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Co-Deformation in Severely Deformed Multiphase Crystalline Metallic Systems: Physical Mechanisms, Intrinsic Limitations, and Perspectives

Xavier Sauvage

The deformation of multiphase metallic alloys or metal matrix composites by forming processes such as rolling or drawing requires the co-deformation of phases. Severe plastic deformation processes have extended possibilities with the ability to produce unique nanoscaled structures through the formation of strain-induced super-saturated solid solutions, the fragmentation of second-phase particles, or ultimate co-deformation down to the nanoscale. Based on various examples, the fundamental mechanisms of co-deformation are addressed. Parameters that promote the extension of existing heterophase boundaries are discussed, and some possible atomic-scale mechanisms involving dislocation activities based on geometrical and energetic considerations are proposed. Finally, the physical limits of co-deformation are considered before suggesting research directions that might be followed to develop fundamental knowledge and applications in the field.

1. Introduction

Cold-drawn pearlitic steels, with a typical yield stress above 3 GPa, are among the strongest metallic materials produced at the industrial scale. They are widely used for suspension cables or as tire cords. Their exceptional mechanical strength results from the nanoscaled structure obtained by deformation during the drawing process. The original pearlitic lamellar structure (ferrite and cementite) progressively aligns along the wire axis, while the interlamellar spacing is progressively reduced from typically 100 nm down to 20 nm.^[1,2] Such a microstructure evolution requires a huge plastic deformation of the two original phases that co-deform leading to a progressive extension of interfaces. A similar strategy is applied to produce, for example, high-

strength Cu/Nb composite wires that combine excellent electrical conductivity and high mechanical strength.^[3]

Severe plastic deformation (SPD) processes that have been thoroughly investigated during the past 25 years^[4] have been applied to many multiphase metallic systems to create unique nanostructured materials. The team of Prof. Reinhard Pippan has carried out a large number of systematic studies on multiphase alloys (such as pearlitic steels) but also on so-called immiscible bi-metallic systems. They have proposed various strategies to promote the reduction of the length scales^[5] and eventually to ultimately achieve super-saturated solid solutions.^[6]


Figure 1 shows the typical evolution of the microstructure of a Cu/Cr composite

material processed by high-pressure torsion (HPT)^[4] to a large level of deformation. Although both phases exhibit significant hardness difference, they co-deform and the length scale of chemical heterogeneities is progressively reduced.^[7] The process is however not smooth, unlike in pearlitic steels processed by drawing^[1,2] or HPT,^[8] where the hard cementite co-deforms rather smoothly with the soft ferrite, the co-deformation process is rather chaotic with obviously much more plastic strain sustained by the soft copper phase. **Figure 2** shows another interesting example where the authors have achieved a Cu/Nb nanoscaled laminate by accumulative roll bonding (ARB)^[4] with a relatively large level of deformation. Although not necessarily regular across the laminate section, the Cu and Nb layer thickness was reduced progressively down to the nanometer scale without significant instabilities.^[9] Even if ARB applied to metallic couples may also easily lead to localized plastic instabilities,^[10] this work clearly demonstrates that the exceptional ability of ferrite and cementite to co-deform in the pearlitic structure is not an exception.

Co-deformation might be desired or not. In the case of drawn pearlite or Cu/Nb composite wires, continuous interfaces and interlamellar spacing reduction are the key factors to enhance the mechanical strength. Therefore, conditions that promote an optimal co-deformation between the two phases are wanted. In the case of nanoscaled materials achieved by SPD, second-phase particle fragmentation could be beneficial to pin grain boundaries (GBs)^[11] or to achieve super-saturated solutions,^[6] and then conditions promoting co-deformation that would eventually lead to the elongation of the different phases along the

