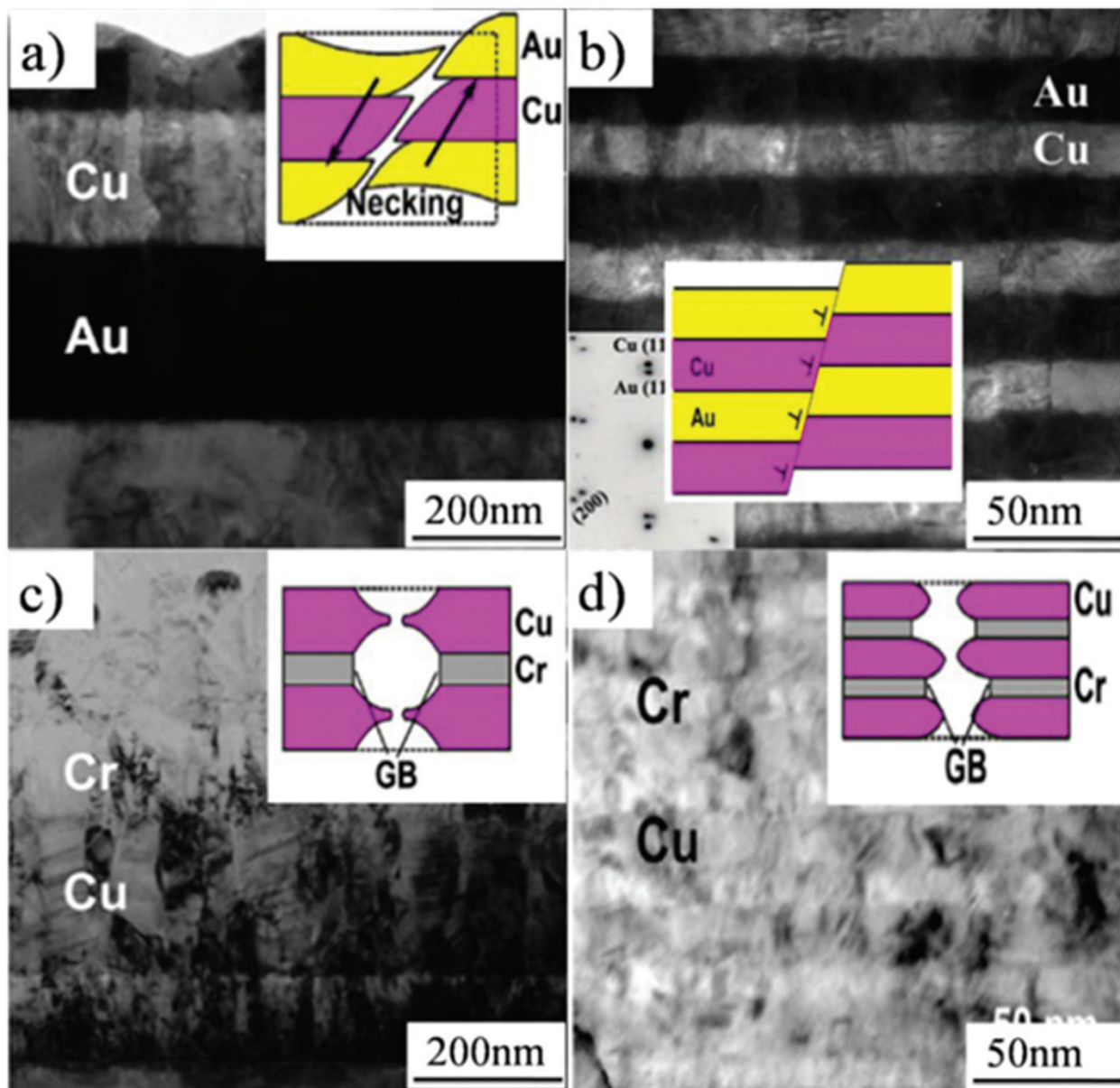


Figure 16. Size effect of laminated composites with layer thickness of (a, b) 200nm and (c, d) 50nm [55].

with decreasing ductile layer thickness by introducing the laminated structure, the confined layer thickness makes dislocations pile up at interface. A low equilibrium number of dislocations can be obtained in the thin ductile layer thickness, which leads to the super-high tensile stress and low fracture toughness at the crack tip, and even cleavage occurs when the tensile stress reaches the theoretical fracture strength. An equilibrium number of dislocations exists at a given load level and ductile layer thickness. Dislocations emitted from the crack tip can not only blunt the crack, but also sent a back stress to the crack tip. Above two effects are mutually competing each other; the former reduces the tensile stress at the crack tip, whereas the latter prevents more dislocation emissions and results into the increase of tensile stress.

