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Multiple continuity of phases in composite materials: Overall property estimates from a laminate system scheme

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ABSTRACT

With the subsequent goal of estimating effective properties of general n-phase composites, a recent (2016) paper from the author and a co-worker firstly addressed all “elementary” situations of one-level phase arrangements from “all to none phases being co-continuous” and proposed relevant property estimates within the mean field approximation homogenization framework. At the first time to the authors knowledge was proposed an estimate for the arrangements (that occur from 3 phases and are the main type above 3) when *several but not all* the assembled phases are co-continuous. Use was made of a “laminate system (*LS*) scheme” method inspired from a literature of the sixties on “fiber system schemes” likely to ensure phase co-continuity. Laminate systems were similarly figured as interpenetrated one-directional layered structures with layer normal oriented in various directions of space, a realizable representation of which can be multiple slip activity in crystals. The relevancy of using such a *LS* scheme to obtain stiffness property estimates that account for phase co-continuity in composites was then successfully exemplified for various, elastic-like or other (piece-wise) linear properties and the possible use of a dual scheme in terms of compliances was shown to likely correspond to a sort of phase “co-discontinuous” converse assemblage. After a few recalls and a clarification of this co-continuity/co-discontinuity duality, we propose a description of a general phase arrangement from a specified “combination” of the elementary ones. Testing it on a two-phase disordered assemblage, a remarkable relation between elementary estimate types is established that comes in support to the relevancy of the proposed description and to the one of the dual *LS* schemes to accounting for multiple phase continuity. A short application on experimental literature data is also exemplified.

1. Introduction and background

1.1. Introduction

Disregarding multi-phase materials with possible multi-scale (multi-level) assemblages of constitutive phases, the present discussion and study starts from the following three observations:

(i) For assembled two-phases A and B, there are from combinatorial analysis and apart of full disorder, four “elementary” one-level (homogeneous) arrangements: either phase A is embedded in phase B or conversely phase B is embedded in phase A, both phases A and B are co-continuous and the dual case of the latter, to be a priori said “none of A and B phases is continuous”, until clarifications given herein. Phase continuity needs be taken in the connectedness topological sense of a 3D through-sample spanning “infinite” cluster of that phase in the material,[1] and “infinite” being to be understood as large enough compared to the characteristic dimension of the composite microstructure. It is worthy to remind that if a phase is not continuous in that sense of a (simply or multiply) connected medium it is discontinuous, say constituted with *disconnected* finite domains (Serra, 1982; Coster and Chermant, 1989; 2001);

(ii) For more than two phases, new elementary arrangements appear in addition to all, none or a single phase being continuous, which all associate several continuous phases with at least one discontinuous (embedded) one, that is arrangements in which *several but not all* phases are co-continuous, to be called “multiply continuous” or “multi co-continuous” arrangements for short;

(iii) In contrast with the since long and widely studied (in the so-called mean field approximation homogenization frame-

[1] In (Torquato et al., 2003), A bi-continuous composite is “one in which both phases are connected across the sample”.

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work) inclusion-reinforced-matrix arrangements which are cases with a single homogeneous continuous phase embedding *all* other ones, it is only recently that the other three arrangement types also have - in that same framework - a relevant scheme for their effective property estimates. Milestones of step-wise contributions from the author and co-workers on that way will be referred to all along this work, with a few pieces of figures borrowed to them.

For two-phase bicontinuous structures, several (quite many) models have been proposed out of that homogenization framework (e.g. Peng et al., 2001; Roberts and Garboczi, 2002; Gong et al., 2005) that we do not include in our present discussion for in general they do not extend to more phases than two and not to the multi-continuous arrangements here of main concern.

After a few recalls of earlier results and an important clarification concerning the dual situation to full phase co-continuity, the main objective of this work is to propose, for effective properties of general n-phase composites not reducible to a single one of its elementary phase arrangements, a description from some combination or "mixture" of them. A formal application of a proposed such a description to a two-phase disordered arrangement reveals that it implies a remarkable relation between elementary property estimates that gives it some support and particular interest. Also, experimental data from (Torres et al., 2012) allowed to present some validating comparisons.

1.2. Background

In the above specified understanding, continuity (connectedness) of a phase can be one-directional (as for infinite parallel fibers), two-dimensional (as for infinite parallel layers or multi-directional infinite fiber arrangement normal to one direction) or three dimensional (as beam networks, sponge-like or foam-like structures etc.). An infinite 3D continuous network "embedded" in an infinite matrix (Franciosi, 2018; Franciosi et al., 2019) is embedded in a limit manner, and becomes co-continuous with the matrix at this limit, as infinite parallel fibers (or layers) can be in the fiber (or in-layer) direction(s). We will assume for sake of simplifying, that morphologically anisotropic embedded infinite 3D networks result from a stretch of an isotropic structure, what means that we consider anisotropic composite structures of ellipsoidal symmetry in the sense of (Ponte-Castaneda and Willis, 1995; Bornert et al., 1996). Properties possibly be given to the constitutive phases of the structures in concern can be considered of the (piece-wise) linear "generalized-elastic-like" type, at the example of Magneto-Electro-Elastic (MEE) coupled properties, what includes most of elastic-like and dielectric-like sub-cases of interest (Kuoi and Huang, 1997; Lee et al., 2005; Franciosi, 2013).

In composites with more than two (n say) phases, while the number of arrangements with a single ($p=1$) continuous embedding matrix increases linearly with n, there is always a single "fully continuous" arrangement ($p=n$) and always a single dual "fully discontinuous" one ($p=0$). The number of multi-continuous arrangements ($1 < p < n$), the rest of the total one-level arrangement number $A(n)$, rapidly increases as that total does. From simple combinatory analysis, $A(n)$ reads:

$$A(n)_{n\geq 2} = \sum_{p=0}^{n} A_p^{(n)} = \sum_{p=0}^{n} \frac{n!}{p!(n-p)!}$$
$$= (1)_{p=0} + (n)_{p=1} + \sum_{p=2}^{n-1} \frac{n!}{p!(n-p)!} + (1)_{p=n}, \qquad (1)$$

as was introduced in (Franciosi and Charles, 2016), and the repartition of these four different arrangement types in $A(n)$ obeys the histogram of Fig. 1.

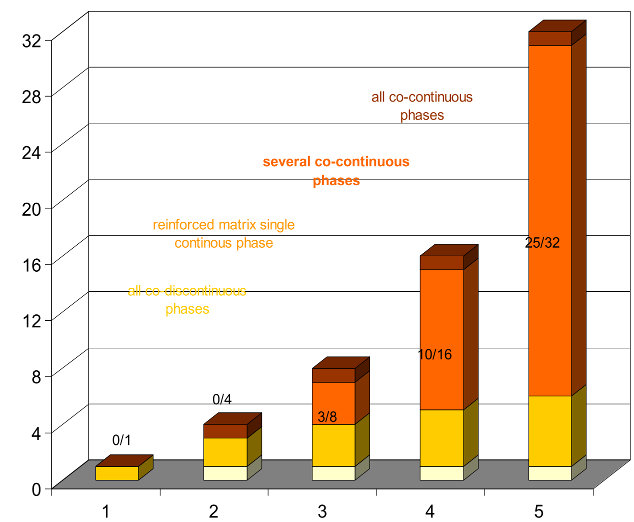

Fig. 1. The various elementary arrangements of n phases up to n = 5; no continuous phase (bottom white); all continuous phases (top brown); a single continuous phase (lower middle yellow); multi-continuous phase arrangements (upper middle orange). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Considering that the Hashin and Shtrikman (*HS*) effective property estimates (Hashin and Shtrikman, 1963; Hashin, 1979; 1983) are reasonably good first order ones in the n cases of multi-reinforced matrix structures ($p=1$), to be called type 1 phase arrangement, the number of other arrangements needing specific estimate schemes is $A(n)-n$: the all co-continuous phase (single $p=n$) case, to be called type 2, the no continuous phase (single $p=0$) case, to be called type 3 or the "multi-continuous" arrangements (all remaining $A(n)-(n+2)$) cases, to be called type 4. A homogeneous medium ($n=p=1$) formally enters the type 1 arrangement as a reinforced matrix at the zero reinforcement limit. It also figures for completeness in the histogram of Fig. 1, although Eq. (1) does not hold for it as written and should be $A(1)=(0)_{p=0}+(1)_{p=1=n}$. However, the forthcoming discussion is concerned with $n \geq 2$ values.

Considering type 2 and type 3 phase arrangements, it is easier, regardless of the value of n, to figure out how the co-continuity of n phases may look like spatially than to figure out how their "co-discontinuity" does. Although an obvious duality of these two arrangements was shown in earlier papers $\forall n$,[2] there is still a missing piece of formalization to properly define the arrangement type that was called "co-discontinuous" as introduced in (Franciosi and El Omri, 2011) as well as the appropriate understanding of this co-discontinuity nature. This is the first point to be addressed for a clarification that highlights the related estimate interpretation.

Considering the number of type 4 arrangements of multiple phase continuity which does not exist for $n=2$, the Fig. 1 and Eq. (1) show that it rapidly increases with n, from 3 assembled phases with 3 over 8 arrangements, 10 over 16 arrangements for $n=4$, 25 over 32 for $n=5$ and so on.

Descriptions of bi-continuous two-phase structures in terms of "Fiber Systems" or "Laminate Systems" were first proposed and examined in (Christensen and Walls, 1972; Boucher, 1974; Christensen, 1979a; b): the "Fiber System" terminology names interpenetrated one-directional bundles of infinite parallel fibers, such that both the matrix and the fiber phases could be considered as continuous in all directions where fibers were oriented, as a multi-directional parallel assemblage of both phases. The effective

[2] Cases with A embedded in B and B embedded in A are only dual ones for n=2.

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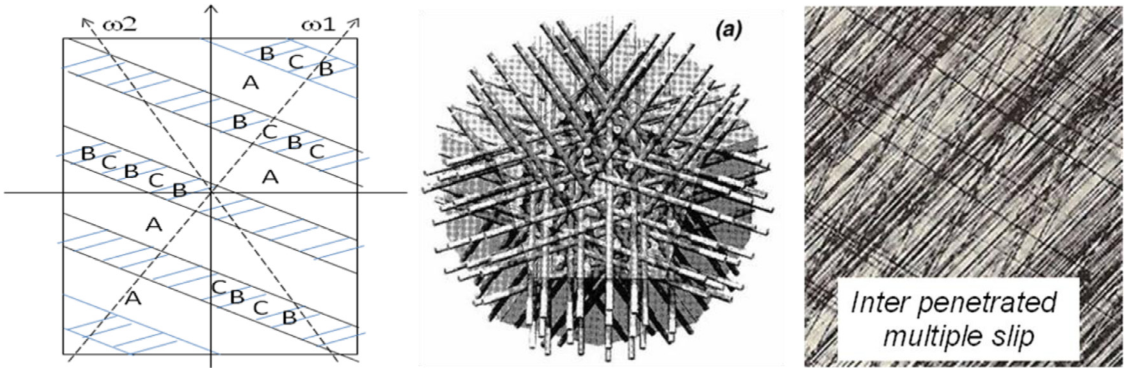

Fig. 2. Examples of (left) hierarchical tri-laminate structure, (middle) isotropic Fiber-System (Dendievel et al., 2002), (right) Laminate System as interpenetrated multiple slip in crystals.

stiffness properties of such a Fiber System were then estimated from the arithmetic average over all the directions $\boldsymbol{\omega}$ on the unit sphere Ω of the 1D bundle effective stiffness $\mathbf{C}^{*F\omega}_{(A,B)} = \mathbf{C}^{*F(\theta,\phi)}_{(A,B)}(\theta,\phi)$ defining the considered system as (when isotropic):

$$\mathbf{C}^{*IFS}_{(A,B)} = (4\pi)^{-1}\iint_{\theta,\phi} \mathbf{C}^{*F(\theta,\phi)}_{(A,B)}(\theta,\phi)\sin\theta d\theta d\phi = (4\pi)^{-1}\int_{\Omega} \mathbf{C}^{*F\omega}_{(A,B)} d\omega \tag{2}$$

Similar moduli for anisotropic structures of that sort result from appropriately weighting the $\mathbf{C}^{*F\omega}_{(A,B)}$ directional contributions in Eq. (2), as will be seen in the following for any phase number n.

Indeed, phase co-continuity is expected to improve the composite stiffness in the direction(s) of co-continuity, as was for example shown in (Torquato et al., 2003) in a search for topological two-phase arrangements that optimize pairs of property moduli such as conductivity and compressibility. This reference geometrically exemplifies such a bi-continuous phase arrangement by a Schwarz two-phase minimal surface of P type (see also Scriven, 1976; Zhou and Li, 2007), to be exemplified in the following as being also a basis for building families of n-phase composites with any p co-continuous ones.

First regardless of practical realization, a similar arithmetic average over a set of directions in space is mathematically possible, summing over the effective stiffness properties of directional layered (laminate) structures, which are planar (2D) assemblages of two phases, while the fiber bundles are (1D). Similarly to the fiber case, making use of the moduli tensors of the $\boldsymbol{\omega}$-oriented laminate (A,B) structure $\mathbf{C}^{*L\omega}_{(A,B)} = \mathbf{C}^{*L(\theta,\phi)}_{(A,B)}(\theta,\phi)$, the moduli estimate reads (for isotropic structures):

$$\begin{aligned}\mathbf{C}^{*ILS}_{(A,B)} &= (4\pi)^{-1}\iint_{\theta,\phi} \mathbf{C}^{*L(\theta,\phi)}_{(A,B)}(\theta,\phi)\sin\theta d\theta d\phi \\ &= (4\pi)^{-1}\int_{\Omega} \mathbf{C}^{*L\omega}_{(A,B)} d\omega,\end{aligned} \tag{3}$$

The obtained effective properties from “Laminate Systems” (*LSs*) defined in (Christensen 1979a;b) as “implying intersecting platelets of some kind” and “suggestive of morphology known as interpenetrating networks of (two) phases”,[3] were likely to account for phase co-continuity in all the in-layer orientations of each directional laminate of the system, as discussed for n phases in (Franciosi and El Omri, 2011).

Laminate System (*LS*) and Fiber System (*FS*) schemes conversely present important differences:

– Laminated structures are very particular ones in the world of composite materials, owing to several specific properties they benefit from, which can be related to characteristics of their representative Green operators (Walpole, 1981);
– while a *n*-phase $\boldsymbol{\omega}$-oriented fiber bundle has not uniquely defined effective directional properties (they depend on which phase(s) act(s) as the necessary fiber-embedding matrix, such that there is an upper *FS(+)* (resp. lower *FS(−)*) estimate when the stiffest (resp. the weakest) phase is chosen as the matrix), all *n* phases play an equivalent role in an $\boldsymbol{\omega}$-oriented laminate structure and the effective properties are uniquely and exactly defined. This makes Laminate Systems more relevant than Fiber Systems when none of the assembled phases is expected to play a particular role;
– Conceiving how laminate structures could be interpenetrated to make a Laminate System - not to be confused with hierarchical laminates (Quintanilla and Torquato, 1996; Milton, 2005), which are multi-scale schemes for microstructure descriptions, as exemplified in Fig. 2 **left** - is harder than conceiving how fiber bundles can be, what was exemplified in (Dendievel et al., 2002) and is shown in Fig. 2**, middle**. A helpful visualization of interpenetrated laminate structures is provided by examining how multiple slip in crystals can be interpenetrated (shown Fig. 2**, right**) and descriptions of slip activity in terms of Laminate Systems were attempted in the context of polycrystal plasticity modeling in (Franciosi and Berbenni, 2007; 2008; Franciosi, 2012a);
– One key point when describing effective properties of a multi-continuous composite from a linear combination of the properties of directional structures is that whether performed in terms of stiffness moduli $\boldsymbol{C}$ or in terms of compliances $\boldsymbol{S} = \boldsymbol{C}^{-1}$, the two schemes and their results are not equivalent: an arithmetic average over stiffness moduli corresponds to a harmonic average over compliance moduli and conversely. Consequently, property estimates from either a Fiber System or a Laminate System scheme go by pairs, (*mFS/cFS*) or (*mLS/cLS*) say, whether stiffness moduli (m) or compliances (c) are considered, with the difference that while the Fiber System pairs are many and in increasing number with *n* (at least equal to the phase number *n* when only considering single phased matrices) and in ranges {*mFS(−),mFS(+)*} and {*cFS(−),cFS(+)*}, the Laminate System pair (*mLS,cLS*) is unique for each n values. This stiffness/compliance duality was neither examined nor commented in the early works on Fiber and Laminate systems for co-continuous phases. It was mentioned in (Franciosi et al., 2011) and studied first time in (Franciosi and El Omri, 2011). Several fundamental inequalities were established rigorously there, showing that for any n-phase set, all property estimates from *FS* and *LS* schemes

[3] What Christensen inappropriately figured as an isotropic aggregate of $\boldsymbol{\omega}$-oriented laminate domains.

