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Opinion

Amorphous Against Crystalline in High-Entropy Alloys

Swe-Kai Chen*

Professor, National Tsing Hua University, 101 Kuang Fu Rd. Sec. II, Hsinchu 30043, Taiwan

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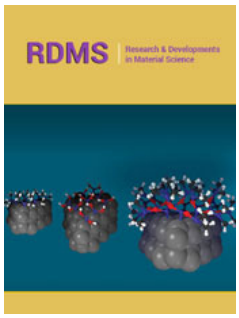

***Corresponding author:** Swe-Kai Chen, Professor, National Tsing Hua University, 101 Kuang Fu Rd. Sec. II, Hsinchu 30043, Taiwan

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Abstract

Amorphous (A) and High-Multicomponent (HMC) or High-Entropy (HE) alloys are from discussion of thermal kinetics and thermodynamics. The S-Z kinetic curves and an Entropy-Enthalpy (EE)-plane elucidate formation of amorphous and HMC or HE alloys, respectively. A Pseudo-Unitary Lattice (PUL) explains the most properties of HEAs. The role of number of components in both A and HMC alloys is also mentioned shortly.

Opinion

Amorphous Alloys (AAs) which have no conventional crystalline structures are strictly a kind of liquid-phase solid materials. Without grain boundaries, they own good corrosion resistance in most industrious applications. They are products of thermal kinetic processes instead of thermodynamical ones. Their structure remained amorphous after producing is controlled by the nose time in kinetic curves during cooling. The longer the nose time the larger the size of the products. Usually, the kinetic curves contain an S-Z shaped curve with the upper portion of S curve and the bottom portion of Z curve in its simplest form. The whole compound curves in the plane of temperature and time coordinates contain a series of producing % crystalline curves usually from the beginning detectable 1% to the finishing 99% stages. When a cooling process across the plane falls in the left-hand side of the 1% kinetic curve till the room temperature the product is amorphous, while that falls to the right-hand side of the 99% curve, the product is crystalline. There are empirical rules like Inoue's rule which tell the numbers of alloy components greater than or equal to three with small, medium, and large atomic sizes, there being much chance to manufacturing large useful amorphous alloys.

However, empirical rules for large sized amorphous alloys do not indicate the number of alloy components should be, but just emphasize the existence of three kinds (rather than number of components) of their atomic sizes have the close relationship to the lengthening of the kinetic nose time necessary to produce the amorphous. Critical cooling rate for the formation of amorphous phase is crucial in this aspect.

Although the large multicomponent alloys (MCAs, now also known as high-entropy alloys, HEAs) have been demonstrated as Crystalline Solid Solutions (CSSs) which give the desired toughness properties of alloy materials, the early developing thinking in MCAs was to design a series of AAs [1]. The aspects of consideration in designing and manufacturing of MCAs and AAs respectively are largely different from each other. MCAs are thermodynamic crystalline phases, while AAs are kinetic amorphous ones. A simple model, entropy energy-to-enthalpy diagram (EED) in Figure 1, for designing the production of CSSs in MCAs has been developed [2,3].

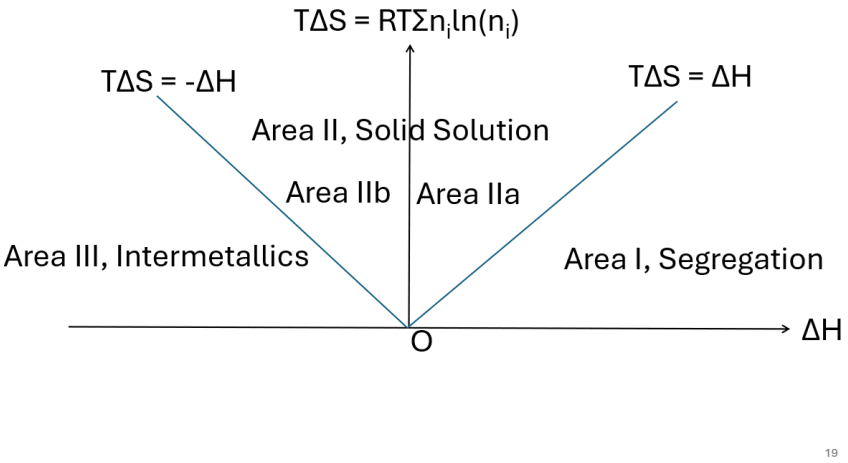

Figure 1: An entropy energy TΔS vs. ΔH diagram for elucidating the enlarged solubility of components in a multicomponent alloy.

