Classification of Displacive Transformations : What is a Martensitic Transformation ?

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Abstract. The displacive transformation classification proposed at ICOMAT 79 [1] is reviewed in light of recent progress in mechanistic understanding. Issues considered include distinctions between shuffle transformation vs. self-accommodating shear, dilatation vs. shear-dominant transformation, and nucleated vs. continuous transformation.

1. HISTORICAL INTRODUCTION

Cohen, Olson and Clapp (COC) at ICOMAT-79 defined a martensitic transformation as a sub-category of a wider field of displacive changes [1]. In the intervening 16 years, there has been appreciable progress in understanding martensitic and related transformations, and it now seems appropriate to re-examine the COC criteria in the light of recent experimental and theoretical work on microscopic mechanisms of transformation.

To some metallurgists, martensite will always be a particular microconstituent in suitably heat-treated ferrous alloys, whilst others may use the apparently tautological definition, "Martensite is the product of a martensitic reaction." The emphasis on the mechanism of transformation rather than the properties of the product phase began with an important paper by Troiano and Greninger [2], who showed that various kinetic and crystallographic characteristics of a martensitic change are markedly different from those observed in other "nucleation and growth" reactions. Solid state reactions were thus divided into two main groups, but Kurdjumov [3] and Kurdjumov and Maximova [4] soon showed that some of the Troiano-Greninger features (e.g., no isothermal transformation) are not applicable to all martensitic transformations. It was later proposed [5,6] that a change of shape of the product crystals should be an experimental test for a martensitic (or more generally a "displacive" [7] or "military" [8]) reaction.

Phase transformations which involve long range diffusion are regarded as reconstructive rather than displacive, but the early work of Garwood [9] showed, and more recent work [10-11] has confirmed, that shape changes occur in some plate-shaped precipitates with substitutional solute contents different from those of their parent solid solutions. These experimental results imply that some transformations requiring diffusion also have displacive character; they are sometimes called "nonferrous bainites," but this can be misleading. To emphasize their mixed characteristics, the term "diffusional-displacive transformations" has recently been suggested [12].