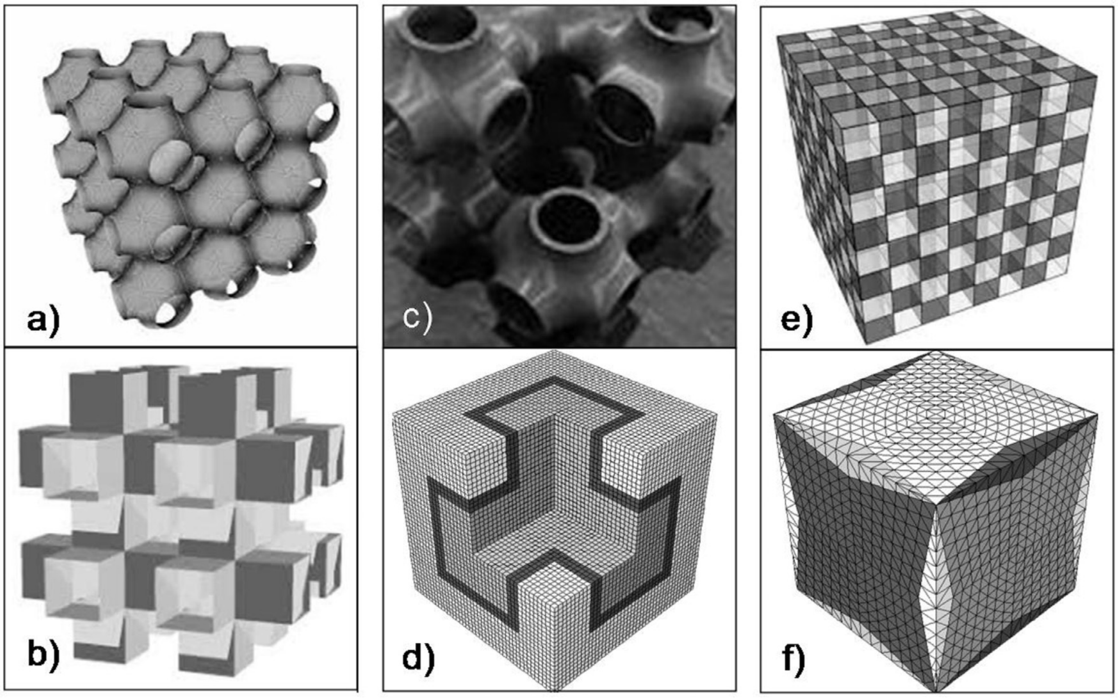

Fig. 3. (a) Minimal Schwarz P surface; (b) a cubic variant of case a; (c) a tricontinuous composite from thickening the case a surface; (d) 1/8th of the unit cubic cell of a tricontinuous composite made from thickening the case b surface; (e) a 3D checkerboard as minimally co-continuous two-phase composite; (f) a tri-phase minimally co-continuous composite (cases d and f borrowed to Franciosi and Charles, 2016).

of both dual (stiffness and compliance) types were - as required - interior to the Hashin & Shtrikman bounds, both defining a subdomain in the *HS* one, with also the (*mLS/cLS*) domain being always interior to the (*mFS(+)/cLS(−)*) one.

Also, as far as these two dual (Stiffness versus Compliance) averaging assumptions are expected to correspond to two dual situations of phase arrangements, if the stiffness arithmetic average finds supports to stand for phase co-continuity, the arithmetic average of compliances must “somehow” correspond to a sort of “phase co-discontinuity”. Hence, for all assembled phases being on equal ground, the two Laminate System estimates determine a domain for phase co-continuity situations, from all to none being co-continuous, “none” being thought to mean all co-discontinuous then in the early proposed interpretation. Co-discontinuity of n phases being questioning, this duality will be here clarified in terms of maximal versus minimal phase co-continuity, with a simple demonstration that the latter arrangement is the closest one to full co-discontinuity in the sense of all co-disconnected phases which is a topological impossibility according to the definitions.

The Fig. 3a-d, present how one can build n-phase fully co-continuous composites from a mathematical minimal Schwarz surface (Scriven, 1976; Torquato et al., 2003; Zhou and Li, 2007), Fig. 3a, of which the Fig. 3b shows a cubic variant. Giving some thickness to such surfaces which share space into two co-continuous subspaces (Fig. 3c and d) yields a tri-continuous phase arrangement (Fig. 3d represents one eighth of a unit cubic cell). Duplicating this surface into $n-2$ layers of different phases yields a family of n co-continuous composites. Fig. 3e exemplifies a so-called “3D checkerboard”, as for example claimed in (Torquato et al., 2003) to correspond with some minimal situation of co-continuous two-phases structures. The characteristic of the interconnections between cubic domains of a same phase is to have null areas (edge-connected phase homologous domains). The Fig. 3f, shows a 3-phase example of such minimal co-continuity, the polyhedral domains of a same phase being point-connected, the cube center being the common top of 3 pairs of same two opposite pyramids whose bases are the cube sides. Fig. 3d and f are from (Franciosi and Charles, 2016).

Coming in addition to the $A(n)$ elementary phase arrangement types of the nomenclature which is presented in Fig. 1, the disordered arrangement of n phases, is reasonably well described in terms of effective property estimate from the use of a Self-Consistent (*SC*) scheme (Kröner, 1958; 1961). In contrast with the *HS* estimates that correspond to a same phase being the (continuous) matrix regardless of its concentration in the composite, disordered composite structures are materials with the phase arrangement being generally dependent on the relative phase concentrations: a dense phase becomes more likely a continuous (connected) one while dilute phases expectedly turn to embedded (disconnected) situations in it. In between, when several phases are in comparable concentrations (provided them be in limited number), disorder can correspond to situations of multiple continuity. For the two-phase case, the phase bi-continuity is a likely arrangement for such a disordered mixture which is often seen as a “percolation” (transition) concentration range separating two domains where the dense (continuous) and dilute (discontinuous) phases interchange their roles progressively with the concentration varying.

If on the contrary the phase number is large, the “co-discontinuous” (minimally co-continuous) arrangement becomes an expectable one in the range of comparable concentrations.

These preliminaries on how a disordered structure may be phase-arranged point out that phase multiple continuity may be evolving with phase concentration changes and that any general structure may be a combination of various elementary arrangements of its phases, with relative weights of these arrangements being phase volume fraction dependent.

The possible description of a general composite as some “mixture” (in a way to be specified) of its elementary phase arrangements is the major point that we finally address in this work.

A comparison of property estimates from the Laminate System (*LS*) schemes with those from the related *SC* estimate in the two-phase case (Franciosi, 2012b) has provided a first morphological argument on how a *LS* estimate from a stiffness moduli averaging (*mLS*) can be seen to well corresponds to (maximally) co-continuously assembled phases and conversely how a *LS* estimate from a compliance averaging (*cLS*) well corresponds to structures with all of the assembled phases being co-disconnected (minimally co-connected) throughout the sample: the *SC*(n) property estimate for a n-phase disordered composite corresponds with all n phases being embedded in a $n+1^{th}$ additional one of infinitesimal volume fraction, the properties of which are those of the composite itself. This $(n+1)$-phase equivalent description for n-phase structures with the $n+1^{th}$ one being a reference embedding (continuous) matrix in infinitesimal concentration is always permitted, whether the composite has a disordered structure or a more specific one: for the *HS* estimates, this $n+1^{th}$ infinitesimal phase is trivially the continuous one itself. The three-phase scheme of the two-phase A, B composite when phase A (resp. phase B) is the matrix, is then A and B phases embedded in an infinitesimal third matrix made of phase A (resp. B).

The determination of this $n+1^{th}$ matrix has been examined (in the cited earlier papers) for the estimates from the Laminate System schemes, first for n = 2 and then for any n value. Then, from the *LS* estimates of n-phases, the reference matrix for the cases mixing several continuous phases with at least one embedded one has also been defined. This will be briefly recalled in Section 2.

Owing to these results, the important request for estimating properties of a general n-phase composite case appears also be to specifying the properties of its specific infinitesimal reference matrix $n+1^{th}$ phase. We propose a possible description of that type for any general one-level n-phase composite, based on the determination of a reference $n+1^{th}$ phase for it, which is assumed to be a disordered mixture of the reference matrices of its $A(n)$ elementary arrangements. The proposed description is first tested for the case of a disordered two-phase arrangement which is supposed to have effective properties given by the *SC* estimate. In so doing, a remarkable relation is established from this description, between the property estimates for the two-phase elementary arrangements. This relation provides support both to the relevancy of the proposed description itself and to the relevancy of the two *LS* schemes to estimate effective properties for all assemblages with multiple phase continuity. An application on literature data is finally reported.

1.3. Paper organization

Section 2 briefly recalls the definitions of the various schemes (*HS(n), SC(n), mLS(n), cLS(n), mLS(p,n)*) of property estimates that will be here of concern, for $n=2$ first and for $n>2$ then. It also briefly recalls insights from the earlier published works about *LS* schemes which will be useful. In Section 3, the *mLS(n)* and *cLS(n)* estimates are reintroduced such as to establish the duality of these *LS* estimates in terms of maximal versus minimal phase co-continuity. Section 4 examines the description of a general n-phase composite in terms of the *A(n)* elementary arrangements of its phases and proposes, based on determining the $n+1^{th}$ reference infinitesimal matrix, a relevant effective property estimate form. In Section 5, the proposed "reference matrix mixture" description is first shown to fulfill a remarkable relation in one-modulus isotropic symmetries. It then reports the performed multi-moduli application on literature data. Section 6 concludes.

2. The laminate system schemes for phase multiple continuity in composites

The discussion is held in the so-called mean field approximation framework for the effective properties of a composite material (m^*) constituted with a "reference matrix" (m^o) of moduli tensor $\boldsymbol{C}^o$ and with assembled n phases (of moduli tensors $\boldsymbol{C}_i$, $i=1, n$). Those are made of non overlapping congruent domains of ellipsoidal shape in homothetic[4] ellipsoidal spatial distribution represented by the same strain "Green operator" (for short[5]) $\boldsymbol{t}_{\boldsymbol{C}^o}^{Vell}$ or Eshelby tensor $\boldsymbol{E}_{\boldsymbol{C}^o}^{Vell}$. We here consider that this reference matrix is a $n+1^{th}$ continuous phase of volume fraction $f_{m^\circ}\approx 0$, supposed to embed all other ones. This includes the possibilities that the infinitesimal matrix identifies with one of the n constitutive phases and that one or more of the embedded phases are co-continuous with it. The phase domains are assumed to be shape-distributed down to infinitesimal in order to allow total space filling. For this background, one can refer to the two-point distribution modeling proposed by (Ponte-Castaneda and Willis, 1995) *(PCW)* of which it is the simple case that also matches with the classical (Hashin-Shtrikman, 1963) (*HS*) frame and can be found in a huge fraction of the later literature concerned with homogenization methods, with also connections to new developments (e.g. Hu and Weng, 2000; Buryachenko, 2007). Dielectric-like properties are a sub-case of elastic-like ones and various coupling types of such properties, as the magneto-electro-elastic (MEE) type, can also be formalized from the elastic type, using the extended notation of (Barnett and Lothe, 1975; Alshits et al., 1992). We therefore develop our discussions using the formalism of effective elastic properties, in which case, the "moduli" strictly speaking refer to stiffness moduli. But moduli of other (linear) properties are similarly represented which may be also called "generalized" stiffness moduli by extension. The same holds for the "inverse moduli" $\boldsymbol{S}^o=(\boldsymbol{C}^o)^{-1}=\overline{\boldsymbol{C}^o}$, which strictly speaking stand for compliances in elasticity but inverse moduli of other properties may also be seen as "generalized" compliances.

Accordingly, generalized stiffness (resp. compliance) tensors link a generalized stress (resp. strain) tensor to a generalized strain (resp. stress) tensor. The Eshelby tensor (Eshelby, 1957) and the strain Green operator for a given ellipsoidal shape (or symmetry) V in the infinite medium (m^o) are linked by the relation $\boldsymbol{t}_{\boldsymbol{C}^o}^{Vell}=\boldsymbol{E}_{\boldsymbol{C}^o}^{Vell}:\boldsymbol{S}^o=\boldsymbol{S}^o:\boldsymbol{E}_{\boldsymbol{C}^o}^{tVell}=\boldsymbol{t}_{\boldsymbol{C}^o}^{tVell}$, where the upper-script "t" stands for "transpose of". In particular, this "strain" Green operator has a dual "stress" one $\boldsymbol{t'}_{\boldsymbol{S}^o}^{Vell}$:

$$\boldsymbol{t'}_{\boldsymbol{S}^o}^{Vell}=\boldsymbol{C}^o-\boldsymbol{C}^o:\boldsymbol{t}_{\boldsymbol{C}^o}^{Vell}:\boldsymbol{C}^o \quad ; \quad \boldsymbol{t}_{\boldsymbol{C}^o}^{Vell}=\boldsymbol{S}^o-\boldsymbol{S}^o:\boldsymbol{t'}_{\boldsymbol{S}^o}^{Vell}:\boldsymbol{S}^o, \tag{4}$$

(Zeller and Dederich, 1973; Walpole, 1981). When elastic-like (say rank four), both Green operators are ij/kl (super) symmetric while the Eshelby tensor is not.[6] Both Green operators furthermore are positive definite, what the Eshelby tensor is not either. Under the specified conditions in this so-called "dilute approximation framework" for the representative volume element (RVE) is an isolated inclusion (or finite pattern) in an infinite medium (Berveiller et al., 1987; Hashin and Shtrikman, 1963; Ponte Castaneda and Willis, 1995; Cherkaev, 2000; Buryachenko, 2001) the effective "stiffness moduli" of *any* anisotropic n-phase composite as described can be

[4] The phase spatial distribution is assumed of same ellipsoidal symmetry as the shape of all embedded domains when any, what allows this same ellipsoidal symmetry to be globally assumed for the considered composite structures.

[5] The twice differentiated strain Green operator integral over the ellipsoidal domain of shape VEll.

[6] Although not used, a "stress Eshelby" dual tensor can be defined in connection to the stress Green operator as the Eshelby tensor connects to the strain Green operator, using compliance moduli instead of stiffness ones.

written:

$$\boldsymbol{C}^{eff(m*)}_{(i,j,\dots,n)\subset m^o} = \boldsymbol{C}^o - \left(\left(\left(\sum_{i=1}^{n} f_i\left((\boldsymbol{C}^o-\boldsymbol{C}_i)^{-1} - \boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}\right)^{-1}\right)^{-1} + \boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}\right)\right)^{-1}; \quad \sum_{i=1}^{n} f_i = 1 - f_{m^\circ} = 1. \tag{5}$$

In Eq. (5), the estimate can be obtained for specified phase properties and concentrations provided the properties $\boldsymbol{C}^o$ of the reference $n+1^{th}$ matrix of infinitesimal volume fraction be identified.[7] The (uniform) Green operator of ellipsoids can be put, thanks to the Radon transform and its inversion formula (Franciosi and Lormand, 2004), under the double integral form $\boldsymbol{t}^{Vell}_{\boldsymbol{C}^o} = \iint_\Omega \boldsymbol{t}^{L(\theta,\phi)}_{\boldsymbol{C}^o}\psi_{Vell1}(\theta,\phi)\sin\theta d\theta d\phi = \int_\Omega \boldsymbol{t}^{L(\omega)}_{\boldsymbol{C}^o}\psi_{Vell1}(\omega)d\omega$. It is a weighted ω integral over the unit sphere Ω of the Green operators $\boldsymbol{t}^{L(\omega)}_{\boldsymbol{C}^o}$ for $\boldsymbol{\omega}$-oriented platelets or laminates, defined with regard to the reference medium (m^o). With the phase domains *Vell* being ellipsoidal, the weight (or shape) function writes $\psi_{Vell}(\omega) = (\frac{3}{4\pi})^2(\frac{v}{3\,D_V(\omega)^3})$, where $D_V(\omega)$ is the half breadth of *Vell* in the ω direction and v its volume.[8] In Eq. (5), the first occurrence of $\boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}$ in the innermost brackets stand for the common shape of all domains and the second occurrence for their spatial distribution. This operator identity is the simplest assumption consistent with total space filling by the inclusions. Non ellipsoidal or multimodal spatial distributions of inclusions or of inclusion patterns (Bornert et al., 1996; Franciosi and Lebail, 2004; Franciosi et al., 2019) are here disregarded.

As already pointed, Eq. (5) provides the well known Hashin-Shtrikman (*HS*) estimates for any choice among the n possibilities of selecting a $\boldsymbol{C}^o = \boldsymbol{C}_i$ $n+1^{th}$ reference matrix from the constitutive phases of the composite.[9] Since $f_{m^o}=0$, Eq. (5) further provides the implicit *SC* estimate when choosing $\boldsymbol{C}^o = \boldsymbol{C}^{m*}_{(i,j,\dots,n)\subset m*}$, the reference infinitesimal matrix (m^o) being the effective medium (m*) itself then. Any other property estimate for the same n assembled phases is expected to correspond to another arrangement type and thus to another specific $n+1^{th}$ reference medium (m^o). The estimates resulting from Laminate System schemes do are characterized by a reference matrix as established in cited previous works and recalled in the following.

2.1. Laminate system schemes and estimates for the two-phase cases

For sake of simplification, omitting (m^o) or (m*) and denoting for room saving $\overline{\boldsymbol{T}}$ instead of $\boldsymbol{T}^{-1}$ the inverse of some tensor forms $\boldsymbol{T}$, we specialize for $n=2$ to write, for $f_A+f_B=1$, Eq. (5) as:

$$\boldsymbol{C}^*_{[A,B]} = \boldsymbol{C}^o - \left(\left(f_A\left((\boldsymbol{C}^o-\boldsymbol{C}_A)^{-1} - \boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}\right)^{-1} + f_B\left((\boldsymbol{C}^o-\boldsymbol{C}_B)^{-1} - \boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}\right)^{-1}\right)^{-1} + \boldsymbol{t}^{Vell}_{\boldsymbol{C}^o}\right)^{-1}, \tag{6a}$$

with the compliance inverse *equivalent* counterpart, also using the stress Green operator $\boldsymbol{t'}^{Vell}_{\boldsymbol{S}^o}$:

$$\boldsymbol{S}^*_{[A,B]} = \boldsymbol{S}^o - \left(\left(f_A\left((\boldsymbol{S}^o-\boldsymbol{S}_A)^{-1} - \boldsymbol{t'}^{Vell}_{\boldsymbol{S}^o}\right)^{-1} + f_B\left((\boldsymbol{S}^o-\boldsymbol{S}_B)^{-1} - \boldsymbol{t'}^{Vell}_{\boldsymbol{S}^o}\right)^{-1}\right)^{-1} + \boldsymbol{t'}^{Vell}_{\boldsymbol{S}^o}\right)^{-1} = \overline{\boldsymbol{C}^*_{[A,B]}}. \tag{6b}$$

The *HS* estimates, that correspond to either phase B embedded in phase A or the converse, result from taking $\boldsymbol{C}^o=\boldsymbol{C}_A$ or $\boldsymbol{C}^o=\boldsymbol{C}_B$ in Eq. (6a), identical to taking $\boldsymbol{S}^o=\boldsymbol{S}_A$ or $\boldsymbol{S}^o=\boldsymbol{S}_B$ in Eq. (6b).