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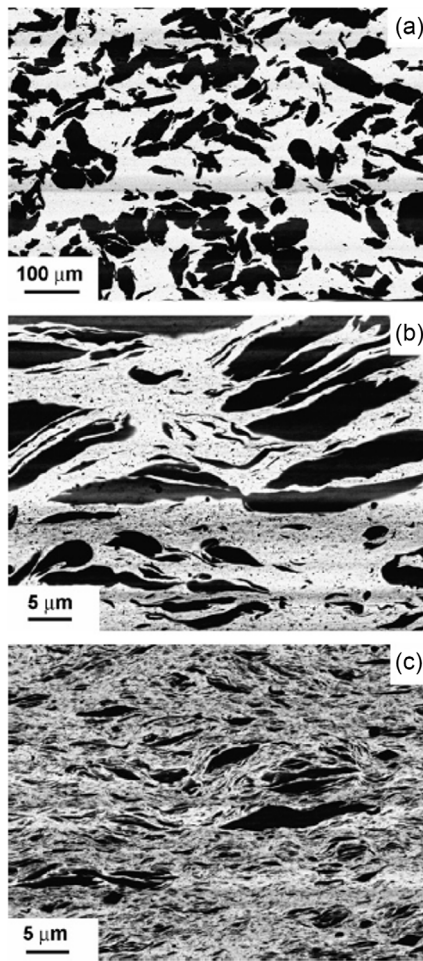


Figure 1. Scanning electron microscopy (SEM) images of the microstructure of a CuCr composite processed by HPT (cross-sectional views with the shear direction horizontal, Cr is darkly imaged): a) initial material; b) shear strain of 50; c) shear strain of 150. Reproduced with permission.^[7] Copyright 2008, Elsevier.

shear direction should be avoided. Therefore, understanding these phenomena is of critical importance, and the following fundamental questions should be addressed: 1) What are the physical parameters that promote (or not) the co-deformation? 2) What are the strain-induced atomic-scale mechanisms leading to an increase in the interface areas between two phases? and 3) How the interface integrity is maintained (or destroyed) so that mechanical alloying is avoided (or achieved)?

2. Parameters Influencing Co-Deformation

In crystalline metallic alloys, plastic deformation is often carried out by mobile dislocations. These defects are mobile when the local shear stress is higher than a critical value. In polycrystalline materials, it is characterized by a yield stress leading to measurable plasticity. Then, it is relatively natural to imagine that materials with similar yield stresses will easily co-deform. However, obviously the pearlitic steels and Cu/Nb composite materials that are mixture of two phases with very different yield

stresses prove that it is not necessary. It has been demonstrated indeed that internal stresses that develop in such systems during deformation lead to a charge transfer promoting the co-deformation and also significantly influence the final mechanical properties.^[3,12]

To achieve an ideal co-deformation that continues under large strains, homogeneous slip is necessary within the multiphase structure at two different scales. First, it is necessary at a scale larger than the size of the different phases to avoid shear localization at the macroscopic scale and fracture. A large hydrostatic pressure such as that involved in SPD processes will be of course favorable, but industrial processes like wire drawing or cold rolling may also provide the necessary conditions. In general, most processing parameters such as strain rate, temperature, deformation path, and amplitude will be important and should be optimized.

Second, homogeneous slip is also necessary within each phase, thus at a much smaller scale, especially when length scales become smaller and smaller after progressive accumulation of strain. It is naturally promoted when the mean free path of dislocations becomes similar to the length scale of structural heterogeneities (phase size) leading to plasticity mechanisms controlled by dislocations–interface interactions. Many dislocations accumulate at interfaces where they can eventually recombine, as it will be discussed later in this manuscript. The influence of the length scale of structural heterogeneities has been demonstrated in drawn pearlite, where a critical original interlamellar spacing does exist beyond which co-deformation of ferrite and cementite is not possible anymore.^[1] Similar observations have also been reported for Cu/Nb laminates processed by ARB.^[9,13]

If length scales are important, it cannot be ignored that during large level of deformation of crystalline materials, some crystallographic textures usually develop. In specific conditions, it could give rise to very specific microstructures like the so-called “Van-Gogh sky structures” in bcc/fcc composite materials processed by drawing.^[14] Systematic studies about the ability to co-deformation of systems with various combinations of crystallographic structure leading to different deformation textures have been extensively studied by Russel et al.^[15] This is probably a key factor leading to the very regular and homogeneous co-deformation reported in Cu/Nb laminates.^[16] As shown in **Figure 3**, high-resolution electron microscopy observations have revealed that the interface developed during deformation between the Cu and the Nb phases exhibits a very specific orientation relationship. This interface structure is deeply connected to the texture that was created in the two phases during rolling, and as it will be discussed in the next section, it probably promotes the atomic-scale mechanisms of the interface area extension.