EED contains a whole horizontal enthalpy (ΔH) and an upper half longitudinal entropy-energy (TΔS) coordinates in the upper half coordinate plane (Figure 1). The plane is derived from the thermodynamic Gibbs function (ΔG) that contains a chemical heat function of enthalpy and a minus physical entropy term, gives the famous equation of ΔG=ΔH-TΔS. The reason for choosing the upper half TΔS is due to the positive term itself by the second law of thermodynamics. At low temperatures T and low ΔS, this physical term is usually neglected as compared with the enthalpy term. One knows that reactions before and after with positive, zero, and negative values of ΔH respectively stand for reluctant, reversible, and spontaneous processes, meaning that one should add energy to a reaction or not in order to guarantee a process to be forward. In general, the condition for a reversible process is ΔG=ΔH-TΔS = 0. If constructs a TΔS-ΔH plane, then one has two areas in the first quadrant, one satisfies ΔH>TΔS>0, designated as Area 1, the other ΔH<TΔS>0, as Area IIa. Compared Areas I to IIa, a reluctant process means not spontaneous or naturally not mixing or segregate, while a spontaneous means mixing. The boundary of Areas I & IIa satisfies ΔH–TΔS = 0, i.e., ΔH=TΔS, which is a condition of reversible reaction. A survey over the Periodic Table of Chemical Elements tells one that the value of enthalpy for intermixing two elements is near zero for the ones belonging to the same column of elements as well as those in the rare-earth metals. This suggests that the ΔH values in the vicinity of zero value favor to form CSSs. As TΔS increases the ΔH width of CSS region for element mixing accordingly enlarges, and thus easily to form CSS, or in turn by saying that the solubility enlarges due to the increase of entropy. At the side of negative ΔH, the processes in this second quadrant are obviously all spontaneous. However, as the absolute value of ΔH is getting larger, an intermetallic reaction occurs since the chemical bond force of the new forming phase is becoming too strong to form CSSs. For simplicity, we just assume that the line boundary of -ΔH=TΔS divides intermetallic area III and CSS area IIb in this second quadrant for the sake of symmetry. To simply increase the number n in a multicomponent alloy is one of the easiest ways to increase the ΔS according to, e.g., R·ln(n) for an equimolar MCA or R·ln(n) = RΣ$n_i$ln(n_i) for the non-equimolar, where R is the gas constant, ln(n) is the nature log of n, and Σ(n_i/n)=1. There is an optimum for ln(n). Further, the other method to increase solubility is obviously to increase the temperature T.

Some words to the properties of crystalline phases in HEAs are given here. Experiments show the phases are commonly body- & face-centered cubic (BCC & FCC), and hexagonal close-packed (HCP) structures with smaller X-ray Diffractometry (XRD) peaks as compared to those of the conventional one- or less-principal-component alloys. A concept of Pseudo-Unitary Lattice (PUL), which consists of all component elements in a single lattice, has been proposed to explain the so-called four (three plus one) effects of sluggish diffusion, largely distortion, and cocktail effect in HEAs due to a multicomponent atom-accompanied diffusion, possible large sized difference of component atoms in the single lattice, and composed components in the characteristic single lattice, respectively, as well as the high solubility high-entropy effect just discussed above. Much mechanical, electrochemical, and physical properties as well as electrical, magnetic, superconductive, thin film, hydrogen absorption and desorption properties have been explosively explored [3,4]. By the way, from the results of Fermi energy detection, HEAs are metallic [4].

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