When assuming $\boldsymbol{C}^o=\boldsymbol{C}^{*SC}_{[A,B]}$ (or equivalently $\boldsymbol{S}^o=\boldsymbol{S}^{*SC}_{[A,B]}=\overline{\boldsymbol{C}^{*SC}_{[A,B]}}$) for the *SC* estimate, the Eq. (6a) takes the implicit form $f_A(\overline{(\boldsymbol{C}^{*SC}_{[A,B]}-\boldsymbol{C}_A)-\boldsymbol{t}^{Vell}_{\boldsymbol{C}^{*SC}_{[A,B]}}}) + f_B(\overline{(\boldsymbol{C}^{*SC}_{[A,B]}-\boldsymbol{C}_B)-\boldsymbol{t}^{Velll}_{\boldsymbol{C}^{*SC}_{[A,B]}}}) = 0$ and conversely Eq. (6b) with regard to the compliance form. After some manipulations and using $\langle\boldsymbol{C}\rangle=\langle\boldsymbol{C}\rangle_{[A,B]}=f_A\boldsymbol{C}_A+f_B\boldsymbol{C}_B$ the material's Voigt upper bound (arithmetic average) for stiffness, $\boldsymbol{C}^{*SC}_{[A,B]}$ can be also written (Franciosi et al., 2011):

$$\boldsymbol{C}^{*SC}_{[A,B]} = \langle\boldsymbol{C}\rangle + (\boldsymbol{C}^{*SC}_{[A,B]}-\boldsymbol{C}_A):\boldsymbol{t}^{Vell}_{\boldsymbol{C}^{*SC}_{[A,B]}}:(\boldsymbol{C}^{*SC}_{[A,B]}-\boldsymbol{C}_B) = \overline{\boldsymbol{S}^{*SC}_{[A,B]}}. \tag{7}$$

This identity between Eqs. (6a) and (7) for $\boldsymbol{C}^{*SC}_{[A,B]}$ in terms of phase moduli can conversely be established in terms of compliances to similarly yield $\boldsymbol{S}^{*SC}_{[A,B]}=\overline{\boldsymbol{C}^{*SC}_{[A,B]}}$, also using the stress Green operator $\boldsymbol{t'}^{Vell}_{\boldsymbol{C}^{*SC}_{[A,B]}} = \boldsymbol{C}^{*SC}_{[A,B]} - \boldsymbol{C}^{*SC}_{[A,B]}:\boldsymbol{t}^{Vell}_{\boldsymbol{C}^{*SC}_{[A,B]}}:\boldsymbol{C}^{*SC}_{[A,B]}$ and $\langle\boldsymbol{S}\rangle=\langle\boldsymbol{S}\rangle_{[A,B]}=f_A\boldsymbol{S}_A+f_B\boldsymbol{S}_B$.

The Eq. (6a) cannot be put under the form of Eq. (7) for the effective properties $\boldsymbol{C}^*_{(a,b)}$ of a general two-phase composite with reference stiffness matrix $\boldsymbol{C}^o$. The only possible cases, as proved in (Franciosi, 2012b), are for (i) the composite whose stiffness reference matrix is $\boldsymbol{C}^o=\{\boldsymbol{C}\}=\{\boldsymbol{C}\}_{[A,B]}=f_B\boldsymbol{C}_A+f_A\boldsymbol{C}_B$ and in dual manner (putting Eq. (6b) into the inverse form of Eq. (7)) for (ii) the composite whose compliance reference matrix is $\boldsymbol{S}^o==\{\boldsymbol{S}\}=\{\boldsymbol{S}\}_{[A,B]}=f_B\boldsymbol{S}_A+f_A\boldsymbol{S}_B$. $\{\boldsymbol{C}\}$ is the Voigt upper stiffness bound for the two-phase [*B*, *A*] "harmonic material" (e.g. the medium with interchanged phases or of same phases but with interchanged concentration proportions) that we denote]*A*, *B*[and $\{\boldsymbol{S}\}$ is its Voigt upper compliance bound, such that $\{\boldsymbol{C}\}=\{\boldsymbol{C}\}_{[A,B]}=\langle\boldsymbol{C}\rangle_{]A,B[}$, $\{\boldsymbol{S}\}=\{\boldsymbol{S}\}_{[A,B]}=\langle\boldsymbol{S}\rangle_{]A,B[}$ and $\{\boldsymbol{S}\}\neq\{\boldsymbol{C}\}^{-1}$.

These reference matrices are those characterizing the two-phase composite effective properties from respectively the stiffness and the compliance Laminate System Scheme (Franciosi et al., 2011; Franciosi and El Omri, 2011; Franciosi, 2012b), which can be finally written:

$$\boldsymbol{C}^{*mLS}_{[A,B]} = \langle\boldsymbol{C}\rangle + (\{\boldsymbol{C}\}-\boldsymbol{C}_A):\boldsymbol{t}^{Vell}_{\{\boldsymbol{C}\}}:(\{\boldsymbol{C}\}-\boldsymbol{C}_B), \tag{8a}$$

$$\boldsymbol{S}^{*cLS}_{[A,B]} = \langle\boldsymbol{S}\rangle + (\{\boldsymbol{S}\}-\boldsymbol{S}_A):\boldsymbol{t'}^{Vell}_{\{\boldsymbol{S}\}}:(\{\boldsymbol{S}\}-\boldsymbol{S}_B) = \left(\boldsymbol{C}^{*cLS}_{[A,B]}\right)^{-1}. \tag{8b}$$

As was established in the cited earlier papers, Eq. (8) result from the more general Eq. (6) owing to fundamental properties of the laminate Green operators, the *exact* effective properties of which obey *both* Eq. (8) equivalently, provided the Green operator of use is the laminate strain (in Eq. (8a)) or stress (in Eq. (8b)) one, say:

$$\begin{aligned}\boldsymbol{C}^{*L(\omega)}_{[A,B]} &= \langle\boldsymbol{C}\rangle - f_Af_B(\boldsymbol{C}_A-\boldsymbol{C}_B):\boldsymbol{t}^{L(\omega)}_{\{\boldsymbol{C}\}}:(\boldsymbol{C}_A-\boldsymbol{C}_B)\\ &= \langle\boldsymbol{C}\rangle + (\{\boldsymbol{C}\}-\boldsymbol{C}_A):\boldsymbol{t}^{L(\omega)}_{\{\boldsymbol{C}\}}:(\{\boldsymbol{C}\}-\boldsymbol{C}_B),\end{aligned} \tag{9a}$$

$$\begin{aligned}\boldsymbol{S}^{*L(\omega)}_{[A,B]} &= \langle\boldsymbol{S}\rangle - f_Af_B(\boldsymbol{S}_A-\boldsymbol{S}_B):\boldsymbol{t'}^{L(\omega)}_{\{\boldsymbol{S}\}}:(\boldsymbol{S}_A-\boldsymbol{S}_B)\\ &= \langle\boldsymbol{S}\rangle + (\{\boldsymbol{S}\}-\boldsymbol{S}_A):\boldsymbol{t'}^{L(\omega)}_{\{\boldsymbol{S}\}}:(\{\boldsymbol{S}\}-\boldsymbol{S}_B) = \overline{\boldsymbol{C}^{*L(\omega)}_{[A,B]}}.\end{aligned} \tag{9b}$$

Thus, *by definition*, the effective properties from the *mLS* and *cLS* schemes of a two-phase material whose overall structure symmetry is characterized by an ellipsoidal shape *Vell* are obtained by a weighted arithmetic averaging of the Eq. (9) with weight function $\psi_{Vell}(\omega)$ of ellipsoidal (including isotropic

[7] If $\boldsymbol{C}^o$ identifies to one of the $\boldsymbol{C}_i$ phase tensors, the term with $\boldsymbol{C}^o-\boldsymbol{C}_i$ vanishes from the sum.

[8] For spherical inclusions and more generally for isotropic symmetry, $\psi_{Sph}(\omega)=\psi_{Iso}(\omega)=1/4\pi \quad \forall\omega=(\theta,\ \phi)$. The integral over Ω is analytic only in particular (the simplest) property symmetries of the reference matrix $\boldsymbol{C}^o$.

[9] It also yields the so-called *HS* lower and upper stiffness bounds when $\boldsymbol{C}^o=\min(\boldsymbol{C}_i)$ or $\boldsymbol{C}^o=\max(\boldsymbol{C}_i)$, corresponding to the upper and lower compliance bounds.

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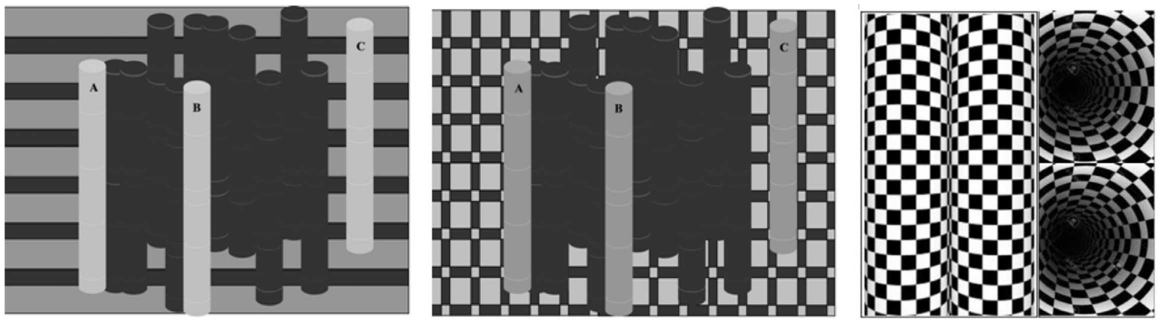

Fig. 4. Examples of third phase wrapping reference matrix for a two-phase fiber bundle with properties ensuring maximal co-continuity (left) and minimal one (middle), (from Franciosi, 2012b) and a zoomed view on two adjacent fibers of the in and out structures of the wrapping layer (right).

$\psi_{Iso}(\omega)=\psi_{Sph}(\omega)=1/4\pi$) symmetry ($\int_\Omega \psi_{Vell}(\omega)d\omega=1$), either in terms of stiffness moduli (for *mLS*) or in terms of compliance ones (for *cLS*). Thus, from Eq. (9), the effective *mLS* and *cLS* estimates for two-phase composites read in terms of stiffness moduli:

$$\boldsymbol{C}^{*mLS}_{[A,B]}=\int_\Omega \psi_{Vell}(\omega)\boldsymbol{C}^{*L(\omega)}_{[A,B]}d\omega;$$
$$\boldsymbol{C}^{*cLS}_{[A,B]}=\overline{\boldsymbol{S}^{*cLS}_{[A,B]}}=\left(\int_\Omega \psi_{Vell}(\omega)\boldsymbol{S}^{*L(\omega)}_{[A,B]}d\omega\right)^{-1}, \tag{10}$$

and conversely in terms of compliances. Owing to the form of the Eq. (10) for the as defined *LS* schemes, the Eq. (8) amount to averaging in Eq. (9) the laminate operators $\boldsymbol{t}^{L(\omega)}_{\{\boldsymbol{C}\}}$ or $\boldsymbol{t}'^{L(\omega)}_{\{\boldsymbol{S}\}}$ (not corresponding to a same medium since $\{\boldsymbol{S}\}\neq\{\boldsymbol{C}\}^{-1}$), over all ω directions around the unit sphere, according to the weight function $\psi_{Vell}(\omega)$. This exactly corresponds with performing the calculation of the strain (resp. stress) Green operator for the ellipsoidal shape *Vell* in term of its Radon polar decomposition in R^3. The comparison with the two-phase related *SC* estimate, i-e the *SC* estimate for the same ellipsoidal symmetry, allowed to establish, with respectively setting $\boldsymbol{C}^o=\{\boldsymbol{C}\}$ and $\boldsymbol{S}^o=\{\boldsymbol{S}\}$, the *mLS* and *cLS* estimates for two-phase composites as:

$$\boldsymbol{C}^{*mLS}_{[A,B]}=\{\boldsymbol{C}\}-\left(\left(f_A\left((\{\boldsymbol{C}\}-\boldsymbol{C}_A)^{-1}-\boldsymbol{t}^{Vell}_{\{\boldsymbol{C}\}}\right)^{-1}\right.\right.$$
$$\left.\left.+f_B\left((\{\boldsymbol{C}\}-\boldsymbol{C}_B)^{-1}-\boldsymbol{t}^{Vell}_{\{\boldsymbol{C}\}}\right)^{-1}\right)^{-1}+\boldsymbol{t}^{Vell}_{\{\boldsymbol{C}\}}\right)^{-1}, \tag{11a}$$

$$\boldsymbol{S}^{*cLS}_{[A,B]}=\{\boldsymbol{S}\}-\left(\left(f_A\left((\{\boldsymbol{S}\}-\boldsymbol{S}_A)^{-1}-\boldsymbol{t}'^{Vell}_{\{\boldsymbol{S}\}}\right)^{-1}\right.\right.$$
$$\left.\left.+f_B\left((\{\boldsymbol{S}\}-\boldsymbol{S}_B)^{-1}-\boldsymbol{t}'^{Velll}_{\{\boldsymbol{S}\}}\right)^{-1}\right)^{-1}+\boldsymbol{t}'^{Velll}_{\{\boldsymbol{S}\}}\right)^{-1}, \tag{11b}$$

which are the special (non equivalent) forms of Eq. (6) for the *mLS* and *cLS* two-phase estimates.

The equivalency of Eq. (8) with Eq. (11) was a first help to figuring the type of material architecture represented by the *LS* estimates (in the two-phase case): they correspond with two embedded phases in an infinitesimal matrix which is an inverse mixture of these two phases in the bulk material, the dilute phase of the bulk being the dense phase in the matrix layer and conversely the dense phase of the bulk being in dilute concentration. With Voigt-like stiffness moduli $\{\boldsymbol{C}\}$ this "harmonic matrix" $[B, A]\equiv\,]A, B[$ ensures at all concentrations for the *mLS* estimate the highest stiffness for the two phases, what is likely corresponding to optimally co-connected phases. With Voigt-like compliances $\{\boldsymbol{S}\}$ (Reuss-like for stiffness) in the cLS Scheme, it conversely realizes the lowest stiffness for the symmetric two-phase assemblage such as to correspond to optimally co-disconnected phases.

From the effective property forms obtained for the reference matrix phase, corresponding structures for isotropic co-continuous and "co-discontinuous" two-phase materials have been tentatively illustrated in (Franciosi and El Omri, 2011) and in (Franciosi 2012b, 2013) for 3D and 2D cases respectively. Basically, an infinitesimal layer of matrix with $\{\boldsymbol{C}\}$ stiffness wrapping parallel fibers can be seen (Fig. 4 **left**) as a sort of 2D welded structure with long stripes of A and B phases normal to the fiber direction, the largest ones for the densest phase in the matrix to connect the more dilute - distant - phase domains in the bulk. A similar 2D layer with $\{\boldsymbol{S}\}^{-1}$ stiffness can be seen (Fig. 4 **middle**) as a double faced, generally irregular, checkerboard with antagonist squares of phase A and phase B through the thickness, such as to prohibit extended interconnections between bulk domains of same phase. The Fig. 4 **right** illustrates an outer and an inner views of two such matrix wrapping for adjacent fibers (the checkerboards can be similar at equal two fiber phase concentrations in the composite bulk).This formally hold for any type of linear properties of general anisotropy. The n-phase cases do not result straightforwardly.