Beyond length scale and texture, the morphology and distribution of phases is crucial. It has been demonstrated, for example, that in pearlitic steels processed by wire drawing^[17] or HPT,^[8] cementite lamellae that are not favorably oriented (perpendicular to the wire axis for drawing and perpendicular to the shear direction for HPT) are fragmented, leading to fine grain structure of ferrite stabilized by nanoscaled cementite fragments. Such a structure is also typical of multiphase alloys that contain intermetallic particles, such as Al-Fe alloys, where the Fe-rich brittle intermetallic phase does not co-deform with the Al matrix but is fragmented down to the nanometer scale.^[11]

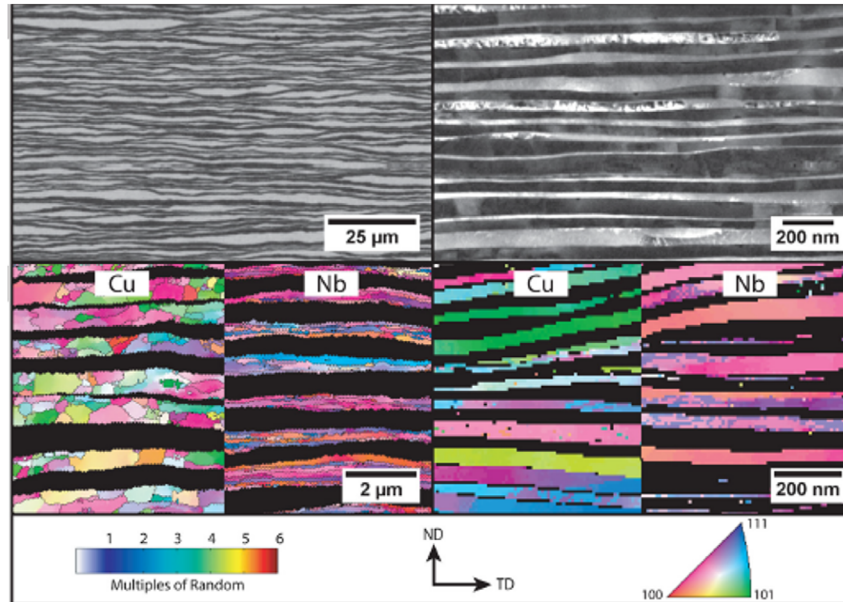


Figure 2. Comparison of coarse and fine Cu/Nb nanolaminate structures achieved by ARB showing a more homogeneous co-deformation of the fine structure (right column). Coarse structure (left): SEM image and electron back scattered diffraction inverse pole figure. Fine structure (right): high-angle annular dark field scanning transmission electron microscopy image and precession electron diffraction maps. Reproduced with permission.^[9] Copyright 2016, AIP Publishing.