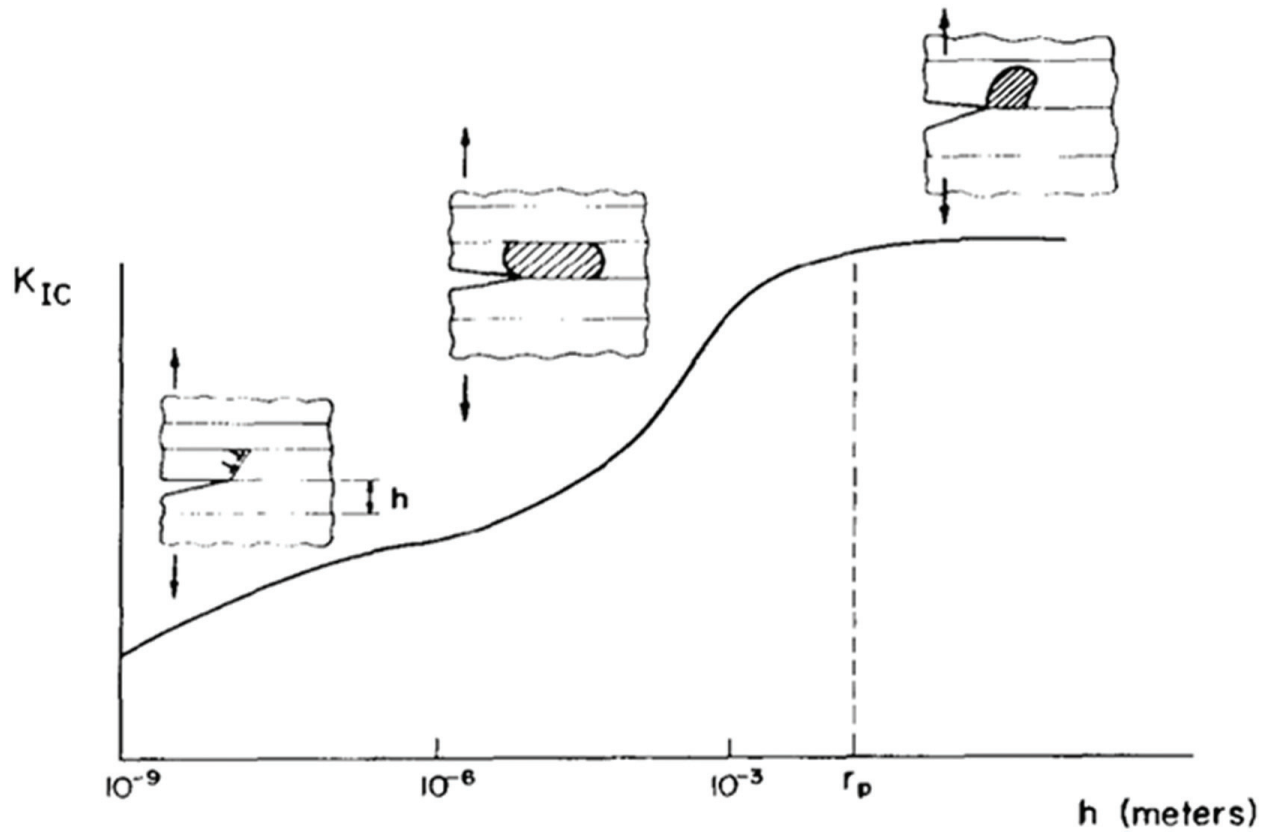


Figure 17. Schematic illustration of fracture energy as a function of layer thickness [67].