2.2. The n-phase fully continuous and "fully discontinuous" cases

The effective properties for *directional n*-phase laminates (including the two-phase case), $C^{*L(\omega)}_{(n)}=(S^{*L(\omega)}_{(n)})^{-1}$, can be exactly obtained from considering stress and strain jump conditions at each planar interface between layers of different phases, without any reference to Green operators (Postma, 1955; Walpole, 1981; El Omri et al., 2000). They read:

$$\begin{aligned}
C^{*L(\omega)}_{c'd'a'b'\,(n)} &= \left.\left\langle (C_{c'd'a'b'})^{-1}\right\rangle^{-1}\right|^{\omega}_{(n)}=\overline{\left\langle \overline{C_{c'd'a'b'}}\right\rangle}^{\omega}_{(n)};\ C^{*L(\omega)}_{abc'd'\,(n)}\\
&= C^{*L(\omega)t}_{c'd'ab\ \ (n)}=\left\langle C_{abp'q'}\overline{C_{p'q'a'b'}}\right\rangle^{\omega}_{(n)}\overline{\left\langle\overline{C_{a'b'c'd'}}\right\rangle}^{\omega}_{(n)};\ C^{*L(\omega)}_{abcd\ \ (n)}\\
&= \left\langle C_{abcd}-C_{abc'd'}\overline{C_{c'd'a'b'}}C_{a'b'cd}\right\rangle^{\omega}_{(n)}\\
&\quad+\left\langle C_{abc'd'}\overline{C_{c'd'p'q'}}\right\rangle^{\omega}_{(n)}\overline{\left\langle\overline{C_{p'q'r's'}}\right\rangle}^{\omega}_{(n)}\left\langle\overline{C_{r's'a'b'}}C_{a'b'cd}\right\rangle^{\omega}_{(n)}.
\end{aligned} \tag{12}$$

Thanks to Eq. (12) there is an exactly known solution for $\boldsymbol{C}^{*L(\omega)}_{(n)}=(\boldsymbol{S}^{*L(\omega)}_{(n)})^{-1}$ and it is then always possible to obtain the $mLS(n)$ and $cLS(n)$ estimates for any n phases from the averaging definitions given in Eq. (10) for two phases, that now becomes for n phases:

$$\boldsymbol{C}^{*mLS}_{(n)}=\int_\Omega \psi_{Vell}(\omega)\boldsymbol{C}^{*L(\omega)}_{(n)}d\omega\ ;$$
$$\boldsymbol{C}^{*cLS}_{(n)}=\overline{\boldsymbol{S}^{*cLS}_{(n)}}=\left(\int_\Omega \psi_{Vell}(\omega)\boldsymbol{S}^{*L(\omega)}_{(n)}d\omega\right)^{-1}. \tag{13}$$

- Full n-continuity: Unfortunately but expectedly, the simpler relations of Eq. (8) obtained for the two-phase cases do not extend in some comparably simple manner for larger phase numbers, for the exact solution for n-phase laminates do not take a form generalizing the two-phase laminate case of Eq. (9). However, $\forall n$ the solution of first (resp. second) of Eq. (13) is still expected to obey the general form of Eq. (5) for some $n+1^{th}$ reference matrix of infinitesimal concentration which has moduli $\boldsymbol{C}_n{}^o$ say (resp. compliances $\boldsymbol{S}_n{}^o$) to be identified. This matrix is the solution of the inverse problem of solving Eq. (5) (resp. its inverse compliance form) for $\boldsymbol{C}_n{}^o$ (resp. $\boldsymbol{S}_n{}^o$) when $\boldsymbol{C}^{eff(m*)}_{(i,j,\ldots,n)\subset m^o}=\boldsymbol{C}^{*mLS}_{(n)}$ (resp. $=\boldsymbol{C}^{*cLS}_{(n)}$) from Eq. (13) and using one of the Eq. (12). This provides an implicit equation that can be solved similarly to the *SC* implicit scheme (Ricotti et al., 2006). Since the matrix concentration is infinitesimal, appropriate iterative schemes need be set up to reach an accurate solution for $\boldsymbol{C}_n{}^o$ (resp. $\boldsymbol{S}_n{}^o$), out of the present purpose.

Although no simple generalization of the two-phase case was found, it was verified from analytically solving a few particular n-phase cases (Franciosi and Charles, 2016) that the properties of the reference matrix still were those of a combination of the n assembled phases and that they were varying oppositely to the corresponding properties of the bulk of the composite.

Fortunately, it is not necessary to have an explicit form of the property tensor of this reference matrix in order to obtain the $\boldsymbol{C}^{*mLS}_{(n)}$ (or $\boldsymbol{C}^{*cLS}_{(n)}$) estimate when all n phases are co-continuous (or co-discontinuous), since the use of Eq. (12) in Eq. (13) does not call for these matrix properties.

-Full n-discontinuity: The realization of "fully co-discontinuous" structures corresponding to the *cLS*(n) estimate, say assemblages of phases capable to prevent any of the phases, even when in high concentration, from connecting into a through-sample spanning cluster is more questioning for the interpretation of phase co-discontinuity is still to be clarified. According to (Scriven, 1976) for examples of such structures, "possibilities are held to be blobs of one composition dispersed in another, or tubules of one threading the other, or lamellae of one alternating with the other". As for the two-phase composites (Fig. 4 right), the proposed idea in (Franciosi and El Omri, 2011) and in (Franciosi, 2012b; 2013) for respectively isotropic and transversally isotropic symmetry is in duality with the co-continuous case. Wrap the phase grains into an infinitesimal coating matrix layer with a phase arrangement such as to prohibit long distance chains of connections between grains of a same phase. As for all co-continuous phases, there is no extension of the reference matrix definition from the two-phase discontinuous case to the one for a general composite with all co-discontinuous n phases. But all what was observed and obtained for the *mLS*(n) estimate holds in dual manner for the *cLS*(n) estimate provided the appropriate substitutions of the involved quantities. The revisited interpretation of this *cLS*(n) estimate to be discussed in Section 3 highlights how the related structures look like.

2.3. The p multi-continuous cases among n>p>1 phases

In the type 4 arrangement of n-phases (n>2) the number p of continuous ones ranges between 2 and $n-1\geq 2$, say $p\in(2, n-1\geq 2)$, the minimum value of $q=n-p$ being now 1, for there must be at least one discontinuous (embedded) phase. In order to write a property estimate for this material in the form of Eq. (5), the existence of an appropriate $n+1^{th}$ infinitesimal reference matrix phase with properties $\boldsymbol{C}_n^{p^o}$ say was questioned in (Franciosi and Charles, 2016). This matrix has to satisfy i) the co-continuity of the p infinite phases and ii) the embedded nature of the $n-p$ remaining ones and this for any (and down to all zero) concentrations of these embedded phases.

The second condition is fulfilled if none of the q embedded phases in the bulk belongs to the constitutive phases of the reference matrix, for no continuity of any of these q phases is then made possible (as is the case for a q-phase reinforced single-phased matrix A, the reference matrix of which is the phase A itself). For the first condition, the study of the fully co-continuous arrangement type 2 has provided a characteristic matrix for n co-continuous phases. Thus, one candidate for the reference matrix defined by properties $\boldsymbol{C}_n^{p^o}$ is the matrix with properties $\boldsymbol{C}_p^o$, say the one for the fully continuous p-composite in same p-phase *relative* proportions as in the n-phase composite, into which q discontinuous phases are additionally embedded.

Thus, the matrix that characterizes the *mLS*(p) estimate fulfills the requests for multi-continuity of p phases among n. It is noteworthy that this same $\boldsymbol{C}_p^o$ matrix for a fixed p-continuous phase assemblage then holds for any q embedded phase number additional to p and in any relative concentrations in addition to those of the p phases. This means $\boldsymbol{C}_n^{p^o}=\boldsymbol{C}_p^o$ $\forall n\geq p$, including the limit of all null concentrations of the included q phases at which $n=p$ and $\boldsymbol{C}_n^{p^o}=\boldsymbol{C}_p^{p^o}=\boldsymbol{C}_p^o$. This conversely implies that any q number of phases added to a fully continuous p-set without contributing to the reference matrix will be necessarily an embedded q-set of phases. Since the $\boldsymbol{C}_p^o$ matrix is characteristic of the *mLS*(p) scheme with effective properties written as $\boldsymbol{C}^{*mLS}_{(p)}$, we denote $\boldsymbol{C}^{*mLS}_{(p,n)}$ the effective properties for that multi-continuous (p, n) phase arrangement type, $\forall n\geq p$. This property estimate reads according to Eq. (5):

$$\boldsymbol{C}^{*mLS}_{(p,n)}=\boldsymbol{C}_p{}^o-\left(\left(\sum_{i=1}^{n}f_i\left(\left(\boldsymbol{C}_p{}^o-\boldsymbol{C}_i\right)^{-1}-\boldsymbol{t}^{Vell}_{\boldsymbol{C}_p{}^o}\right)^{-1}\right)^{-1}+\boldsymbol{t}^{Vell}_{\boldsymbol{C}_p{}^o}\right)^{-1};$$

$$\sum_{i=1}^{n}f_i=1-f_{m_{p^o}}\approx 1, \tag{14a}$$

with Eq. (5) also fixing $\boldsymbol{C}_p{}^o$ from inversely solving, using $\boldsymbol{C}^{*L(\omega)}_{(i,j,\ldots,p)}$ from Eq. (12) in Eq. (13):

$$\boldsymbol{C}^{*mLS}_{(p)}=\int_\Omega\psi_{Vell}(\omega)\boldsymbol{C}^{*L(\omega)}_{(i,j,\ldots,p)}d\omega$$

$$=\boldsymbol{C}_p{}^o-\left(\left(\sum_{i=1}^{p}F_i\left(\left(\boldsymbol{C}_p{}^o-\boldsymbol{C}_i\right)^{-1}-\boldsymbol{t}^{Vell}_{\boldsymbol{C}_p{}^o}\right)^{-1}\right)^{-1}+\boldsymbol{t}^{Vell}_{\boldsymbol{C}_p{}^o}\right)^{-1};$$

$$\sum_{i=1}^{n}F_i\approx 1. \tag{14b}$$

In the simplest case of two co-continuous phases A, B with q $(=n-2)$ embedded ones D^i down to zero concentration of all of them, in *both* A and B (D^i can possibly be found either fully in phase A or in phase B and partly in both when along the A/B interface), the infinitesimal reference matrix has properties $\boldsymbol{C}_2{}^o=\{\boldsymbol{C}\}_{A,B}$. Note that if one of the D^i phases becomes continuous together with phases A and B, the reference matrix will be modified from $\boldsymbol{C}_2{}^o=\{\boldsymbol{C}\}_{A,B}$ to $\boldsymbol{C}_3{}^o$. If all the $(n-2)$ included phases become co-continuous with A and B, the reference matrix will change to $\boldsymbol{C}_n{}^o$ and conversely. Consistently, if one of the two co-continuous phases (B say) totally vanishes too (in addition to all the D^i phases), the reference infinitesimal matrix becomes $\boldsymbol{C}_1{}^o=\boldsymbol{C}_A$, that is a layer of the homogeneous phase A wrapping grains of phase A and grains of the phases D^i. This corresponds to the single continuous phase A embedding all other ones with properties obeying a *HS* estimate. Incidentally, this means that for $p=1$ one can write $\boldsymbol{C}^{*mLS}_{(1=A,n)}=\boldsymbol{C}^{*HSA}_{(n)}$, with the matrix phase $A\in(1,n)$, so making a formal place for the *HS* estimates in the $\boldsymbol{C}^{*mLS}_{(p,n)}$ series.

In summary, $\forall n$, for $p=2$, a $\boldsymbol{C}^{*mLS}_{(2,n)}$ stiffness tensor estimate can be explicitly obtained from Eq. (5) with using as $n+1^{th}$ reference matrix the medium with stiffness properties $\boldsymbol{C}_2{}^o=\{\boldsymbol{C}\}_{A,B}$, while

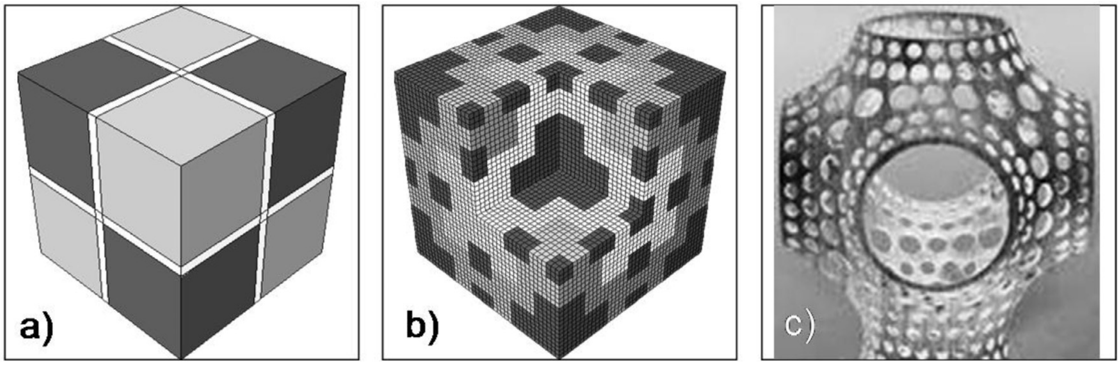

Fig. 5. (a) Two embedded phases in one continuous matrix; (b) 1/8th of cubic cell for a 3-phase composite structures with one embedded phase in two co-continuous ones from Fig. 3d (Franciosi and Charles, 2016); (c) a still tricontinuous composite with holes in the interfacial phase.

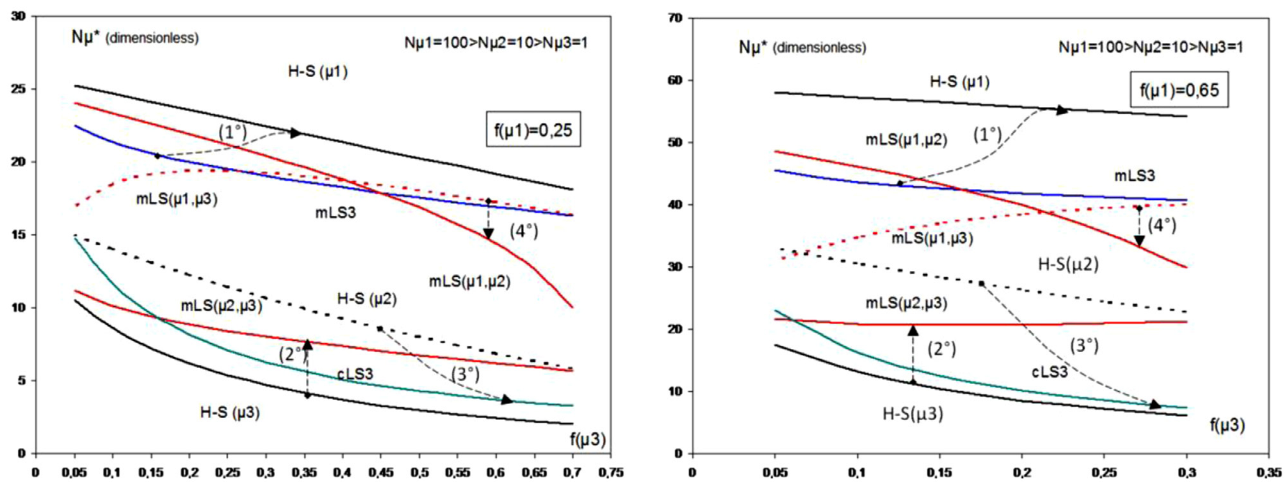

Fig. 6. The 8 estimates for the elementary arrangements of 3 isotropic and incompressible elastic phases at 25% (left) and 65% (right) of the stiffest phase 1 (modified from Franciosi and Charles, 2016) showing 4 transition examples in between pairs.

for $p > 2$ one must first solving Eq. (14b) for $\boldsymbol{C}_p{}^o$ prior to solve Eq. (14a) for $\boldsymbol{C}^{*mLS}_{(p,n)}$, this inverse problem for obtaining $\boldsymbol{C}_p{}^o$ needing to first solve for $\boldsymbol{C}^{*mLS}_{(p)}$ in using Eq. (13) left. Some particular (isotropic dielectric-like or elastic-like) cases have been shown (Franciosi and Charles, 2016) to have an explicit $\boldsymbol{C}_p{}^o$ solution up to $p = 5$, $\forall n$, what represents a substantial quantity of situations.

2.4. Continuous versus discontinuous phase status changes and percolation-like transitions

As a new complement to what mainly is up to that point a summary of earlier results, it is noteworthy in the proposed formula for the $\boldsymbol{C}^{*mLS}_{(p,n)}$ estimate (for any values of p and $n>p$) that any time one (or several) of the assembled phases change(s) from discontinuous to continuous or conversely, at fixed n value and without changes in the phase concentrations, there is an expected sharp discontinuity in the corresponding effective properties which "jump" or "drop" from one phase arrangement description and estimate to another in the $A(n)$ set. When the change is smooth with phase concentration changes, it is a percolation-like process, with a gradual transition over some concentration range similar to the documented ones for two-phase structures. We exemplify this in making use of the three-phase situation examined in (Franciosi and Charles, 2016) for which the four estimate types result in 8 estimate "branches" according to Fig. 1 and Eq. (1). The two cases of tri-continuous (type 2) and "tri-discontinuous" (type 3) assemblages correspond to the cubic cells exemplified in Fig. 3d and f respectively. The corresponding cells for the type 1 and type 4 assemblages are reported on Fig. 5, which shows (5a) one continuous phase separating disconnected cubes of the two other phases, the classical inclusion-reinforced matrix structure, and (5b) a bicontinuous arrangement of two phases (obtained from the tri-continuous structure of Fig. 3d) embedding a third one (which occupies the cube centre and corners on the Figure) as finite domains located either along their interface or in their bulks.

The Fig. 5c recalls that holes in a continuous layer are not enough to make it disconnected (the two different phases occupying each subspace are at direct contact in the interface holes). This structure changes from tri-continuous to 3-phase bicontinuous when the interface becomes made of disconnected pieces. A degrading joint or weld between two materials may correspond to such a tri-continuous to bi-continuous arrangement change (regardless of additional voids or cracks).

Jumps, drops and smooth, percolation-like, transitions in terms of property estimate changes are exemplified in Fig. 6 which show the 8 estimate "branches" taken from (Franciosi and Charles, 2016) for the unique effective shear modulus for 3 isotropic and incompressible (I-I) elastic phases, at two fixed values of the median phase concentration of 25% (left) and 65% (right). High "stiffness" contrasts (μ1=100>μ2=10>μ3=1) are considered. On the borrowed plots have been drawn four same examples of sharp (vertical) and smooth (wavy oblique) transition paths between shear estimate pairs:

(1) the upward property transition from the $mLS(3)$ - tri-continuous - estimate to the $HS(+)$ upper bound (continuous stiffest phase 1) would result from (gradual) continuity loss of both phases 2 and 3. In order to cross and pass above the $mLS(\mu1,\mu2)$ - bi-continuous - estimate, phase 2 must become discontinuous together with phase 3 while only phase 3 needs be discontinuous otherwise;
(2) the upward "jump" from the $HS(-)$ lower bound (softest phase 3) to the $mLS(\mu2,\mu3)$ estimate would result from a sharp discontinuous to continuous transition of the median phase 2 in addition to the soft phase 3, what increases the effective property;
(3) the downward transition from the median $HS(\mu2)$ estimate to the $cLS(3)$ "tri-discontinuous" estimate by gradual continuity loss of the median phase 2 what decreases the effective property;
(4) the downward "drop" from the $mLS(\mu1,\mu3)$ estimate to the $mLS(\mu1,\mu2)$ one by (dis)continuity interchange of phases 2 (median) and 3 (softest) at constant stiffest phase 1 vol fraction. In this not obvious transition, the effective property is decreased when the median phase 2 (in low concentration) becomes continuous at the place of the softest one 3 (in high concentration). At low volume fraction of the soft phase 3 (left hand sides of both figures), the drop becomes a jump.