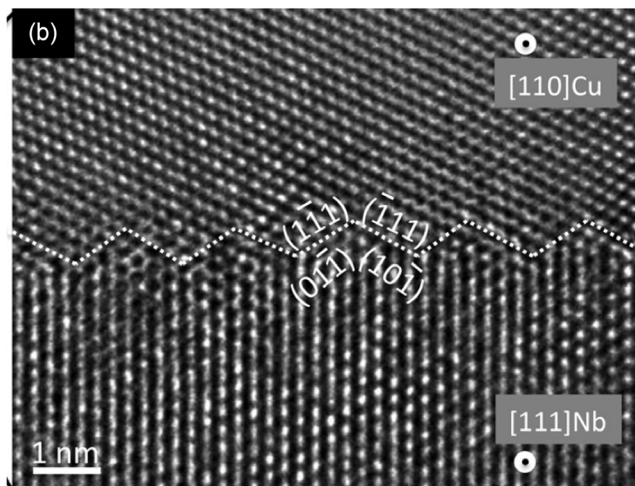


Figure 3. High-resolution transmission electron microscopy image showing the atomic-scale configuration of Cu/Nb interfaces with a specific orientation relationship in Cu/Nb nanolaminates obtained by ARB. Reproduced with permission.^[16] Copyright 2013, Springer.

3. Extending Interfaces at the Atomic Scale via Dislocation Storage

Co-deformation is characterized by the homogeneous, regular, and concomitant plastic deformation of two adjacent phases under stress. Thus, it requires plastic events in both phases and some mechanisms at the interface between the two phases to extend the interface area. In crystalline metallic alloys or

composites deformed at ambient temperature at a reasonable strain rate, plastic deformation is often carried out by dislocation activity. This deformation mechanism will be considered in the following. Under local shear stress, dislocations are mobile, propagating shear events until they get pinned by some obstacles. In multiphase materials, interphase boundaries are strong obstacles and different mechanisms could occur when a dislocation meets such an interface: 1) formation of dislocations pile-ups; 2) transmission of the dislocation in the other phase and shearing of the interface (may eventually occur if the interface between the two phases is coherent, and in such a case, fragmentation is promoted); and 3) recombination with other dislocations or defects at the interface. In metal matrix composites that combine a crystalline and an amorphous phase,^[18,19] dislocations emitted in the crystalline phase might be stored or absorbed at interfaces and eventually trigger shear bands in the amorphous phase leading also to an extension of interfaces.

Figure 4 shows the schematic representation of mechanisms involving dislocations that may contribute to the necessary increase in the interface area between two crystalline phases to achieve co-deformation. The first is based on the combination of two shear events occurring in a close location. They are induced by dislocations from two different sources and transmitted across the interface (Figure 4a–d). The combination of the two shear events leads to an increase in the interface length of $\sqrt{2}b$ where b is the burger's vector of mobile dislocations (assumed to be similar in both phases). In multiphase systems favorable to co-deformation (such as pearlite or Cu/Nb composites), there is little evidence of ledges at interfaces and the probability of two independent shear events at the same location is quite unlikely. Therefore, this mechanism, even if possible, is