4.4. Interface layer

4.4.1. Interfacial bonding strength

Nambu et al. [38] reported that laminated composites with strong interfacial bonding strength exhibit superior longitudinal fracture elongation compared with the laminated composites with weak interfaces. For a metal film deposited on an elastic substrate, Li et al. [68] predicted that strong interface can delay strain localization by finite element simulation (FEM). However, if the metal film approaches to form interfacial delamination crack due to weak interfacial bonding strength, the metal film becomes freestanding and is easy to form a localized necking. Koseki et al. [40] proved that delamination crack is related to the low interfacial toughness, while laminated composites with high interfacial toughness reveal high fracture elongation before failure, leading to localized necking and ductile fracture surface, exhibiting interfacial delamination crack and brittle fracture of brittle layer when the interface becomes unstable with the increase of strain. The debonding criterion is expressed as follows:

$$\Gamma_{\text{int}} \geq 0.26 \frac{\pi t_B \sigma^2}{2E_B} \quad (9)$$

where Γ_{int} is the interfacial toughness and σ is the stress imposed on the brittle layer. This criterion provides a reasonable conditions of sufficient interfacial toughness to inhibit the interfacial delamination phenomenon.

4.4.2. Interfacial reaction phase

The interfacial reaction between two different layers can result in decreasing mechanical properties of laminated composites [50]. There are a series of studies on the mechanical response of laminated composites based on Ti-Al [69], Ti-Cu [70], Ti-Ni [71], Ni-Al [72], Cu-Al [73], Ti-Nb [74], Fe-Al [75, 76] and Fe-Cu [77] systems. **Table 2** shows the room temperature tensile properties of these laminated composites. The laminated composites reveal low fracture elongation compared to the constituent metals, which is attributed to the sintering voids and in situ reacted brittle intermetallic layer.

The size of reacted intermetallic layer can also affect the mechanical properties of laminated composites. Pang et al. [78] and Huang et al. [79] reveal that laminated Ti-(SiCp/Al) and Ti/Al composites exhibit high fracture ductility due to the in situ reacted nanosized TiAl₃ intermetallic layer, whose fracture elongation is even higher than that of the constitute layers. Guo et al. [74] reported that the intermetallic reacted layer can influent the fracture mechanism of adjacent layer. The plastic deformation behavior of adjacent Al layer can be constrained by the hard brittle intermetallic reacted Al₂Cu layer, leading to brittle fracture morphologies with crack propagating along the grain boundary of Al. Liu et al. [54] proposed that addition of thinner Al layer can enhance the tensile plasticity of cold-rolled Cu without losing strength by delaying onset of premature localized necking. Overall, the constrained interface can effectively improve the strain delocalization of soft layers and delay the formation of tunnel cracks [79].

4.5. Strain hardening exponent (n) and strain rate parameter (m)

For the strong interface without transition phase, the deformation behavior of laminated composites can depend on the strain hardening exponent (n) and strain rate parameter (m) of the constitute layers. Based on the power-law Hollomon equation, the flow stress (σ) of the individual layer can be expressed as follows [50]:

$$\sigma = K \left[\varepsilon^n + m \ln \left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_r} \right) \right] \quad (10)$$

where K is the strength constant and $\dot{\varepsilon}_r$ is a reference strain-rate. Herein, n and m exert the most dramatic influence, high value of n indicates the high uniform fracture elongation, and

Laminated systems	Brittle intermetallic layer	Maximum elongation at room temperature (%)
Ti-TiBw/Ti (present work)	—	27.5
Ti-Al [69]	Al ₃ Ti, Al ₂ Ti, TiAl, TiAl ₃	3.4
Ti-Cu [70]	Ti ₂ Cu, TiCu, TiCu ₃ , TiCu ₄	4.5
Ti-Ni [71]	TiNi ₃ , Ti ₂ Ni, TiNi	≤3
Ti-Nb [72]	NbTi	3.6
Ni-Al [73]	Ni ₃ Al, Al ₂ Al ₃ , NiAl	12
Cu-Al [74]	Al ₂ Cu, AlCu, Al ₃ Cu ₄ , Al ₂ Cu ₃ , Al ₄ Cu ₉	11
Fe-Al [75, 76]	Fe ₂ Al ₅ , Fe ₄ Al ₁₃	18
Fe-Cu [77]	—	60

Table 2. The elongation of laminated composites at room temperature in reported different system.

high value of m will even the material superplastic. The strain-rate hardening increases the elongation, which not only delays the onset of instability, but also by retarding the consequences of necking. The laminated composites with superior tensile ductility can be obtained by adjusting the values of n and m of constitute layer [80].