Another particular example mentioned in (Franciosi, 2012b) is worthy to be recalled: if under some loading, a sample spanning continuous 3D network of fractures appears in a (homogenous or not) undamaged material, the mLS estimate predicts a finite drop of the effective stiffness from infinitesimal crack opening or void fraction.

All such transitions (by changes of phase "connectedness" status) occur in various physical, metallurgical, mechanical processes, as during metal solidification/fusion (Limodin et al., 2007; Liang et al., 2008; Pavot et al., 2015), phase precipitation/dissolution in alloys (Mac Cue et al., 2015) or transition in polymers (Veenstra et al., 2000) according to various extrinsic condition changes. They also occur during metallic powder compaction process (Poquillon et al., 2002; Martin and Bouvard, 2006; Mazaheri et al., 2009) or metal fragmentation under (blowing or pressure) load. Reciprocal events correspond with the converse continuous to discontinuous transitions. All are examples of application fields for the knowledge of effective properties corresponding to all elementary phase arrangements in n between which transitions can occur. The case of a porous material with a varying porosity status will be examined in section 5.

A formally important last case is the transition from one to zero matrix phase for which the clarification given in next Section 3 on the understanding of "co-discontinuity" will come in support to the qualitative analysis here performed. Two distinct possibilities correspond to this transition from a $HS(n)$ estimate ($p=1$, $\forall n \geq 2$). The first one corresponds to a continuity loss of the matrix phase (A say) which remains present in the assemblage ($p=1 \to p-1=0$ at $n \geq 2$), and the second possibility corresponds with the limit of the matrix decreasing concentration down to zero, the composite structure changing then from n phases to $n-1$ ones ($p=1 \to p-1=0$ and $n \geq 2 \to n-1 \geq 1$). In both possibilities, excepted if $n-1=1$ in the second one (a two-phase $B\subset A$ composite that becomes single - homogeneously, say continuously - phased B), such a transition would yield a n-phase composite with none of the phases being continuous, if none of the discontinuous phase takes over to become continuous in turn. This corresponds to the so far called "fully co-discontinuous" situation associated with the $cLS(n)$ estimate type. But this also amounts to giving to phase co-discontinuity the meaning of simultaneous disconnectedness for all the assembled phases, what is impossible in strict topological sense. Among the $q=n-1$ phases which were discontinuous when embedded within the continuous A one, all cannot remain discontinuous if A ceases be continuous or vanishes. A*t least one* of these q phases is expected to now take the status of being continuous to fill the new open gaps between the disconnected (or vanished) elements of phase A. It is also noteworthy that these transitions from one to zero continuous phase would amount to writing for $p=0$, $\forall n \geq 2$ $\boldsymbol{C}^{*mLS}_{(0,n)} = \boldsymbol{C}^{*cLS}_{(n)}$ such that, with $\boldsymbol{C}^{*mLS}_{(n)} = \boldsymbol{C}^{*mLS}_{(p=n,n)}$ and since for $p=1$, $\forall n \geq 2$ we arrived at $\boldsymbol{C}^{*mLS}_{(1=A,n)} = \boldsymbol{C}^{*HSA}_{(n)}$, the four arrangement types would have found a place in the same type 4 series. But from the topological arguments, the identity $\boldsymbol{C}^{*mLS}_{(0,n)} = \boldsymbol{C}^{*cLS}_{(n)}$ is clearly *not* correct. Hence, the $cLS(n)$ estimate type does not correspond to full co-discontinuity in the full co-disconnectedness sense and the mLS/cLS duality is still to be clarified. In relation to what precedes, even if a multi continuous $(p,\ n)$ composite structure of p co-continuous phases among n, with $q=n-p$, is also a multi discontinuous $(q,\ n)$ one, this does not correspond to an identified duality of the $\boldsymbol{C}^{*mLS}_{(p,n)}$ estimate with some $\boldsymbol{C}^{*cLS}_{(q,n)} = (\boldsymbol{S}^{*cLS}_{(q,n)})^{-1}$ one in the sense of the mLS/cLS duality. An obvious dual of a multi-continuous $(p,\ n)$ structure is a multi-continuous $(q,\ n)$ structure, with p and q permuted phase types with effective properties $\boldsymbol{C}^{*mLS}_{(q,n)}$, of the same type as $\boldsymbol{C}^{*mLS}_{(p,n)}$, using in Eq. (12) the matrix $\boldsymbol{C}_q{}^o$ for the q continuous phases in appropriate concentrations. A dual estimate to $\boldsymbol{C}^{*mLS}_{(p,n)}$ taking the form $\boldsymbol{C}^{*cLS}_{(q,n)} = (\boldsymbol{S}^{*cLS}_{(q,n)})^{-1}$has no interpretation so far of the structure type it may represent, if any.

The next section introduces the $mLS(n)$ and $cLS(n)$ estimates in a clarifying way regarding the interpretation of the underlying phase arrangements. It incidentally points a possible understanding for the $\boldsymbol{C}^{*cLS}_{(q,n)} = (\boldsymbol{S}^{*cLS}_{(q,n)})^{-1}$ estimate type, to be let in purpose of further developments.

3. Maximal and minimal p-phase co-continuity in composites

We here first propose a proof that composites having all their phases being fully co-continuous as defined, must have effective properties taking the form of the stiffness moduli Laminate System (mLS) scheme recalled in Section 2, Eq. (10) left for $n=2$ and Eq. (13) left for $n>2$. The proof is first given for $n=2$, and is next generalized to p-phase co-continuous materials, without or with q embedded phases. From this first insight, it can then be proven that *both* linear averaging of p-laminate stiffness moduli and linear averaging of p-laminate compliance moduli correspond to co-continuity of the p phases, the difference being that stiffness linear averaging corresponds to maximal (strong) co-continuity while compliance linear averaging corresponds to minimal (weak) co-continuity. Minimal co-continuity of p phases is finally shown to be the best representative of their topologically impossible full co-disconnectedness.

3.1. Multiple continuity of phases implies laminate green operators

The main property between laminate operators can be written for any two (A,B) media:

$$\boldsymbol{t}_A^{L(\omega)} - \boldsymbol{t}_B^{L(\omega)} = \boldsymbol{t}_A^{L(\omega)} : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \boldsymbol{t}_B^{L(\omega)}$$
$$\Leftrightarrow \boldsymbol{t'}_A^{L(\omega)} - \boldsymbol{t'}_B^{L(\omega)} = \boldsymbol{t'}_A^{L(\omega)} : (\boldsymbol{S}_B - \boldsymbol{S}_A) : \boldsymbol{t'}_B^{L(\omega)}. \tag{15}$$

Within the here used homogenization framework from which effective properties of n-phase materials can be characterized by Eq. (5), the specific effective (stiffness-like or compliance-like) properties of n-phase laminates can be expressed equivalently from any choice of one of the n constitutive phases (moduli $\boldsymbol{C}_i = \boldsymbol{S}_i^{-1}$) as reference matrix in finite concentration $1 > f_i \geq 0$, say:

$$\boldsymbol{C}_{(n)}^{*L(\omega)} = \boldsymbol{C}^r - \left(\left(\sum_{\substack{i=1 \\ i \neq r}}^{n} f_i \left((\boldsymbol{C}^r - \boldsymbol{C}_i)^{-1} - \boldsymbol{t}_{\boldsymbol{C}^r}^{L(\omega)} \right)^{-1} \right)^{-1} + \boldsymbol{t}_{\boldsymbol{C}^r}^{L(\omega)} \right)^{-1}, \quad \forall r \in \{1, n\}, \tag{16a}$$

$$\boldsymbol{S}_{(n)}^{*L(\omega)} = \boldsymbol{S}^r - \left(\left(\sum_{\substack{i=1 \\ i \neq r}}^{n} f_i \left((\boldsymbol{S}^r - \boldsymbol{S}_i)^{-1} - \boldsymbol{t}'^{L(\omega)}_{\boldsymbol{S}^r} \right)^{-1} \right)^{-1} + \boldsymbol{t}'^{L(\omega)}_{\boldsymbol{S}^r} \right)^{-1}, \quad \forall r \in \{1, n\}. \tag{16b}$$

Eq. (16) hold - in addition to Eq. (12) - thanks to the laminate property of Eq. (15) which is the sufficient condition. We here demonstrate (with details in Appendix A) this sufficient condition to also be the necessary one. That is if a n-phase medium has effective properties identically estimated by Eq. (5) for several (p say) choices of the matrix phase among the n ones, then all involved operators must fulfill the laminate operator property between all phase pairs among p.

Considering first two-phase [A, B] bi-continuous composites ($p = n = 2$), the possibility of choosing either phase A or phase B as the matrix, yields to assuming the equality:

$$\begin{aligned} \boldsymbol{C}^*_{A \subset B} &= \boldsymbol{C}_B - f_A \left((\boldsymbol{C}_B - \boldsymbol{C}_A)^{-1} - f_B \boldsymbol{t}_B^V \right)^{-1} \\ &= \boldsymbol{C}^*_{B \subset A} = \boldsymbol{C}_A - f_B \left((\boldsymbol{C}_A - \boldsymbol{C}_B)^{-1} - f_A \boldsymbol{t}_A^V \right)^{-1}. \end{aligned} \tag{17}$$

Eq. (17) is shown (Appendix A) to assign Eq. (15) left and thus $\boldsymbol{t}_A^V = \boldsymbol{t}_A^{L(\omega)}$ (resp. B), $\forall \omega \in \Omega$.

Lets consider next 3-phase [A,B,D] composites with phases A and B being co-continuous while phase D is either co-continuous a well or discontinuous, that is embedded.

- If phase D is co-continuous with phases A and B, three conditions of co-continuity similar to Eq. (17) can be written. Assuming them to be simultaneously fulfilled amounts to writing each of them, for example between A and B phases (all involved operators are assumed identical for the composite is taken to satisfy a simple ellipsoidal spatial distribution symmetry of all 3 phases) as:

$$\left. \begin{aligned} \boldsymbol{C}^*_{(A,D) \subset B} &= \boldsymbol{C}_B - \left(\left(f_A \left((\boldsymbol{C}_B - \boldsymbol{C}_A)^{-1} - \boldsymbol{t}_B^V \right)^{-1} + f_D \left((\boldsymbol{C}_B - \boldsymbol{C}_D)^{-1} - \boldsymbol{t}_B^V \right)^{-1} \right)^{-1} + \boldsymbol{t}_B^V \right)^{-1} \\ = \boldsymbol{C}^*_{(B,D) \subset A} &= \boldsymbol{C}_A - \left(\left(f_B \left((\boldsymbol{C}_A - \boldsymbol{C}_B)^{-1} - \boldsymbol{t}_A^V \right)^{-1} + f_D \left((\boldsymbol{C}_A - \boldsymbol{C}_D)^{-1} - \boldsymbol{t}_A^V \right)^{-1} \right)^{-1} + \boldsymbol{t}_A^V \right)^{-1} \end{aligned} \right\}, \tag{18a}$$

$$\boldsymbol{C}^*_{(A,D) \subset B} = \boldsymbol{C}_B - \left(\overline{\langle \boldsymbol{T} \rangle_{(A,D) \subset B}} + \boldsymbol{t}_B^V \right)^{-1} = \boldsymbol{C}^*_{(B,D) \subset A} = \boldsymbol{C}_A - \left(\overline{\langle \boldsymbol{T} \rangle_{(B,D) \subset A}} + \boldsymbol{t}_A^V \right)^{-1} \Big\}. \tag{18b}$$

Eq. (18) yield after some simple manipulations (Appendix A2) and (I, D)⊂J written (J):

$$\begin{aligned} \boldsymbol{t}_A^V - \boldsymbol{t}_B^V &- \boldsymbol{t}_B^V : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \boldsymbol{t}_A^V \\ &= \overline{\langle \boldsymbol{T} \rangle_{(B)}} : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \overline{\langle \boldsymbol{T} \rangle_{(A)}} + \boldsymbol{t}_B^V : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \overline{\langle \boldsymbol{T} \rangle_{(A)}} \\ &\quad + \overline{\langle \boldsymbol{T} \rangle_{(B)}} : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \boldsymbol{t}_A^V + \overline{\langle \boldsymbol{T} \rangle_{(B)}} - \overline{\langle \boldsymbol{T} \rangle_{(A)}}. \end{aligned} \tag{19}$$

This being expected to hold down to a zero volume fraction of the third phase D, it must be consistent with the two-phase co-continuity of A and B and therefore the left hand side must be zero, with the implied conditions from the two-phase case, that only laminate operators fulfil this relation type. We are thus left with the additional condition (proved in Appendix A):

$$\begin{aligned} &\overline{\langle \boldsymbol{T} \rangle_{(B)}} : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \overline{\langle \boldsymbol{T} \rangle_{(A)}} \\ &\quad + \boldsymbol{t}_B^V : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \overline{\langle \boldsymbol{T} \rangle_{(A)}} \\ &\quad + \overline{\langle \boldsymbol{T} \rangle_{(B)}} : (\boldsymbol{C}_B - \boldsymbol{C}_A) : \boldsymbol{t}_A^V + \overline{\langle \boldsymbol{T} \rangle_{(B)}} - \overline{\langle \boldsymbol{T} \rangle_{(A)}} = \boldsymbol{0}, \end{aligned} \tag{20}$$

and the two complementary ones by circular *A, B, D* permutations for full tri-continuity.

Thus, when the three-phase (A,B,D) material is a laminate, that is when the three Green operators are a same laminate one, this set of equations is simultaneously fulfilled. Other possibilities are unexpected as far as the Green operator pair relation of Eq. (17) is only fulfilled for laminates.

- If phase D is an embedded one while phases A and B are co-continuous, there are no complementary equations to Eq. (20) which however assigns the operator pair $\boldsymbol{t}_A^V, \boldsymbol{t}_B^V$ to be laminate operators. There is no such assignment on the operator that represents the shape of the embedded phase D domains. Eq. (18 to 20) hold as well for D standing for several embedded phases D^i, all the terms possibly being included in the $\overline{\langle \boldsymbol{T} \rangle_{(A,(D_i)) \subset B}}$ notation type.

It results that beyond purely directional laminate structures, Eq. (17) (and the p-set ones similar to the Eq. (20) for $n > p = 2$) can only hold for effective stiffness properties that take the form of a linear combination (thus an arithmetic mean) of effective properties of directionally laminated structures, what correspond to Eq. (13) left, exotic cases excepted if any.

3.2. Both linear combinations of strain or stress green operators hold as well

For the dual situation a priori taken to correspond with all co-discontinuous (all co-disconnected) phases to be associated with an arithmetic average of laminate compliance properties over directions in space, there would not be any reasoning similar to the performed one for the stiffness average, for as far as none of the constitutive n phases can be taken as a continuous matrix, there would be no initial relation similar to Eq. (17) to start from. However, the laminate property recalled in Eq. (15) holds as well when it is inversely written in terms of the dual compliance moduli and consequently the compliance counterpart of Eq. (17) also holds equivalently with regard to the stress Green operator (Eq. (15) right). It results of this stiffness/compliance equivalency for *directional* laminates that all composites whose effective properties can be written as an arithmetic average of the compliances of directional laminates (according to Eq. (13) right) also obey the necessary condition for phase co-continuity. It corresponds to an harmonic average of the stiffness moduli, say the weakest average in contrast with the arithmetic average which is the strongest one. As a consequence, the duality of the two (arithmetic and harmonic) stiffness moduli averaging does not correspond to fully continuous (all co-continuous) versus fully discontinuous (all co-discontinuous) n-phase structures but to maximally stiff (strong) versus maximally compliant, minimally stiff, (weak) all n co-continuous phase structures. This is consistent with the conclusions arrived at in Section 2.4 from considering the consequences of a continuous to discontinuous transition of a single matrix phase in a composite assemblage. Some additional support will be given from the last section analyses.

3.3. Minimal phase co-continuity as the closest to impossible full co-disconnectedness

From a minimal 3D continuity definition for one phase as being a sample spanning cluster made of point- (or line-) connected finite domains of that phase into a 3D network, 3D discontinuity of one phase will correspond to 3D disconnections of this minimal network. Hence, *full co-discontinuity of all phases in the co-disconnectedness sense in an assemblage is impossible* for one phase at least needs be available to fill the gaps in between the disconnected ones. Full disconnectedness can only at the best be represented by the arrangement of minimal co-continuity, say all phases being clusters of point-connected (or line-connected, say null area connections) domains, that is all the phases in their weakest continuity state at once. Thus minimal co-continuity does not identify to full co-discontinuity but can be seen as the closest-to-it phase arrangement. This refined understanding of the initially proposed interpretation of the co-continuity versus co-discontinuity duality corresponding to the $mLS(n)$ and $cLS(n)$ estimates facilitates how to figure out structures corresponding to minimal co-continuity and to the $cLS(n)$ estimate type. The example of "tri-discontinuous structure" given in Fig. 3f well corresponds to minimally co-connected phases.

4. General n-phase composites in terms of the A(n) elementary n-phase arrangements

For a general n-phase composite whose (one-level) microstructure does not match well enough with any of the elementary arrangements, some "mixture" of those is likely representative. Such a mixture can also be useful in composites where the continuous/discontinuous status of certain phases is not well known, as for the porosity in the experimental example from (Torres et al., 2012) that will be examined next. We consider whether some $P(N)$ probability-weighted combination of the $N{=}A(n)$ arrangements can provide a microstructure description and a property estimate, still in the considered homogenization framework where effective properties result from Eq. (5) or equivalent ones. Treating this composite as being statistically homogeneous, each point $\boldsymbol{r}$ in it may be given such a probability $P(J)$ to match with the J elementary arrangements among the $N{=}A(n)$ ones, whether most of them be zero or not. Matching can a priori either be directly in terms of their effective properties or more basically in terms of their "microstructural characteristics" that we here understand in the sense of phase spatial arrangements as defined. With the needed key information being to determine an infinitesimal reference $n+1^{th}$ matrix, the question turns into whether one can describe the composite, from its $N{=}A(n)$ elementary arrangements, either in terms of their effective properties or in terms of their reference matrices, provided a set of $P(N)$ probabilities.