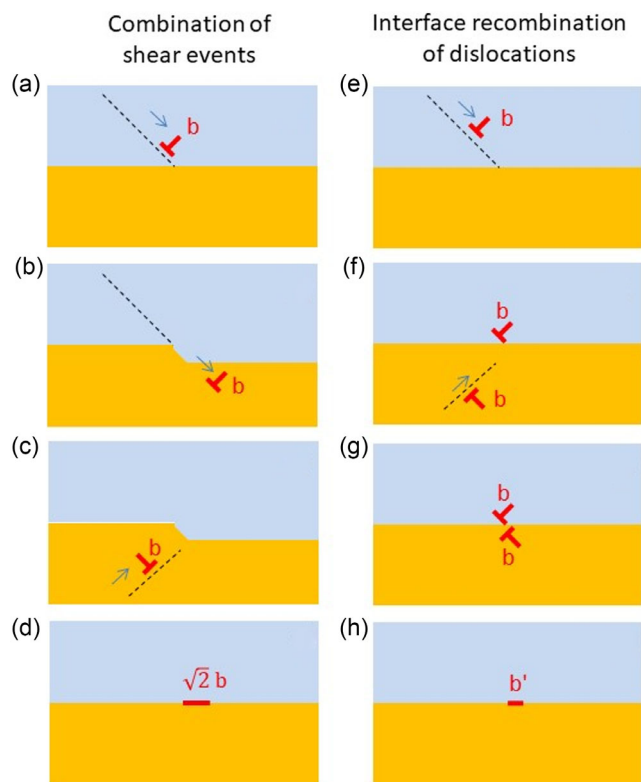


Figure 4. Schematic representation of possible atomic-scale mechanisms leading to the strain-induced extension of interfacial area between two phases that co-deform (for the sake of simplicity, dislocations with the same burger vector in both phases are considered). Left column, combination of shear events: a) activation of a slip system in the upper phase, b) transmission of the dislocation across the interface and creation of step at the interface, c) activation of a slip system in the lower phase, d) transmission of the dislocation across the interface and extension of the interface. Right column, interface recombination of dislocations: e) activation of a slip system in the upper phase, f) dislocation captured by the interface and activation of a slip system in the lower phase, g) both dislocations are captured at the interface, and h) recombination of the dislocation and extension of the interface.

probably not frequent. The second possibility (Figure 4e–h) is the absorption of lattice dislocations by the interface followed by the recombination with a dislocation coming from the adjacent phase. The recombination of the two interface dislocations leads to an increase in the interface length. When a favorable orientation relationship exists between the two phases or when a specific texture is developed under the macroscopic plastic deformation,^[16] such mechanism would be favored.

To evaluate how realistic might be such a recombination process, one may consider the energetic point of view. The elastic energy per unit length of a dislocation can be written as:

$$E_{\text{dislo}} = \frac{\mu}{4\pi} b^2 \ln\left(\frac{R}{r_0}\right) \quad (1)$$

where μ is the shear modulus, b is the burger vector, r_0 is the radius of the dislocation core (typically $r_0 \approx b$), and R is the distance on which the elastic field extends (typically half the mean

distance between dislocations). As a rough estimate, with $R > 100 b$, $\mu \approx 100$ GPa, $b \approx 2 \cdot 10^{-10}$ m, then $E_{\text{dislo}} > 10^{-9}$ J m⁻¹.

This should be compared to the energy required to expand the interface boundary along the same unit length with a width equal to b (scenario in Figure 4) which can be written as:

$$E_{\text{int}} = b\gamma \quad (2)$$

where γ is the interfacial energy. Then, with $\gamma \approx 1$ J m⁻², $E_{\text{int}} = 2 \times 10^{-10}$ J m⁻¹ which is significantly lower than E_{dislo} indicating that the recombination schematically represented in Figure 4 is indeed highly favorable on an energetic point of view.

Nevertheless, there are situations where the co-deformation occurs relatively smoothly (Cu/Nb laminates in Figure 2), but there are also situations where the harder phase does not sustain as much plastic strain as the softer phase, leading to some relatively complex structures. This is the typical case of the Cu/Cr composite displayed in Figure 1, and similar features have been recently also reported in the Cu/Mo system.^[20] This situation is schematically represented in Figure 5, where some particles are distributed within a matrix with a lower yield stress. Depending on the distribution of the particles and their volume fraction, some localization of plastic strain may eventually occur in the matrix (Figure 5a), while some dislocations may create pile-ups at matrix–particle interfaces (Figure 5b). These pile-ups promote the emission of dislocations inside the particle and activate its plastic deformation. At a given macroscopic plastic strain, the matrix has sustained much more plastic deformation, and the

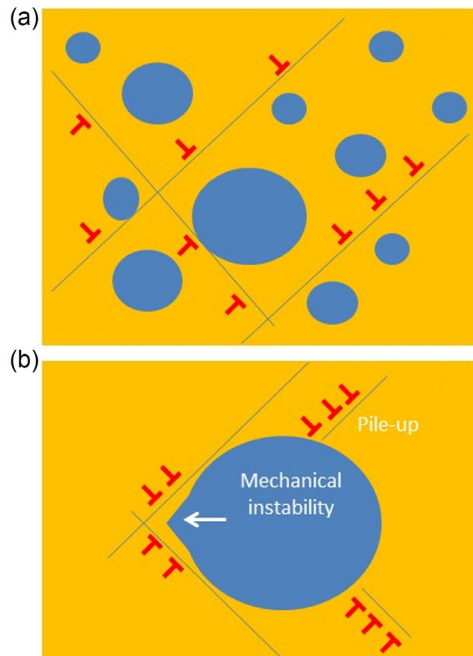


Figure 5. a) Schematic representation of shear localization in a material with a matrix containing particles with a higher yield stress. b) Interactions between matrix dislocations and one specific particle. Some dislocation pile-ups build up at the interface increasing locally the stress and triggering shear events in the particles (not depicted for clarity). Ahead of the particle, localized shear events in the matrix lead to some plastic instabilities and a localized deformation of the particle.