Figure 18 shows the fracture elongation map of many power-law hardening monolithic materials [80, 81]. It is interesting to note that a low ductility material laminated with another material that can coax the laminated composites toward higher elongation. For example, austenitic stainless steel and commercial purity iron make them attractive options as the ductility-enhancing component within a laminated metal composites, and this rule can also be indicated in the stainless steel/ultrahigh carbon steel multilayered materials during the high-temperature tensile testing process [32].

4.6. Tunnel crack

In 2008, Inoue et al. [37] reported that fracture elongation is gradually increased with the decrease of layer thickness, which is attributed to decreasing size of tunnel crack. For the

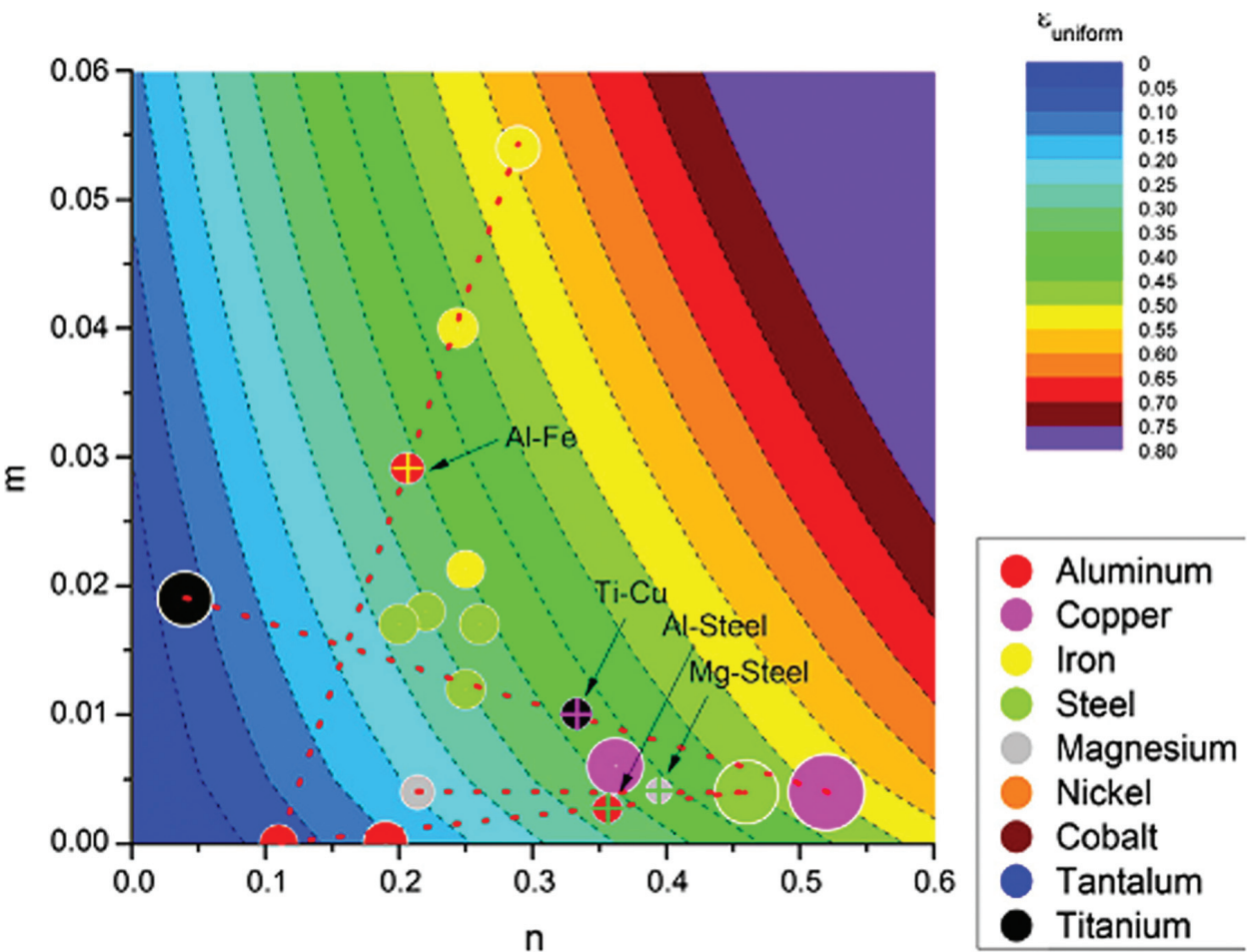


Figure 18. The fracture elongation map of many power-law hardening monolithic materials [80].