For such a general (one-level) n-phase composite of effective properties $\boldsymbol{C}^*=(\boldsymbol{S}^*)^{-1}$ and of $n+1^{th}$ reference matrix with properties $\boldsymbol{C}^o=(\boldsymbol{S}^o)^{-1}$, Eq. (5) can be rewritten equivalently as:

$$\left((\boldsymbol{C}^o-\boldsymbol{C}^*)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^o}\right)^{-1}=\sum_{i=1}^{n} f_i\left(\left(\boldsymbol{C}^o-\boldsymbol{C}^i\right)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^o}\right)^{-1}, \tag{21a}$$

$$\left((\boldsymbol{S}^o-\boldsymbol{S}^*)^{-1}-\boldsymbol{t}'^V_{\boldsymbol{S}^o}\right)^{-1}=\sum_{i=1}^{n} f_i\left(\left(\boldsymbol{S}^o-\boldsymbol{S}^i\right)^{-1}-\boldsymbol{t}'^V_{\boldsymbol{S}^o}\right)^{-1}. \tag{21b}$$

The equivalency between Eqs. (21a) and (21b) is straightforward once having noticed that any difference of the form $\overline{\Delta\boldsymbol{C}^*}=(\boldsymbol{C}^o-\boldsymbol{C}^*)^{-1}$ transforms into its dual form $\overline{\Delta\boldsymbol{S}^*}=(\boldsymbol{S}^o-\boldsymbol{S}^*)^{-1}$ as any strain Green operator $\boldsymbol{t}^V_{\boldsymbol{C}^o}$ transforms into its related stress one $\boldsymbol{t}'^V_{\boldsymbol{S}^o}$ (see in Section 2) say $\overline{\Delta\boldsymbol{S}^*}=\boldsymbol{C}^o-\boldsymbol{C}^o:\overline{\Delta\boldsymbol{C}^*}:\boldsymbol{C}^o$ or conversely $\overline{\Delta\boldsymbol{C}^*}=\boldsymbol{S}^o-\boldsymbol{S}^o:\overline{\Delta\boldsymbol{S}^*}:\boldsymbol{S}^o$, such that:

$$\begin{aligned}\overline{\Delta\boldsymbol{S}^*}-\boldsymbol{t}'^V_{\boldsymbol{S}^o}&=\boldsymbol{C}^o-\boldsymbol{C}^o:\overline{\Delta\boldsymbol{C}^*}:\boldsymbol{C}^o-\left(\boldsymbol{C}^o-\boldsymbol{C}^o:\boldsymbol{t}^V_{\boldsymbol{C}^o}:\boldsymbol{C}^o\right)\\&=-\,\boldsymbol{C}^o:\left(\overline{\Delta\boldsymbol{C}^*}-\boldsymbol{t}^V_{\boldsymbol{C}^o}\right):\boldsymbol{C}^o.\end{aligned}$$

When neither $\boldsymbol{C}^*=(\boldsymbol{S}^*)^{-1}$nor $\boldsymbol{C}^o=(\boldsymbol{S}^o)^{-1}$ are known, the addressed question is essentially of formal interest as a method yielding first a $\boldsymbol{C}^o$ estimate and then a $\boldsymbol{C}^*$ one. It may also be helpful when only the composite effective properties are known but not the reference matrix for estimating which reference matrix characterizes the composite structure. It can be of practical interest to help determining missing connectedness characteristics of the constitutive phases. The here examined question is the relevancy of either one of the two options below (only expressed in stiffness form for room saving, thanks to the equivalency for the compliant form).

- The first option can be written, from the $\boldsymbol{C}^{*J}$ properties for each J of the $A(n)$ arrangements:

$$\left(\left(\boldsymbol{C}^{\widehat{o}}-\boldsymbol{C}^*\right)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^{\widehat{o}}}\right)^{-1}=\sum_{J=1}^{N=A^{(n)}} P_J\left(\left(\boldsymbol{C}^{\widehat{o}}-\boldsymbol{C}^{*J}\right)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^{\widehat{o}}}\right)^{-1}, \tag{22a}$$

with probabilities P_J that fix the composite properties $\boldsymbol{C}^*$ provided a reference matrix $\boldsymbol{C}^{\widehat{o}}=(\boldsymbol{S}^{\widehat{o}})^{-1}$ which is not necessarily identical to $\boldsymbol{C}^o$;

- The second option reads similarly, with the $\boldsymbol{C}^{oJ}$ being the properties of the reference matrices of the elementary arrangements that are supposed to define the composite reference matrix $\boldsymbol{C}^o$ for probabilities P_J and provided a $\boldsymbol{C}^{oo}=(\boldsymbol{S}^{oo})^{-1}$ $N+1^{th}$ reference "super matrix":

$$\left((\boldsymbol{C}^{oo}-\boldsymbol{C}^o)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^{oo}}\right)^{-1}=\sum_{J=1}^{N=A^{(n)}} P_J\left(\left(\boldsymbol{C}^{oo}-\boldsymbol{C}^{oJ}\right)^{-1}-\boldsymbol{t}^V_{\boldsymbol{C}^{oo}}\right)^{-1}. \tag{22b}$$

Lets a priori consider that all the N elementary arrangements possibly contribute, that is $P_J\geq 0,\ \forall J\in(1,\ N)$ in $\sum_{J=1}^{N=A^{(n)}} P_J=1$. In both options, substituting the n-phase initial composite (of $N{=}A(n){>}n$ arrangements) with an equivalent N-phase one for which the $A(N){=}A(A(n))>A(n)$ arrangement number rapidly increases appears as a wrong route unless some relevant assumption allows overcoming the difficulty of specifying which arrangement(s) needs be selected in $A(N)$.

An assumption which can be used for both options regardless of the number of non zero contribution among N is to consider the only one-level arrangement not contained in the $A(N){=}A(A(n))$ set, that is a disordered arrangement of the N elementary ones. If the SC estimate is assumed to hold in that case, it provides a null left hand side in both Eq. (22), as well as for the compliance corresponding forms. This assumption yields to assuming that any n-phase composite could be described, provided probabilities $P(N)$ be specified, from a *disordered* "mixture" of its N-arrangements, in terms of either the elementary effective properties (option 1) or the elementary reference matrices (option 2). Note that the "mixture" neither applies on stiffness tensors nor on compliance ones but on the "neutral" quantity defined in Eq. (21).

Given this $P(N)$ set, the SC solution for the "effective properties" first option, Eq. (22a), corresponds to setting $\boldsymbol{C}^{\widehat{o}}=\boldsymbol{C}^*$ (or $\boldsymbol{S}^{\widehat{o}}=\boldsymbol{S}^*$ in the equivalent compliance equation) in the right hand side. This provides a $\boldsymbol{C}^*$ effective stiffness tensor according to a direct SC scheme without any new access to the related reference matrix $\boldsymbol{C}^o$ than the already evoked inverse problem.

For the second "reference matrices" option from Eq. (22b), the SC solution corresponds to setting $\boldsymbol{C}^{oo}=\boldsymbol{C}^o$ (or $\boldsymbol{S}^{oo}=\boldsymbol{S}^o$ in the compliance equation) in the right hand side. This provides first (from a direct SC scheme) a reference matrix $\boldsymbol{C}^o$ for the composite of concern, of which it is next possible to obtain (also in direct manner) the composite $\boldsymbol{C}^*$ properties using Eq. (21).

Alternative simple assumptions to this disorder one do not easily come out: in the first option, it could be assumed for example, without a priori referring to any particular (SC or else) estimate type for the combination of elementary effective properties, that the same reference matrix should hold for both the n-phase composite and its N-arrangement representation, that is assuming $\widehat{\boldsymbol{C}^{o}}=\boldsymbol{C}^{o}$ in Eq. (22a). It would then be possible to formally obtain a solution for $\boldsymbol{C}^{o}$ in equaling Eqs. (21a) to (22a) what is, as the SC estimate, an inverse problem to solve which remains quite complicated in general and without an ensured solution existence, in contrast with a direct SC scheme to solve. There is no equivalent to this assumption for the second option.

No other simple enough assumptions have been found worthy of being here examined.

In order to check these two (indeed empirical) description proposals (options 1 and 2), a validation test needs be applied to some n-phase composite of well enough known microstructure in terms of the probability set $P(N)$ as well as of well enough known effective properties to perform comparisons. This is not easy to find out from literature data owing to the problem difficulties to solve even for $n=2$ and it is not easy to realize either, neither experimentally nor numerically. Prior to any experimental campaign and to comparison tries with numerically realized structures, both being quite complex to perform with the required accuracy in terms of structure control and property measurements, we report a check on a purely theoretical case. This case has allowed to establish the remarkable relation that we found worthy to be presented. The two proposed optional descriptions have been applied to a two-phase composite assumed to be disordered in the sense that its effective properties from Eq. (21) with $n=2$ are also assumed given by the SC estimate. This assumption amounts to assuming $\boldsymbol{C}^{o}=\boldsymbol{C}^{*}$ (or $\boldsymbol{S}^{o}=\boldsymbol{S}^{*}$) such that the left hand side of Eq. (21a) is zero together with the one of Eq. (22a) in option 1 or of Eq. (22b) in option 2. Both description options (1,2) in terms of the $A(2)$=4 phase elementary arrangements then read:

$$0=\sum_{i=1}^{n} f_i\left(\left(\boldsymbol{C}^{*}-\boldsymbol{C}^{i}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*}}\right)^{-1} \text{ with}$$
$$0=\sum_{J=1}^{N=A^{(n)}} P_J\left(\left(\boldsymbol{C}^{*}-\boldsymbol{C}^{*J}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*}}\right)^{-1}, \tag{23a}$$

$$0=\sum_{i=1}^{n} f_i\left(\left(\boldsymbol{C}^{*}-\boldsymbol{C}^{i}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*}}\right)^{-1} \text{ with}$$
$$0=\sum_{J=1}^{N=A^{(n)}} P_J\left(\left(\boldsymbol{C}^{*}-\boldsymbol{C}^{oJ}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*}}\right)^{-1}. \tag{23b}$$

From having examined both inverse problems of finding the $P(4)$ solution set, the first part of Section 5 reports (with help in Appendix B) the proof of the remarked relation that comes in support to the “reference matrix mixture" option (2) as well as to the validity of the two dual Laminate System schemes for estimating effective properties of “multi-continuous” composites.

This relation is that in the case of isotropic incompressible (I-I) two-phase elastic-like composites, or isotropic (I) dielectric-like ones, the probability set $P(4)$ which is solution of Eq. (23b) is independent of the phase (shear or conductivity) modulus contrast. This contrast independency is achieved because the properties *of the reference matrices* (*not* the effective properties) for the four elementary arrangements of two phases (A,B) fulfill a particular match in relation with those of the SC estimate. These four property estimates from the elementary two-phase arrangements are those of the two HSA and HSB estimates (the reference matrices of which are the phases A and B themselves, with stiffness tensors $\boldsymbol{C}_A$ and $\boldsymbol{C}_B$) plus the two $mLS(2)$ and $cLS(2)$ estimates from the dual Laminate System schemes (the reference matrices of which are the matrices with stiffness tensors $\{\boldsymbol{C}\}=\{\boldsymbol{C}\}_{[A,B]}$ and $\{\boldsymbol{S}\}^{-1}=(\{\boldsymbol{S}\}_{[A,B]})^{-1}$ respectively). As far as the $HSA=HS(2)_{B\subset A}$ and $HSB=HS(2)_{A\subset B}$ estimates well hold for the two arrangements of the reinforced-matrix type and that the $SC=SC(2)_{A,B}$ estimate well holds for a disordered arrangement, this contrast independency property would not hold if the $mLS=mLS(2)_{A,B}$ and $cLS=cLS(2)_{A,B}$ estimate pair was not representing the two cases of phase bi-continuity (maximal co-continuity) and “bi-discontinuity” (minimal co-continuity).

This contrast independency is in particular checked to not being fulfilled by the probability set $P(4)$ found to fulfill Eq. (23a) for the “effective property” description option (1). Such a contrast independency is unexpected in multiple moduli problems, a theoretical problem whose examination is beyond the present scope. Expectedly, the more a composite microstructure is characterized in the details, the more its effective properties have interdependencies. This is why, if not impossible, the finding of “simple" (one-level) n-phase mixtures to simultaneously match a given set of properties, is unlikely. In Section 5, one first establishes the contrast-independent one-modulus relation. We next present a direct application of the “matrix mixture method” on literature data for a porous Ti composite structure characterized in (Torres et al., 2012), the porous phase of which is partly connected and partly disconnected.

5. The “matrix mixture method”: a formal problem and one application example

The SC estimate $\boldsymbol{C}^{*SC}_{[AB]}$ for two-phase materials $[A, B]$ of general anisotropy is solution of:

$$\sum_{i=1}^{2} f_i\left(\left(\boldsymbol{C}^{*SC}_{[AB]}-\boldsymbol{C}^{i}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*SC}_{[AB]}}\right)^{-1}=f_A\left(\left(\boldsymbol{C}^{*SC}_{[AB]}-\boldsymbol{C}^{A}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*SC}_{[AB]}}\right)^{-1}$$
$$+f_B\left(\left(\boldsymbol{C}^{*SC}_{[AB]}-\boldsymbol{C}^{B}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*SC}_{[AB]}}\right)^{-1}=0, \tag{24}$$

as the particular case for $n=2$ of Eq. (22) and (23) left. In order to examines the two description options introduced in Section 4, the corresponding SC formula for the $A(2)=4$ elementary arrangements of the 2 phases, with either $\widehat{\boldsymbol{C}}^{J}=\boldsymbol{C}^{*J}$ or $\widehat{\boldsymbol{C}}^{J}=\boldsymbol{C}^{oJ}$ for option 1 (Eq. 22 right) or option 2 (eq. 23 right), with notations $\widehat{\boldsymbol{N}}^{X}_{[AB]}=((\boldsymbol{C}^{*SC}_{[AB]}-\widehat{\boldsymbol{C}}^{X}_{[AB]})^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*SC}_{[AB]}})^{-1}$ and $P_{CA}=P_{DA}=1-P_{CA}$ for the probability that phase A (resp. B) is continuous or discontinuous, can be written:

$$\sum_{J=1}^{4} P_J\left(\left(\boldsymbol{C}^{*SC}_{(4)}-\widehat{\boldsymbol{C}}^{J}\right)^{-1}-\boldsymbol{t}^{V}_{\boldsymbol{C}^{*SC}_{(4)}}\right)^{-1}=$$
$$\boldsymbol{0}=P_{CACB}\widehat{\boldsymbol{N}}^{mLS}_{[AB]}+P_{DADB}\widehat{\boldsymbol{N}}^{cLS}_{[AB]}+P_{CADB}\widehat{\boldsymbol{N}}^{HSA}_{[AB]}+P_{DACB}\widehat{\boldsymbol{N}}^{HSB}_{[AB]}, \tag{25a}$$

$$\boldsymbol{0}=P_{CA}P_{CB}\left(\widehat{\boldsymbol{N}}^{mLS}_{[AB]}-\widehat{\boldsymbol{N}}^{HSA}_{[AB]}-\widehat{\boldsymbol{N}}^{HSB}_{[AB]}+\widehat{\boldsymbol{N}}^{cLS}_{[AB]}\right)$$
$$+P_{CA}\left(\widehat{\boldsymbol{N}}^{HSA}_{[AB]}-\widehat{\boldsymbol{N}}^{cLS}_{[AB]}\right)+P_{CB}\left(\widehat{\boldsymbol{N}}^{HSB}_{[AB]}-\widehat{\boldsymbol{N}}^{cLS}_{[AB]}\right)+\widehat{\boldsymbol{N}}^{cLS}_{[AB]}. \tag{25b}$$

With unconditional probabilities $P_{CACB}=P_{CA}P_{CB}$, $P_{CADB}=P_{CA}(1-P_{CB})$, $P_{CBDA}=P_{CB}(1-P_{CA})$, $P_{DADB}=(1-P_{CA})(1-P_{CB})$ and writing $d\widehat{\boldsymbol{N}}^{X}_{[AB]}=\widehat{\boldsymbol{N}}^{X}_{[AB]}-\widehat{\boldsymbol{N}}^{cLS}_{[AB]}$, Eq. (25b) can be put under the form:

$$P_{CA}P_{CB}\left(d\widehat{\boldsymbol{N}}^{mLS}_{[AB]}-d\widehat{\boldsymbol{N}}^{HSA}_{[AB]}-d\widehat{\boldsymbol{N}}^{HSB}_{[AB]}\right)$$
$$+P_{CA}\left(d\widehat{\boldsymbol{N}}^{HSA}_{[AB]}\right)+P_{CB}\left(d\widehat{\boldsymbol{N}}^{HSB}_{[AB]}\right)=-\widehat{\boldsymbol{N}}^{cLS}_{[AB]}. \tag{26}$$

We address the simple situations of a single modulus to fix in finding the probability set $P(4)$.