The COC classification scheme for separating martensitic from other displacive transformations is reproduced in Figure 1. Displacive transformations dominated by atomic shuffles rather than by lattice deformation are first eliminated, and only those lattice distortions in which shear strains are more important than dilatational strains are accepted as martensitic. Finally, a division is suggested between true martensites, in which the strain energy has a major influence on the transformation kinetics and product morphology, and quasimartensites with very small lattice deformations, in which transformation may be continuous. Whilst this scheme attempted to minimize the need for detailed mechanistic information, emphasizing instead kinematic and morphological information deducible from relatively macroscopic observation, it has since been suggested that nucleation is an essential characteristic of a martensitic reaction [13]. A concise definition of a martensitic transformation is then a "shear dominant, lattice distortive, diffusionless transformation occurring by nucleation and growth" [13]. Some points worthy of further consideration are (a) the definition of a shuffle and the validity of the distinction between shuffle-dominant and strain-dominant mechanisms, (b) self-canceling microshears which give
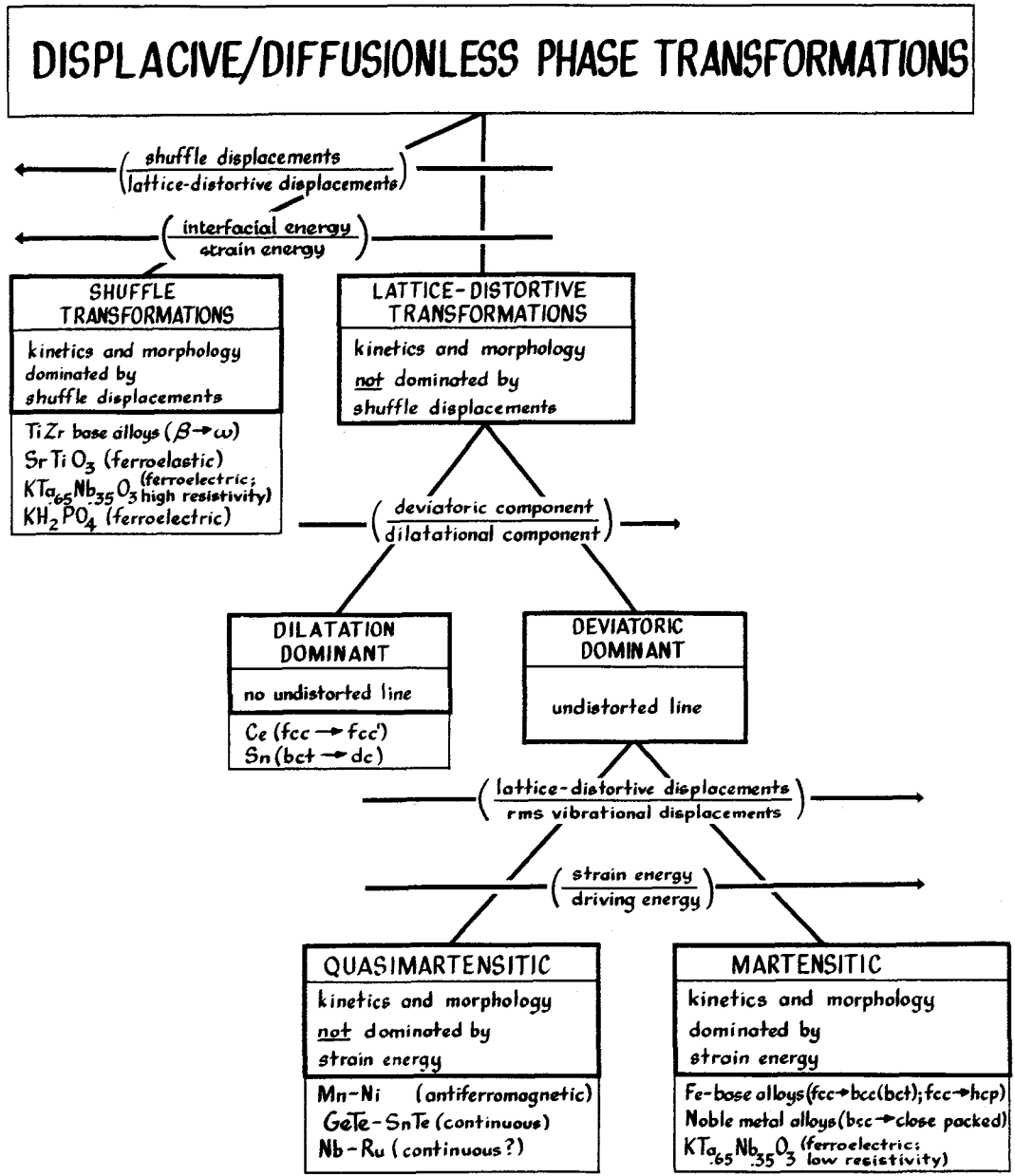
DISPLACIVE/DIFFUSIONLESS PHASE TRANSFORMATIONS

$$\left(\frac{\text{shuffle displacements}}{\text{lattice-distortive displacements}}\right)$$

$$\left(\frac{\text{interfacial energy}}{\text{strain energy}}\right)$$

SHUFFLE TRANSFORMATIONS

kinetics and morphology dominated by shuffle displacements

Ti Zr base alloys ($\beta \rightarrow \omega$)
Sr Ti O_3 (ferroelastic)
$KTa_{.65}Nb_{.35}O_3$ (ferroelectric; high resistivity)
KH_2PO_4 (ferroelectric)

LATTICE-DISTORTIVE TRANSFORMATIONS

kinetics and morphology <u>not</u> dominated by shuffle displacements

$$\left(\frac{\text{deviatoric component}}{\text{dilatational component}}\right)$$

DILATATION DOMINANT

no undistorted line

Ce (fcc $\rightarrow$ fcc')
Sn (bct $\rightarrow$ dc)

DEVIATORIC DOMINANT

undistorted line

$$\left(\frac{\text{lattice-distortive displacements}}{\text{rms vibrational displacements}}\right)$$

$$\left(\frac{\text{strain energy}}{\text{driving energy}}\right)$$

QUASIMARTENSITIC

kinetics and morphology <u>not</u> dominated by strain energy

Mn−Ni (antiferromagnetic)
GeTe−SnTe (continuous)
Nb−Ru (continuous?)