elastic-plastic laminated composites, the following criterion has already been proposed for suppressing crack propagation [40]:

$$t_B \leq t_{critical} = \frac{2\sqrt{3}K_{IC}^2\sigma_{Y,D}}{\sigma^3} \quad (11)$$

where t_B , K_{IC} and σ are the thickness, fracture toughness and the ultimate strength of the brittle phase layer, respectively. $\sigma_{Y,D}$ is the yielding strength of ductile phase layer, and $t_{critical}$ is the critical size. The above equation indicated that if the thickness brittle phase layer is lower than the critical thickness size, the tunnel crack in brittle phase layer can be effectively suppressed by the ductile phase layer. Therefore, a notable transition in the fracture morphologies of S420 brittle layer from brittle cleavage to ductile shear and dimples is identified as the layer thickness is reduced.

Meanwhile, Liu et al. [65, 82] reported that the laminated Ti-TiBw/Ti composites with thinner layer thickness possess higher fracture elongation than the laminated composites with thick layer thickness due to presence of more tunnel cracks. **Figure 19** shows the in situ tensile fracture process of laminated Ti-TiBw/Ti composites with layer thickness of 500 and 300 μm . It is revealed that the laminated composites with 500 μm layer thickness sharply fail at the tensile strain of 12.7%, and only a tunnel crack propagates into the overall sample as shown in **Figure 19(f)**. That is to say, when the brittle layer thickness extends beyond the critical thickness size, the ductile layer cannot prevent/blunt the propagation of tunnel crack, and the laminated composites will sharply fracture. However, the laminated composites with layer thickness of 300 μm revealed that one tunnel crack is presented at the strain of 13.4% as shown in **Figure 19(k)**. Finally, the laminated composites fail with two or more tunnel cracks at the strain of 15.4% as shown in **Figure 19(l)**, which is taken as a noncatastrophic fracture stage.