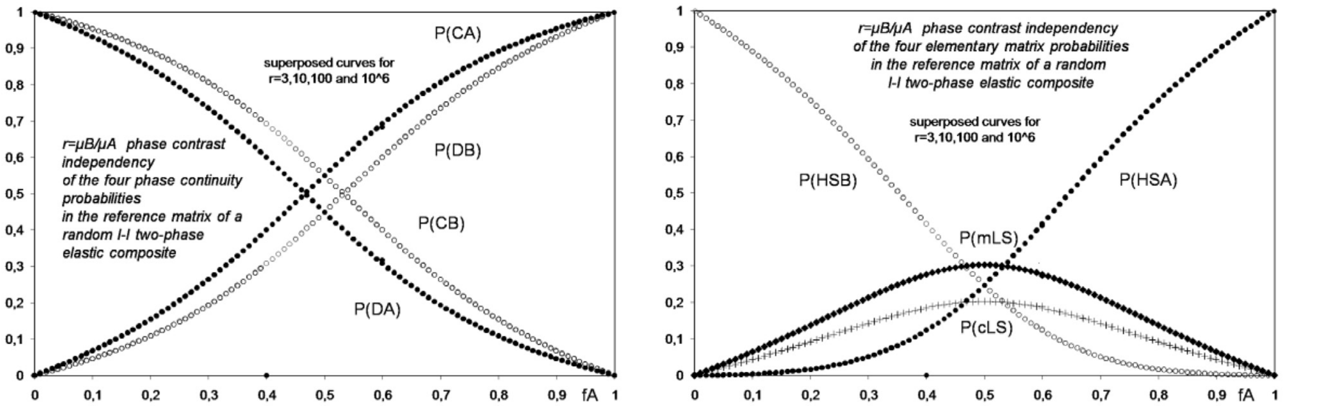

Fig. 7. The 4 (contrast independent) continuity and discontinuity probabilities of the two phases of a disordered two-phase I-I composite (left) and the related probabilities of each elementary reference matrix contributions in the composite reference matrix (right).

5.1. Single modulus (I-I) elastic or (I) dielectric problem(s)

Material phases with isotropic elastic properties have at most two independent (shear and compressibility "bulk") moduli and a single -shear - one when both phases are isotropic and incompressible, the I-I case. There is also a single modulus for dielectric-like isotropic properties. These moduli being tensor eigenvalues, Eq. (26) takes a scalar form for each, with only the Green operator term being different (Appendix B).

For cases with a single modulus, the two probabilities P_{CA},P_{CB} can be connected from assuming symmetry in an $A \overset{\rightarrow}{\underset{\leftarrow}{}} B$ phase interchange such that $P_{CA}(f_A = f) = P_{CB}(f_B = f) = P_{CB}(f_A = 1 - f)$. Thus, also considering the dual (harmonic) material $[B, A] =]A, B[$ with P_{CA},P_{CB} $\equiv P, P'$ the same equation as Eq. (26) for $f_A = f = 1 - f_B$ holds with permuted volume fractions such that $f_A = 1 - f$ and permuted probabilities $P, \overset{\rightarrow}{\underset{\leftarrow}{}} P'$. This symmetry assumption yields the system:

$$PP'\left(d\widehat{N}^{mLS}_{[AB]} - d\widehat{N}^{HSA}_{[AB]} - d\widehat{N}^{HSB}_{[AB]}\right) + P\left(d\widehat{N}^{HSA}_{[AB]}\right) + P'\left(d\widehat{N}^{HSB}_{[AB]}\right) = -\widehat{N}^{cLS}_{[AB]}. \tag{27a}$$

$$PP'\left(d\widehat{N}^{mLS}_{[AB[} - d\widehat{N}^{HSA}_{]AB[} - d\widehat{N}^{HSB}_{]AB[}\right) + P'\left(d\widehat{N}^{HSA}_{]AB[}\right) + P\left(d\widehat{N}^{HSB}_{]AB[}\right) = -\widehat{N}^{cLS}_{]AB]}, \tag{27b}$$

with scalar coefficients. Eliminating the PP' terms from the form:

$$\begin{cases} PP'[a] + P[b] + P'[d] = [k] \\ PP'[a] + P[d] + P'[b] = [k] \end{cases}, \tag{27c}$$

yields a relation between P and P' that reads $P' = \alpha + \beta P$, with scalar α and β coefficients. In Eq. (27), the quantities $\widehat{N}^X - \widehat{N}^Y$ read in terms of shear moduli:

$$\widehat{N}^X - \widehat{N}^Y = \frac{\mu^o - \widehat{\mu}^{*X}}{3\mu^o + 2\widehat{\mu}^{*X}} - \frac{\mu^o - \widehat{\mu}^{*Y}}{3\mu^o + 2\widehat{\mu}^{*Y}} = \frac{\mu^o\left(\widehat{\mu}^{*Y} - \widehat{\mu}^{*X}\right)}{\left(\left(1 - \frac{2}{5}\right)\mu^o + \frac{2}{5}\widehat{\mu}^{*X}\right)\left(\left(1 - \frac{2}{5}\right)\mu^o + \frac{2}{5}\widehat{\mu}^{*Y}\right)}, \tag{28}$$

with the coefficient 2/5 being characteristic of the involved Green operator term for this I-I elastic shear case. This term being defined on the disordered material of reference with effective shear modulus μ^o that results from a *SC* scheme, the resolution of Eq. (27) in terms of existing probabilities (P, P') is dependent on the expressions for the five $\mu^o = \mu^{*SC}$, $\widehat{\mu}^{HSA}$, $\widehat{\mu}^{HSB}$,$\widehat{\mu}^{*mLS}$,$\widehat{\mu}^{*mLS}$ moduli involved, with either $\widehat{\mu}^* = \mu^*$ (effective properties) in option 1 or $\widehat{\mu}^* = \mu^o$ (reference matrices) in option 2 (for the *SC* estimate, $\mu^o = \mu^{OSC} = \mu^{*SC}$).

The analytical resolution (sketched in Appendix B) of this I-I case establishes the contrast independency of the solution which corresponds to:

$$\beta = \frac{3f_B - 2f_A}{3f_A - 2f_B};\ \alpha = \frac{2(f_A - f_B)}{3f_A - 2f_B} = -f_A\beta + f_B \text{ with } \frac{\alpha}{\beta} = -f_A + \frac{f_B}{\beta} = \frac{2(f_A - f_B)}{3f_B - 2f_A}. \tag{29}$$

The resulting solution for the $P = P_{CA}$,$P' = P_{CB}$ probabilities reads:

$$P = -\frac{1}{2}\left(\frac{4f_A - 5}{3 - 5f_A} \pm \sqrt{\left(\frac{4f_A - 5}{3 - 5f_A}\right)^2 - 4\frac{3f_A}{3 - 5f_A}\left(\frac{3 - 5f_A}{3 - 5f_A}\right)}\right)$$

$$\text{for } f_A \begin{cases} < 3/5 \\ > 3/5 \end{cases},\ P' = \alpha + \beta P. \tag{30}$$

The four probabilities P_{CA},P_{CB},P_{DA},P_{DB} as well as the four probabilities (and proportions) of the elementary references matrices P_{CACB},P_{CADB},P_{DACB},P_{DADB} in the reference matrix of two-phase I-I disordered elastic composites are plotted in Fig. 7 **left** and **right** respectively, for several numerically checked moduli contrast values superposed with the analytical solution from Eqs. (29,30). It is observed on Fig. 7 right that, i) as expected, the *HSA* (resp. *HSB*) matrix type becomes more and more dominant when the phase A (resp. B) becomes the densest one with the probabilities of all three other elementary arrangements going to zero; ii) the intermediate range combines the four matrix arrangements in more or less equal quantities (near 25%) with a slightly larger "bi-continuous" *mLS* part at 30% and a (only slightly) smaller "bi-discontinuous" *cLS* part at 20%, far from being a domain where phase co-continuity dominates.

Hence, contrast independency clearly results from the use of the *mLS*(2) and *cLS*(2) estimates together with the three *HSA*, *HSB*, *SC* ones and it is lost when using other pairs of estimates instead of the *mLS*(2) and *cLS*(2) one. This contributes to validate these two *LS* estimates as representative of the properties of "bi-continuous" (maximally continuous) and "bi-discontinuous" (minimally continuous) two-phase structures first as well as their consistent *mLS*(*n*), *cLS*(*n*) and *mLS*(*p, n*) extensions for multi-continuous n-phase composites. In passing, if the latter extension well holds as an

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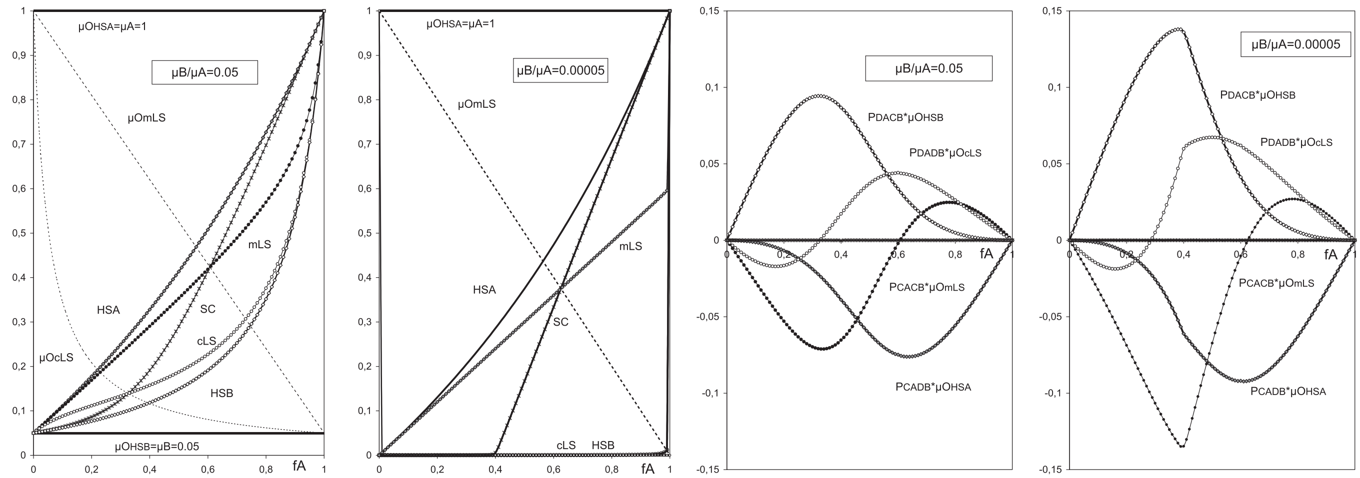

Fig. 8. From left to right; effective shear modulus and related reference matrix modulus for the five estimates of two-phase isotropic incompressible composites μA,μB at (i) moderate and (ii) high phase contrast and the corresponding (iii,iv) weighted four elementary matrix contributions to the reference matrix modulus of a disordered arrangement of the two-phases. The weights are the four functions of Fig. 7 right at any contrast.

estimate for $q = n - p$ embedded phases in maximally co-continuous p ones, the dual estimate $cLS(p,\ n)$ evoked in Section 2.4 as formally existing finds an interpretation as corresponding to minimally co-continuous p phases embedding the $q = n - p$ ones. This also reinforces the max/min co-continuity duality for the stiffness and compliance *LS* schemes.

Generalizing the I-I elastic case, one easily verifies from the Appendix details that if changing the Green operator coefficient 2/5 of the I-I shear modulus case into some $0 \le w \le 1$ value (e.g. in taking a different (as cuboidal) Green operator[10] or the one of a different one-modulus similar problem), the contrast-independent solution of Eqs. (29,30) still applies in substituting 2/5 with w (for isotropic dielectric-like moduli, $w = 1/3$[11]). The contrast-independent solution from the description option 2 thus is also shown morphology- and property-dependent.

This characteristic relation between five "elementary property estimates" for two-phase composites is illustrated in Fig. 8 for two shear moduli contrasts μB/μA= 0.05 and 0.00005. The two Fig. 8 left show the reference matrix and effective shear moduli pairs for the four phase arrangements of the two phases together with the pair for the composite which are superimposed since assumed given by the *SC* estimate. The 9 curves are well distinct at moderate contrast while at high contrast several curves are too close to zero to be distinguished but they remain in same order. The two Fig. 8 right show for both contrasts how the four matrix contributions from the two-phase elementary assemblages yield a null sum for the composite reference matrix (according to Eq. (23b)), when using the same four fixed probabilities from Eqs. (29,30) and shown in Fig. 7 right (on both Fig. 8 right, the plotted null sum confuses with the abscissa axis). In brief analysis, 3 contributions over 4 are negative (resp. positive) at low hard (resp. soft) phase fractions and two are of each sign in the median zone. The sharp transitions at high contrast near 40% of hard phase correspond to the sharp slope change of the *SC* estimate. As far as the modulus of the composite reference matrix is low (resp. high), the negative term(s) come from the stiff (rep. weak) elementary matrices.

[10] All Green operators with a single term symmetry in the I-I elastic or I dielectric cases yield a different w value.

[11] From the second part of the (iiii) term for elastic-like rank-four operators.

5.2. A multi-moduli problem example on an isotropic-compressible (I-C) elastic composite

The direct problem of finding a probability set $P(N)$ to fix a reference matrix for any n-phase composite following the proposed "reference matrix mixture" method according to Eq. (23b) applies for any phase moduli symmetries. Two experimental comparisons have been possible from literature data concerning a particular type of 3-phase bicontinuous composite, made of a porous medium (commercially pure titanium) in which the porosity is partly connected and partly disconnected (Torres et al., 2012). It is a multi-moduli I-C elastic situation, with a priori 6 distinct phase moduli. This microstructure is representative of the type 4 phase arrangement presented in Sections 1 and 2. The reported literature data concerning both the three phase relative concentrations and the corresponding effective Young moduli allow to first compare with the proposed $\mathbf{C}^{*mLS}_{(p,n)} = \mathbf{C}^{*mLS}_{(2,3)}$ estimate recalled in Section 2.3 for which the fourth reference matrix phase of infinitesimal concentration has the properties of the reference matrix $\mathbf{C}^{*mLS}_{(2)}$ for the bicontinuous composite part, according to Eq. (14). If the data are not considered precise enough to ensure the elementary arrangement $\mathbf{C}^{*mLS}_{(2,3)}$ be the best estimate, it is possible to apply the "matrix mixture method" from determining a likely probability set $P(4)$ also based on the available data. We here report and compare the obtained results from these two estimates.

In the Isotropic-Compressible (I-C) elastic cases, considering both shear μ and bulk θ moduli eigenvalues, the tensorial Eq. (22b) for the moduli estimates from the reference matrix mixture using the *SC* scheme yields to two similar scalar ones, one for each modulus, as:

$$0 = \sum_{J=1}^{N=A^{(n)}} P_J \left(\left(\mu^* - \mu^{oJ} \right)^{-1} - t^{Iso}_{\mu^*} \right)^{-1}$$
$$= \sum_{J=1}^{N=A^{(n)}} P_J \left(\frac{1}{\mu^* - \mu^{oJ}} - \frac{2}{5\mu^*} \left(\frac{6\mu^* + \theta^*}{4\mu^* + \theta^*} \right) \right)^{-1}, \tag{31a}$$

$$0 = \sum_{J=1}^{N=A^{(n)}} P_J \left(\left(\theta^* - \theta^{oJ} \right)^{-1} - t^{Iso}_{\theta^*} \right)^{-1}$$
$$= \sum_{J=1}^{N=A^{(n)}} P_J \left(\frac{1}{\theta^* - \theta^{oJ}} - \frac{3}{4\mu^* + 3\theta^*} \right)^{-1}. \tag{31b}$$

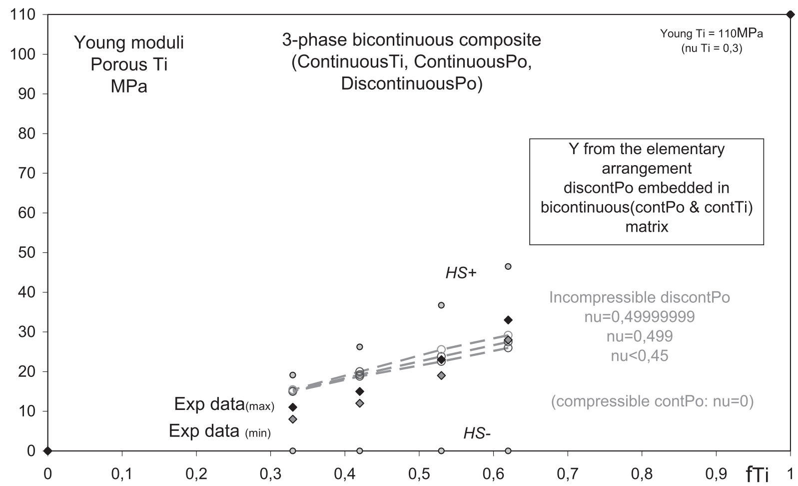

Fig. 9. Effective Young moduli estimates from a cLS(2,3) scheme for a 3-phase bicontinuous composite associating connected and disconnected porosities in a titanium solid phase. Experimental moduli and structure characteristics correspond to data from (Torres et al., 2012).

The particular explicit two-phase forms of this *SC* estimate are given in Appendix C which sketches briefly the route to investigate the two moduli inverse problem corresponding to the one-modulus I-I case treated in the previous sub-section. The involved Green operator terms in Eq. (31) take the coupled forms with regard to the moduli eigenvalues μ and $\theta = \frac{2\mu(1+\nu)}{1-2\nu}$:

$$t^{Iso}_{\mu^*} = \frac{2}{15\mu}\left(\frac{4-5\nu}{1-\nu}\right) = \frac{2}{5\mu}\frac{6\mu+\theta}{4\mu+\theta};\ t^{Iso}_{\theta} = \frac{1-2\nu}{2\mu(1-\nu)} = \frac{3}{4\mu+3\theta}, \tag{32}$$

that result from the terms of the isotropic (spherical) strain Green operator recalled in (Franciosi and Lormand, 2004; Franciosi, 2010):

$$\begin{cases} t^{Sph}_{1111} = \frac{1}{5}\left(\frac{-1}{2\mu(1-\nu)}\right) + \frac{1}{3}\left(\frac{1}{\mu}\right) = \frac{7-10\nu}{30\mu(1-\nu)} \\ t^{Sph}_{1212} = \frac{1}{15}\left(\frac{-1}{2\mu(1-\nu)}\right) + \frac{1}{6}\left(\frac{1}{\mu}\right) = \frac{4-5\nu}{30\mu(1-\nu)} \end{cases},$$

$$t^{Sph}_{1122} = t^{Sph}_{1111} - 2^{Sph}_{1212} = \frac{-1}{30\mu(1-\nu)}. \tag{33}$$

According to the *SC* scheme when n>2, Eq. (31) are solved from a common iterative procedure applied on the scalar forms of Eq. (5) in starting for any arbitrary matrix in the admissible (i.e. within *HS* bounds) range and which easily converges to a moduli pair (μ^{*SC},θ^{*SC}).