MARTENSITIC

kinetics and morphology dominated by strain energy

Fe-base alloys (fcc$\rightarrow$bcc(bct); fcc$\rightarrow$hcp)
Noble metal alloys (bcc$\rightarrow$close packed)
$KTa_{.65}Nb_{.35}O_3$ (ferroelectric; low resistivity)

Figure 1: Classification scheme proposed at ICOMAT 79 [1].

near zero shape change, (c) transformations with very small principal strains, and (d) diffusional-displacive transformations.

2. SHUFFLES AND SELF-COMPENSATING SHEARS

A lattice deformation produces changes in the sizes and/or the shapes of a set of unit cells (not necessarily primitive cells) of the parent lattice; a shuffle only rearranges the atom positions within a unit cell. In principle, a labelled set of atoms defining a primitive reference lattice (the crystal lattice or some superlattice thereof) in the parent structure will, after incorporation into the product, form a new primitive lattice, and the labels will identify corresponding vectors, planes and cells. Any such atom in an initial position $\mathbf{u}$ will have a final position $\mathbf{v}=\mathbf{Su}+\mathbf{t}$ where $\mathbf{S}$ represents the lattice deformation and $\mathbf{t}$ is a non-lattice-repeat vector. The remaining atoms of the parent structure are situated on identical, interpenetrating lattices, and to produce the correct product structure, each such lattice must be deformed as above and then translated relative to the reference lattice. These translations constitute the shuffles. If there are n atoms in corresponding unit cells of minimum volume, the minimum fraction of atoms required to shuffle is $1-1/n$ (apart from some special configurations). Shuffles can only be avoided if both structures have primitive unit cells containing only one atom. A corollary of this is that displacive changes involving structures with large unit cells will tend to be shuffle-dominated, and COC-defined martensitic transformations would then be favored by structures with only a few atoms per unit cell. Shuffles are *relative* displacements; it is equally valid to regard any other of the n sets of atoms as the non-shuffling reference set. Indeed if a reference lattice is chosen with an origin not on an atomic site, all atoms must shuffle.

An interesting test case for the relative roles of lattice deformation and shuffles is the δ (fcc) to α (monoclinic) transformation in Pu alloys in which the monoclinic α structure includes 16 atoms per unit cell. Despite the large amount of shuffling, a large lattice deformation apparently dominates the transformation behavior giving the same kinematic and morphological characteristics as classical martensites [14,15] with kinetic parameters very similar to martensitic transformations in ferrous alloys [16].

The separation of the atomic displacements into lattice deformations and shuffles is not always unambiguous; for example, the deformation may apply only to the lattice points with each atom in the motif unit given the same (lattice) displacement, or the atoms might be regarded as embedded in a deformable continuum, so that the various atoms in a motif unit will undergo different lattice displacements. Now consider the well-known mechanism for fcc↔hcp changes in which a dislocation (approximately a Shockley partial) moves through every two close-packed atomic planes of the original structure, converting it into the other structure. This is equivalent to the motion of an interface step, or "transformation dislocation," between planes (say) 1 and 3 in which the slip is entirely between planes 1 and 2. Thus, the whole structural change *apparently* requires no shuffling. But now consider the lattice deformation to be homogeneous, i.e. produced by the motion of a half-Shockley dislocation through every atomic plane. Every other atomic plane is now correctly placed but the other planes are stacked in intermediate positions, e.g., $\frac{1}{2}$ (B+C), and to complete the change, these atoms must shuffle into either B or C positions. Although the "no-shuffle" description seems preferable, the distinction is not physically meaningful.