co-deformation is limited by the interface area increase that requires dislocation recombination and thus sufficient activity inside particles (Figure 1b). All particles do not exhibit the same elongation, and the morphology of particles indicates that the local co-deformation may vary within particles. This is probably the result of localized shear events intersecting near particles, which might create some instabilities (Figure 5b).

4. Intrinsic Limitations of Co-deformation and Perspectives for Future Research

As long as the co-deformation conditions discussed earlier do exist (sufficient hydrostatic pressure, enough plasticity in both crystalline phases, optimal orientation of phases relative to the shear stress, etc.), the different phases in the deformed material will flow, the total interfacial area will increase, and the length scale of the material heterogeneity will decrease. In principle, this could last as long as no significant damage occurs and as long as the surface integrity is kept. Indeed, if interfaces are sheared by transmitted dislocations, some fragmentation or some local mixing that would affect the interfacial region will possibly occur. During a process such as ARB (Figure 2), where the thickness of layers is reduced at each step, the obvious physical limit is when the layer thickness of one phase reaches a value close to the lattice parameter. A similar limit does exist also for wire drawing and for any other SPD process that increases interfacial areas between phases. However, this extreme situation seems to never be reached experimentally, where at the nanometer scale, the surface integrity is usually lost. Some fragmentation and mechanical mixing are often reported,^[6,11,20] even for systems that exhibit an excellent ability to co-deformation such as fine pearlitic steels^[21] or Cu-based nanocomposites.^[7,22] This might be attributed to some mechanical instabilities or changes in deformation mechanisms. Molecular dynamic simulations have been used to clarify atomic-scale mechanisms of mechanical mixing.^[23] Nevertheless, further work should probably be carried out for a better understanding of the exact mechanisms involved at interfaces (schematically described in Figure 4 and 5) and their limits at the nanometer scale where surface integrity is lost.

Single-phase polycrystalline materials exhibit some interfaces, namely GBs. Under large levels of deformation by traditional processes such as wire drawing or rolling, the morphology of grain does change; they become elongated, leading to an increase of the total GB area. Thus, the situation looks relatively similar to multiphase or composite materials where co-deformation processes lead also to a morphological change and an increase of interfacial areas. However, GBs exhibit a much higher mobility than heterophase boundaries and dynamic recrystallization phenomena (even at low homologous temperature) usually limit the evolution of the grain aspect ratio (unless some impurities or particles pin them),^[24] which is not the case in multiphase materials. Thus, controlling and taking advantage of co-deformation mechanisms is still a promising way to achieve unique ultrafine grain structures. For example, it has been shown recently how the combination of co-deformation and reactive interdiffusion during SPD could extend the possibilities for the design of high-strength metallic alloys with relatively high ductility.^[25] But more

generally, there is a wide field of possible applications (recently reviewed in ref. [26]) that could certainly take benefit of a better understanding of basic mechanisms of co-deformation for the optimization of structures down to the nanoscale. The design of new metal matrix composites, such as those recently achieved thanks to the ARB process,^[27] is an interesting and promising way of development. The elaboration of hybrid^[28] or architected^[29] materials, eventually based on inspiration from nature,^[30] based on co-deformation is also worth being studied more extensively, since such structures offer a much larger range of possibilities for structural and functional properties.

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Conflict of Interest

The author declares no conflict of interest.

Keywords

co-deformation, metallic alloys, multiphase systems, nanoscaled structures, severe plastic deformation

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