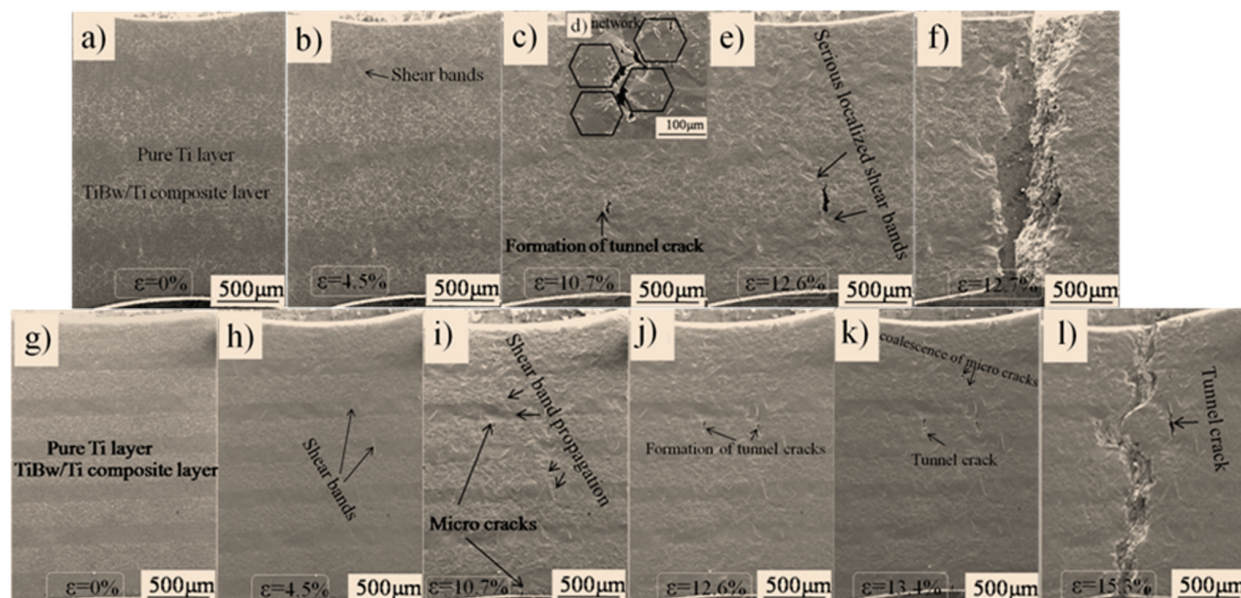


Figure 19. The fracture processes of the laminated Ti-TiBw/Ti composites with different layer thicknesses at the in situ tensile strains of (a, f) 0%; (b, h) 4.5%; (c, d, i) 10.7%; (e, j) 12.6%; (f) 12.7%; (k) 13.4%; (l) 15.3%. (a–f) For the laminated composites with 500 μm layer thickness; (g–l) for the laminated composites with 300 μm layer thickness [65].

Therefore, the laminated composites with 300 μm layer thickness can obtain higher fracture elongation than the composites with 500 μm layer thickness, which is attributed to more tunnel cracks formed in the thin TiBw/Ti layers.

In the laminated Ti-TiBw/Ti systems, Liu et al. reported [47, 83–87] that the formation and number of tunnel crack are also related to interfacial bonding strength and residual stress, and the ductile Ti layer exhibits ductile-brittle transition at the tunnel crack with the decreasing Ti layer thickness and the increasing volume fraction of TiB reinforcement, which is attributed to that the plastic deformation of thin Ti layer is constrained by TiBw/Ti layer, resulting in high triaxial tensile stress at the tunnel crack. Therefore, the laminated composites with thin Ti layer thickness reveal brittle fracture characteristics. With increasing volume fraction from 5 to 12%, the fracture manner in the TiBw/Ti layer changes from quasi-cleavage fracture to flat brittle fracture. Moreover, Ti layer reveals obviously ductile-brittle transition manner: the ductile dimples, crack bifurcation and lath-like intergranular fracture are presented in the laminated composites with volume fraction of 5%, 8% and 10%, respectively, and a typical brittle fracture manner is existed in the Ti layer. Huang et al. [85] reported that the plastic deformation zone can effectively delay the propagating behavior of tunnel crack. However, the size of plastic zone is decreased with the increasing volume fraction of TiB. Therefore, the tunnel crack is difficult to be blunted and easy to propagate in a brittle manner in the laminated composites with high volume fraction of reinforcements.

5. Conclusions

A background to the development of toughening mechanisms of metal matrix composites with multiscale hierarchical structure and laminated structure was overviewed in this article as follows:

1. The development of metal matrix composites approaches to the microstructure configuration design, and titanium matrix composites can achieve both high strength and toughness by adjusting the multiscale hierarchical structure.
2. The design ideal of metal matrix composites with laminated structure is aiming to achieve high strength, toughness and ductility. It is revealed that elastic property, and yield strength of laminated composites follow the “rule of average.” However, the estimation of fracture elongation and fracture toughness is complex, which is inconsistent with the “rule of average.”
3. The fracture elongation and toughness of laminated composites are related to interfacial bonding strength and tunnel crack number, and the fracture elongation of laminated composites with strong interface can be controlled by adjusting the strain hardening exponent n and strain rate parameter m .
4. The tensile properties and fracture characteristics of laminated composites reveal obvious size effect, with the decrease of layer thickness; the tensile strength is gradually improved, whereas the fracture elongation is first increased in microscale and then sharply decreased in the submicroscale and nanoscale.

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Author details

Baoxi Liu¹, Lujun Huang^{2*}, Lin Geng² and Fuxing Yin¹

*Address all correspondence to: huanglujun@hit.edu.cn

1 Tianjin Key Laboratory of Materials Laminating Fabrication and Interfacial Controlling Technology, School of Materials Science and Engineering, Research Institute for Energy Equipment Materials, Hebei University of Technology, Tianjin, China

2 School of Materials Science and Engineering, Harbin Institute of Technology, Harbin, China

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