A pure shuffle transformation requires that some unit cell of one lattice is almost identical with a cell of the other lattice. Apart from small differences in the spacing of the close-packed layers, this condition is met in various transformations between structures which have different stacking sequences of two-dimensional hexagonal atomic nets. In Co-Ni alloys, for example, a cell six atomic {111} planes high in the fcc structure is almost identical with a similar cell in the hcp phase, so that a shuffle-dominated transformation may be feasible. If A layers on planes, 0, 6, 12 etc. define the cell, and the hcp stacking is ...ABAB..., the required shuffles on planes 2, 3, 4 and 5 are C→A, A→B, B→A, and C→B respectively. The shuffle vectors may thus be of type $\pm\frac{1}{6}<111>$ or $\pm\frac{1}{6}<11\bar{2}>$. However, these shuffles require atoms to pass each other at separations much smaller than the interatomic distance, and this implies a very large increase in energy. The necessity to avoid configurations with unrealistic interatomic distances is probably the most important factor in prohibiting certain shuffles whilst permitting others, and the same restriction prevents certain shears. For example, shears of $2^{1/2}$ or $2^{-1/2}$ in the antitwinning direction of a fcc structure would produce, in principle, a fcc twin and a hcp structure respectively, but both involve A-A stacking at an intermediate configuration and are not observed. Shears of $2^{-1/2}$ and $8^{-1/2}$ in the opposite (twinning) direction give the same structures without intermediate A-A stacking and both are observed. In assessing the feasibility of shuffle mechanisms, the primary consideration may thus be neither the fraction of atoms which shuffle nor the magnitude of the shuffle vector, but rather the maximum disruption caused by the displacement of a single atom or of several atoms cooperatively.

The dislocation mechanism for the fcc-hcp change leads to a shape change with a large shear component of $8^{-1/2}$ as is observed in some alloys. However, in the absence of stress fields, any of the three <112> directions in a {111} plane may be the shear direction, and if the direction changes every z planes, the shape change will be nearly zero over distances greater than about $3z$ planes. In particular, if $z\sim1$, the zero shape change will apply to the unit cell discussed above in terms of shuffles. If the shears of the three layers occur consecutively, the mechanism is still intrinsically martensitic. But is it shear dominated? When z is appreciably larger than unity, the periodical changes in shear direction are undoubtedly driven by the accumulating strain energy, but for $z=1$ it is possible that 3-layer pure steps form spontaneously and effect transformation with zero shape change, thus producing no strain energy.

Hitzenberger and Karnthaler [17] have shown that the fcc phase forms from hcp in thin foils of Co-Ni alloys as lamellae with planar (not tapering) growth fronts and a minimum thickness of 6-10 planes, consistent with the pure step mechanism. Thicker lamellae (up to 50 planes) showed contrast effects indicative of a near zero net Burgers vector. Similar effects have recently been studied by Waitz and Karnthaler [18] in a dhcp-fcc transition in Co-Fe alloys. Two quasi-Shockley partials, gliding on adjacent atom planes, will produce an extrinsic fault, and repetition of this double shear every four planes will effect the change from ...ABC... to ...ABAC... stacking, and vice versa. The adjacent shears may be in the same direction, $\ell_1 = \frac{1}{6}<11\bar{2}> = \frac{1}{3}<10\bar{1}0>$, or in equivalent directions, ℓ_1 and ℓ_2; in the latter case, the net shear is parallel to $-\ell_3 = \ell_1 + \ell_2$ since $\Sigma\ell_i = 0$. Note that $-\ell_3$ is the antitwinning vector dismissed as impossible above, and its core is now spread over two atomic planes, and produces quite a different configuration. *In situ* electron microscopy has shown that the fcc phase also forms from dhcp as thin lamellae, usually 12 or 24 planes thick, with little evidence of large strains near their planar ends. The mechanism allows the shear to be compensated over a minimum of 12 planes, and is thus consistent with observations.

These observed pure shuffle mechanisms may be promoted by the thin foil geometry of the specimens through free-surface partial-dislocation source action. The observations raise the interesting question of the extent to which such transformations would still exhibit traditional martensitic characteristics. The absence of self energy would certainly provide a significant reduction of the nucleation barrier in the homogeneous limit, but perhaps more importantly the absence of a shape strain eliminates the strong elastic interactions which promote autocatalytic heterogeneous nucleation of many martensites.

3. DILATATION VS. SHEAR

The COC condition for a martensitic transformation is that the lattice deformation must have an undistorted line; this excludes only rather exceptional cases in which all principal strains have the same sign. The simplest such change is a pure dilatation, as in the transition between two fcc allotropes of Ce (γ and α) with very different atomic volumes. An invariant line strain is specified for martensite because it may be factored into a simple shear and an IPS, thus fulfilling the requirements of the crystallographic theories in which the habit plane is an IPS of the shape deformation, and is glissile, i.e., the number of atoms is conserved as the interface is displaced. In general, any large misfit between two phases can be tolerated elastically only in a particle of small dimension in the direction of the misfit. Thus, large uniform volume changes will only occur coherently in very small particles such as GP zones; larger particles will have to incorporate misfit dislocations in their interfaces so that the growth becomes non-conservative and requires an atomic flux. The best known example of a very large volume change (>20%) is the allotropic transformation in tin, where the low temperature (grey tin) form is very difficult to nucleate, and is obtained from bulk samples as a powder or as a severely cracked and crumbling aggregate. The pure dilatational γ-α transition in Ce alloys is apparently accomplished displacively without cracking, at least in thin foils [19] but the mechanism is not clear; in pure Ce it is preceded by an incomplete martensitic change from γ to a dhcp (β) allotrope. The previously mentioned δ-α transformation in Pu alloys also involves a 20% volume change, but in this case it has been clearly established [15] that a lattice correspondence is adopted involving sufficiently large shear deformations that the invariant line condition is met and the transformation exhibits conventional martensitic IPS characteristics.

The case of pure dilatational transformation offers simplifying features for theoretical investigation of some behaviors of general relevance to lattice-distortive transformations. This has been used by Chu et al [20] to perform precise numerical calculations of homogeneous "nonclassical" nucleation behavior near a lattice instability, comparing systems with smooth vs. cusped surfaces of energy vs. strain. Both types of system show decreasing strain amplitude of the critical nucleus as instability is approached, but only the smooth system shows the Cahn-Hilliard divergence of nucleus size and interfacial width. An interesting behavior shown by the dilatational transformation is that the localized deformation of a

nucleus is restrained by nonvanishing elastic shear constants such that the nucleation barrier does not necessarily vanish at the point of dilatational instability. This illustrates a significant role in nucleation of mechanical transformations of elastic constants beyond those directly associated with the primary order parameter of the system.

4. NUCLEATED VS. CONTINUOUS DISPLACIVE TRANSFORMATIONS

The COC classification distinguishes between martensitic and continuous displacive products ("quasimartensitic") in which nucleation is not required since variant domains appear virtually simultaneously throughout the material by continuous strain modulation [21]. This analogue of spinodal decomposition is theoretically possible for a first order transformation if the parent phase approaches a mechanical stability limit where an appropriate elastic modulus changes sign. However, even when the isothermal modulus is negative , rapid growth of a domain structure may be limited because the reaction is effectively adiabatic, i.e. energy is conserved and the growth rate of a perturbation is constrained by the necessary heat flow. Umantsev and Olson [22] have performed normal mode and weakly non-linear analyses of the dynamics in unstable systems where the homogeneous free energy φ is a smooth function of a strain parameter ζ and of temperature T. The partial second derivatives of this function, $\varphi_{\zeta\zeta}$, $\varphi_{\zeta T}$ and φ_{TT}, define the isothermal and adiabatic moduli, $\varphi_{\zeta\zeta}$ and $\varphi_{\zeta\zeta}(1-M)$ where $M=(\varphi_{\zeta T})^2/\varphi_{TT}\varphi_{\zeta\zeta}$. After incorporating a strain gradient energy, solutions may be obtained for the amplification factor β as a function of the wave number of the strain modulation, k.

Figure 2 is a stability diagram in the plane of $\varphi_{\zeta\zeta}$ vs. $\varphi_{\zeta T}$ which is divided into various unstable regions by the plotted values of M; in each region, the schematic β-k relationship is shown as an insert.
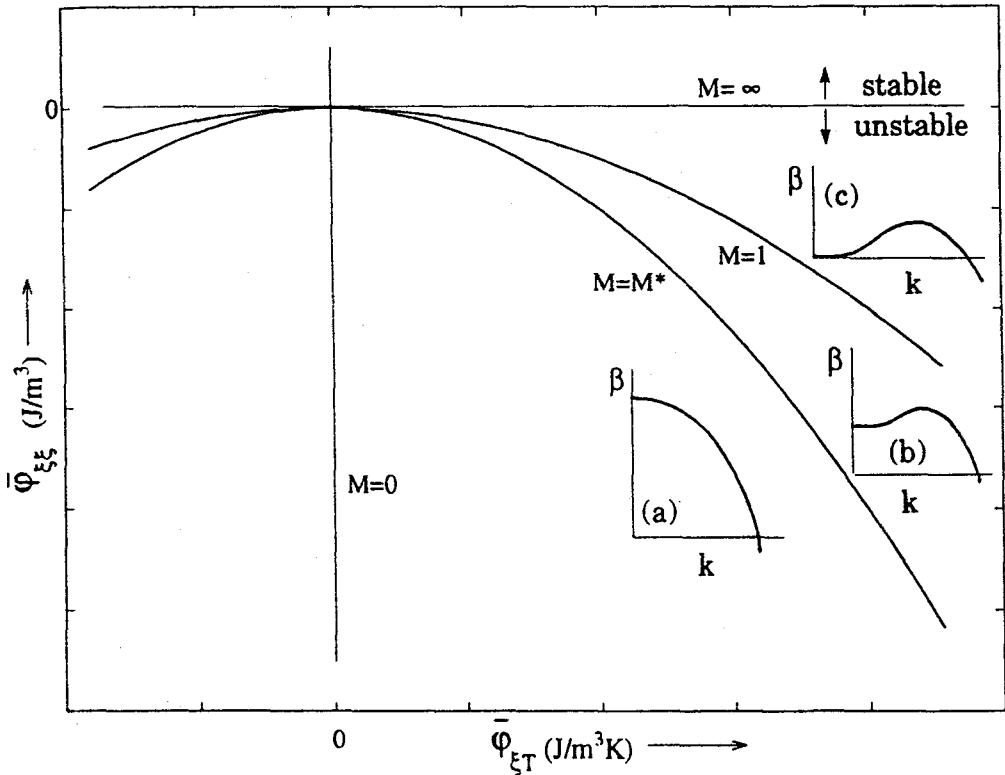

Figure 2: Stability diagram for continuous modulation in unstable systems, showing schematic curves of amplification factor β vs. wave number k [21].

In case (a), $0<M<M^*=(1+R)^{-1}$ where R is the ratio of thermal and mechanical relaxation diffusivities assuming linear response functions. Both moduli are strongly negative and the fastest growth is for a homogeneous distortion (k=0). Actually, studies of hydroelastic mechanical relaxation in such systems [23] suggests that inertial effects would probably lead to a dynamic strain conservation condition and give a $\beta(k)$ relation similar to that of case (c) below [24]. Case (b) with $M^*<M<1$ also corresponds to negative values of both moduli, but fastest growth occurs for a modulation of finite k. Finally in region (c) close to the stability limit, $M>1$ and the adiabatic modulus becomes positive. The homogeneous mode is now completely suppressed, and the fastest growing modulation is controlled entirely by heat transfer. A weakly non-linear analysis leads to a Cahn-Hilliard type equation, and indicates that the system is an exact thermal diffusion analogue of conventional spinodal decomposition. Later stages of continuous transformation have been studied by numerical methods [25].

The unstable states (a) and (b) of Figure 2 may not be experimentally accessible, but case (c) is of considerable interest. An attempt to impose this mechanism on the first order, cubic-tetragonal change in FePd alloys, for which estimates of gradient and elastic energies are available [26,27] was abandoned because of the very high predicted transformation rate; it has been suggested that insulating systems might provide a better experimental test [28].

An ultimate aim of these theoretical exercises is to understand whether there is a critical level of transformation strain below which a continuous reaction supersedes a heterogeneously (or in special circumstances [29] homogeneously) nucleated reaction, and whether this condition can be examined experimentally. Theoretical treatment of this competition will have to include the role of fluctuations in the metastable regime where the relevant elastic moduli are all positive. More elaborate calculations will be required to make a quantitative comparison between the rates of these two modes of transformation.

5. DIFFUSIONAL-DISPLACIVE TRANSFORMATIONS

This mixed category is clearly non-martensitic, but it has a crystallography which apparently follows that of a comparable martensitic transformation [11] and this must imply fully coherent or semicoherent interfaces. A kinetic theory has been developed for the special case where only interstitial atoms are mobile [30,31], as in ferrous bainites, but recent work demonstrates the possibility of a martensitic-type shape change in substitutional alloys at temperatures where all atoms are mobile. These alloys are often called "non-ferrous bainites," but in view of the probable mechanistic differences, this term may be inappropriate. As in martensite, such diffusional-displacive transformation implies a local lattice correspondence, but it is not clear why this correspondence should be maintained in cases where growth involves substitutional diffusion. It has been suggested that the term "atomic site correspondence" be used to emphasize that there is no correlation in the positions of individual atoms. However, in some cases, both diffusional and martensitic, only a fraction of the atom sites may be directly interrelated by a correspondence matrix, so that it is more logical to describe both situations as a lattice correspondence, with the atoms able to move freely over the sites in the diffusional-displacive case. The remaining question is then why do not these mobile atoms occupy additional sites which are situated, for example, at the surface of a single interface or in extended regions of an internally constrained particle, leaving unoccupied other surface sites or sites in severely compressed regions.

6. CONCLUSIONS

Review of the COC classification in light of recent progress supports the slightly amended definition of a martensitic transformation as a shear-dominant, lattice-distortive, diffusionless transformation occurring by nucleation and growth. Further progress in mechanistic understanding will help to quantify the gray areas separating different subclasses of displacive transformation. For planar stacking transformations, a distinction between shuffle and self-compensating shear transformations can be made by requiring the shuffle transformation to involve cancellation of shears over the smallest possible cell size, although such cases will still possess many features in common with shear transformation. Recent examples suggest the undistorted line criterion (lattice deformation principal strains of unlike sign) is still useful for separating shear-dominant transformation from the rarer case of dilatation-dominant displacive transformation. Further analysis is needed to more precisely define physical criteria governing the competition between nucleated vs. continuous transformation in weak first-order systems.

APPENDIX: PANEL DISCUSSION

In discussions at the conference, several speakers presented lists of attributes of martensitic transformations similar to those discussed in an earlier classification scheme by Lieberman [32]. These notably included the tendency toward an invariant-plane shape strain. In our discussion here, we have tried to identify the minimum number of primary criteria from which the full range of attributes follow, giving emphasis to criteria which can be tested in a practical manner.

In considering tests of diffusionless character, some discussion concerned the acoustic emission accompanying rapid radial growth as a definitive signature of diffusionless behavior [33]. It was also pointed out that higher temperature lath martensites in steels may involve growth times allowing some carbon partitioning. COC [1] proposed that a transformation is "virtually" diffusionless as long as any incidental diffusion that may occur does not assist the transformation kinetics.

Final discussion raised questions regarding the needed level of precision in defining transformations, with concerns expressed that overly restrictive definitions can have undesirable bureaucratic consequences, potentially including lower funding and smaller ICOMAT conferences. In response, it was emphasized that such difficulties would not arise provided ICOMATs continue to cover martensitic and related transformations. A certain precision of definitions will always be necessary for effective human communication, and this need is particularly acute if we are to protect technical inventions with patents.

Finally, clear definitions need not limit the very fruitful activity of exploring the applicability of principles established in martensite to a broader range of transformations.

ACKNOWLEDGMENTS

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