- ***The porous titanium as a 3 phase bicontinuous composite.*** The difference with the exemplified curves for the sole shear modulus in Fig. 6 for a I-I elastic material is that here the three (isotropic) phases have different compressibility moduli $\theta = Y/(1-2\nu)$, where $Y = 2\mu(1+\nu)$ is the Young modulus, with at most one phase being nearly incompressible ($\nu \cong 0,5$), say the discontinuous (embedded) part of the porosity. Compressibility and shear moduli for *Ti* are $\theta_{Ti} = 275MPa$ and $\mu_{Ti} = 42,31MPa$ respectively from the *given* Young modulus $Y_{Ti} = 110MPa$ and a literature Poisson ratio $\nu_{Ti} = 0,3$. The connected (continuous) porosity phase has been taken of null Poisson ratio (compressibility modulus $\theta_{Pocont} = 2\mu_{Po}$). The whole porous phase has an assumed shear modulus $\mu_{Po} = 1.10^{-3}MPa << \mu_{Ti}$. The shear moduli ratio $\frac{\mu_{Po}}{\mu_{Ti}} = \frac{1.10^{-3}}{42,31} \approx 2,3.10^{-5}$ is arbitrarily chosen and not varied. The only varied modulus in our performed simulations is the Poisson ratio for the incompressible porosity embedded part ν_{Podisc} from 0.4 to 0.5, such that the compressibilty modulus range is $\theta_{Podisc} \in \{0,03-15.10^6\}$. According to Eq. (14), the two moduli μ, θ of

Table 1
Data for the curves plotted in Fig. 8 (the asterisks indicate the parameters estimated from the data in (Torres et al., 2012) and estimated Young moduli from the mLS(2,3) scheme for a three-phase bicontinuous I-C elastic composite.

fPo *	*fTi* *	*fPocont* *	*fPodisc* *	*fBicontMatr* *	Y MPa *	Y MPa *mLS*(2,3)
0,38	0,62	0,312	0,068	0,932	28–33	26–29,2
0,47	0,53	0,3896	0,0804	0,9196	19–23	22,5–25,6
0,58	0,42	0,5452	0,0348	0,9652	12–15	18,8–20
0,67	0,33	0,6539	0,0161	0,9839	8–11	15,1–15,4

the infinitesimal reference matrix fourth phase made of the two co-continuous phases take the form $\{x\}_{Ti,Pocont} = \frac{f_{Pocont}x_{Ti}+f_{Ti}x_{Pocont}}{f_{Pocont}+f_{Ti}}$. Then the effective moduli are obtained from embedding the three (*Ti, Pocont, Podisc*) phases in this reference matrix.

The plotted comparisons with $\mathbf{C}^{*mLS}_{(2,3)}$ estimated Young moduli in Fig. 9 for 4 different phase relative volume fractions correspond to the data collected in Table 1 for the phase concentrations and for the effective Young moduli, including the obtained $\mathbf{C}^{*mLS}_{(2,3)}$ estimate. The *HS* bounds are also indicated. The composite is issued from a powder compaction technique that we do not enter in. The selected set of comparison data corresponds to those obtained for the highest compaction pressure of 800 MPa used in elaboration. They do not differ noticeably from a second group (obtained at 600 MPa pressure) in terms of phase concentrations, but the measured Young moduli slightly differ. The min/max reported interval in Figs. 9 (and 10) for the measured Young moduli to compare with estimates is based on these two (800–600) MPa data series.

With regard to general accuracy of porosity measurements, the obtained $Y^{*mLS}_{(2,3)}$ estimate pretty well matches the measured moduli and any better matching attempt (especially at the lowest *Ti* volume fraction points) needs more accurate phase (moduli and concentrations) characteristics which are the only information contributions to insert in the estimate expression from Eq. (31) for the two (shear and bulk) moduli eigenvalues. In particular, varying $\theta_{Podisc} \in \{0,03-15.10^6\}$ in varying the Poisson ratio to the closest to 0.5 is shown to have very little effect on the resulting Young moduli estimates (bounded at $Y^* = 3\mu^*$) and all other moduli cannot be varied much.[12]

[12] Taking $\nu_{Ti} = 0,31$ also frequently found in literature yields $\theta_{Ti} = 290MPa$ and $\mu_{Ti} = 42MPa$.

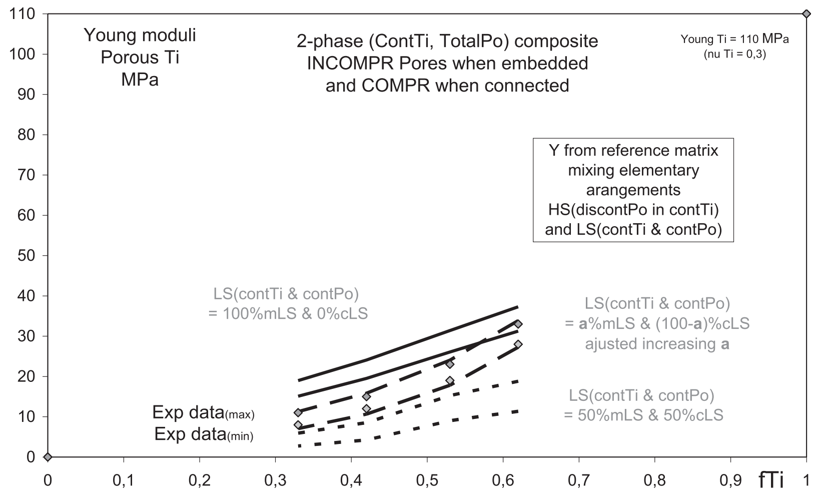

Fig. 10. Effective Young moduli estimates for a (porous titanium) two-phase composite, based on the proposed "matrix mixture description" from the four elementary two-phase arrangements with given probabilities. Experimental moduli and structure characteristics correspond to data from (Torres et al., 2012).

- ***The porous titanium as a two-phase composite*** of which the connected versus disconnected porosity status is not precisely known. For application of the "reference matrix mixture" method as proposed, the need is a four-probability set $P(4)$ to obtain a reference matrix for the (porous titanium) composite as the SC effective moduli of the assemblage of the reference matrices of the four two-phase elementary arrangements. These four reference matrices have the moduli pairs which are those of the two constitutive phases, with the additional consideration that the pore compressibility depends on the continuous versus discontinuous nature of its arrangement, as done for the 3-phase analysis reported first. For the HS estimates, the phase moduli are (μ_{Ti},θ_{Ti}) and (μ_{Po},θ_{Po}) with θ_{Po} being incompressible when included and compressible if it were (although unlikely as seen next) embedding the Ti phase. Similarly for the $mLS(2)$ and $cLS(2)$ estimates, the moduli pairs are $(\{\mu\}_{Ti,Po}, \{\theta\}_{Ti,Po})$ and $(\overline{\{\bar{\mu}\}}_{Ti,Po}, \overline{\{\bar{\theta}\}}_{Ti,Po})$, with the given definitions of the Voigt and Reuss averages on the harmonic two-phase medium, for both moduli, $\{x\}_{Ti,Po} = f_{Po}x_{Ti} + f_{Ti}x_{Po}$ and $\overline{\{\bar{x}\}}_{Ti,Po} = (f_{Po}(1/x_{Ti}) + f_{Ti}(1/x_{Po}))^{-1}$. θ_{Po} is taken compressible in the bicontinuous $mLS(2)$ scheme and incompressible in the $cLS(2)$ one.

Then taking the probability $P_{Cont}(Ti) = 1 - P_{Disc}(Ti) = 1$, yields for the probability of the lower HS estimate that $P(HS^-)_{Ti,Po} = P_{Ti\subset Po} = 0$, regardless of the probabilities for the pore phase $P_{Cont}(Po) = 1 - P_{Disc}(Po)$ to be continuous or discontinuous. If these probabilities were unconditional to those on Ti (what is not known), the probability of bicontinuity would be $P_{Cocont}(Ti,Po) = P_{Cont}(Ti) \times P_{Cont}(Po) = P_{Cont}(Po)$ and the one of "bi-discontinuity" $P_{Codisc}(Ti,Po) = P_{Disc}(Ti) \times P_{Disc}(Po) = 0$. Similarly for probability $P(HS^+)_{Ti,Po} = P_{Po\subset TI}$ it would read $P_{Cont}(Ti) \times P_{Disc}(Po) = P_{Disc}(Po)$. In which case, the non zero probabilities are $P(HS^+)_{Ti,Po} = P_{Disc}(Po)$, and $P_{Cocont}(Ti,Po) = P(mLS)_{Ti,Po} = P_{Cont}(Po)$. This is already different from the previous pure bicontinuous description plotted in Fig. 9. The addition of a probability that the structure may have a part with all embedded porosity makes it stiffer, as expectedly.

Considering the third probability $P(cLS)_{Ti,Po}$ to not be necessarily zero can for example be justified by conditional probabilities (that we do not know either). It is noteworthy that while the co-discontinuous interpretation of the phase arrangement type associated to the $cLS(2)$ estimate prohibits a non zero $P(cLS)_{Ti,Po}$ value for Ti cannot be discontinuous, the interpretation of minimal co-continuity allows it, corresponding to a weakly continuous part of the Ti phase mixed with a weakly connected porous part. In order to compare the first estimate (with $P(cLS)_{Ti,Po} = 0$)

Table 2
Estimated probabilities for the four elementary two-phase arrangements to contribute to the two-phase porous titanium structure examined and characterized in (Torres et al., 2012).

$fPo*$	$\frac{fPodisc}{fPo}*$	$\frac{fPocont}{fPo}*$	$P(HS^+)$	$P(mLS)$	$P(cLS)$	$P(HS^-)$
0,38	0,18	0,82	0,18	$0{,}82 \times (A)$	$0{,}82 \times (1-A)$	0
0,47	0,17	0,83	0,17	$0{,}83 \times (A)$	$0{,}83 \times (1-A)$	0
0,58	0,06	0,94	0,06	$0{,}94 \times (A)$	$0{,}94 \times (1-A)$	0
0,67	0,024	0,976	0,024	$0{,}976 \times (A)$	$0{,}976 \times (1-A)$	0

with estimates obtained with a non zero $P(cLS)_{Ti,Po}$, one introduces a coefficient A in order to consider probability forms as $P(mLS)_{Ti,Po} = P_{Cont}(Po) \times A$ and $P(cLS)_{Ti,Po} = P_{Cont}(Po) \times (1-A)$, while keeping $P(HS^+)_{Ti,Po} = P_{Disc}(Po)$. In Table 2 are collected the data concerning the continuous and discontinuous parts of the porosity as measured in (Torres et al., 2012). Using the assumption that $P_{Cont}(Po) = \frac{f(Pocont)}{f(Po)}$ and $P_{Disc}(Po) = \frac{f(Podisc)}{f(Po)}$, the Table 3 summarizes the 3 cases of A values from which estimated Young moduli are compared with the measured ones. Since the compressibility of the porosity has been taken different whether it is connected or not, pairs of Young moduli have been estimated for the two-phase composite, corresponding to a pore phase being either totally incompressible or totally compressible. The obtained values are plotted in Fig. 10, in comparison with the measured moduli: case 1, with $A = 100\%$, corresponds to the strict two non zero probability case which yields an effective modulus interval stiffer than measured one, with a globally increasing tendency. Case 2 assumes a constant $A = 50\%$ value independently of the (Ti, Po) partition. This estimate interval also increases regularly but in remaining below the experimental interval. The third test makes use of roughly adjusted A values on the measured Young moduli to determine which variation with the Ti volume fraction must have the ratio $\frac{P(cLS)_{Ti,Po}}{P(mLS)_{Ti,Po}} = \frac{1-A}{A}$ (note that the estimated two (lower and upper) moduli, for compressible and incompressible pores, approximately match the min/max experimental interval limits such as to compare the middle lines of both intervals, but it is not meant that there must be a one-to-one matching of these curve pairs). One can notice that the satisfyingly matching A values are larger than the fTi volume fraction they correspond to and that, in the fTi range of the data, A increases between the two previously tested constant values of 50% and 100%. Hence, the fraction of phase arrangement represented by the $cLS(2)$ estimate decreases drastically in comparison to the increase of the one represented by the $mLS(2)$

Table 3
Data for the curves plotted in Figure 13 (the asterisks indicate the parameters estimated from the data in (Torres et al., 2012) and estimated Young moduli from the "matrix mixture" method for a two-phase I-C elastic composite.

fPo *	*fTi* *	*A* values Case 1	*A* values Case 2	*A* values Case 3	*Y* MPa Case 1	*Y* MPa Case 2	*Y* MPa Case 3	*Y* MPa *
0,38	0,62	1	0,5	0,87	31,25–37,3	11,37–18,8	27,3–33,9	28–33
0,47	0,53	1	0,5	0,72	26,1–31,37	9–15,25	17,84–24	19–23
0,58	0,42	1	0,5	0,68	19,5–24,06	4,25–8,54	10,8–15,85	12–15
0,67	0,33	1	0,5	0,65	15,1–18,96	2,75–5,9	7,07–11,3	8–11

estimate. An interpretation would be that a weakly co-connected composite fraction is significantly present at medium *Ti* concentrations and decreases with the decrease of the total porosity, such that only a strongly co-connected composite remains at higher Ti concentrations. This encouragingly shows that the $mLS(p, n)$ estimate type appears relevant for stiffness estimate of multi continuous composites and that the proposed "matrix mixture method" is consistent as well and is also capable of providing insights on phase arrangement characteristics in composite structures. Further examination of this method will be the topic of forthcoming papers.

6. Conclusion

Starting from previously obtained new results concerning the effective property estimates for the various one-level (opposed to multi-scaled) phase arrangement possibilities in *n*-phase composite structures, we here elaborate further on multiple phase continuity in composites as viewed from Laminate System (*LS*) schemes. These Laminate Systems were defined first as interpenetrated directional laminate structures to represent 3D co-continuity of several assembled phases, none of them playing a priori a particular role. This phase co-continuity having been shown to enhance the composite effective stiffness in a way well represented by arithmetic (weighted) average of the stiffness properties of directional laminate structures constitutive of the system, the dual approach considering arithmetic averaging of compliances (harmonic average of stiffness) were first associated to the converse phase co-discontinuity assumption without clear representation.

Continuity being to be taken in the sense of connectedness, phase discontinuity is necessarily a set of disconnected domains of same phase. This definition makes impossible full co-discontinuity in the sense of simultaneous disconnectedness of n assembled phases. The minimum of connectedness for a single phase being 3D through-sample spanning infinite clusters made of point-connected finite domains, minimal co-continuity of n phases appears to be the closest possible arrangement corresponding to co-discontinuity. Hence, the duality co-continuity versus "co-discontinuity" is substituted with maximal versus minimal co-continuity. This is one first clarification of the present work to associate the arithmetic stiffness averaging to maximal phase co-continuity and conversely the arithmetic compliance averaging to minimal phase co-continuity.

This refined interpretation makes easier to figure out minimally co-continuous phase arrangements. The made availability of estimates for any possible elementary phase arrangement among *n* opens on potential applications in physical processes involving possible changes of connectedness status for some of the constitutive phase of a mixture. This happens during solidification/fusion, precipitation/dissolution, compaction/fragmentation and other many phenomena characterized by percolation-like sharp or gradual transitions.

Beyond this, the essential purpose of this work is a description proposal for any composite when not well defined in terms of its elementary phase arrangement as a combination or "mixture" of them. In examining how a two-phase disordered composite could be so described, it is first shown that a description based on defining a reference matrix for the composite as a disordered assemblage of the reference matrices of its elementary phase arrangements provides access to an admissible solution. This description is supported by a remarkable relation found in the simple cases of disordered two-phase materials with Isotropic (dielectric-like) or Isotropic-Incompressible (elastic-like) one-modulus (physical) symmetry: a definition of the reference matrix of such a disordered two-phase composite as a disordered mixture of the reference matrices of its elementary phase arrangements proves to be independent of the phase moduli contrast. This relation between the elementary estimate types for two-phase composites is a result of the use of the two dual Laminate System (*LS*) estimates for representing the effective properties of bi-continuous (maximally continuous) and of bi-discontinuous (minimally continuous) phase arrangements. They complement the Hashin-Shtrikman (*HS*) estimates for the two other arrangements of the well known reinforced-matrix types to provide that unique relation with the *SC* estimate. Application to experimental data from literature on a 3-phase material both supports the relevancy of the proposed estimate for multi-continuous composites and of the proposed "reference matrix mixture" method for estimating effective properties of general composites.

Appendix A. Effective properties of multi-continuous materials obey LS schemes

The first considered co-continuous two-phase case is next extended to p co-continuous ones among n.

A1: for two-phase cases (A,B), assuming Eq. (15) yields the relations (with $\Delta\boldsymbol{C}_{BA} = \boldsymbol{C}_B - \boldsymbol{C}_A$):

$$\begin{aligned}\Delta\boldsymbol{C}_{BA} &= f_A\left(\Delta\boldsymbol{C}_{BA}{}^{-1} - f_B\boldsymbol{t}_B^V\right)^{-1} + f_B\left(\Delta\boldsymbol{C}_{BA}{}^{-1} + f_A\boldsymbol{t}_A^V\right)^{-1} \\ &= \Delta\boldsymbol{C}_{BA} : \left(f_A\left(\boldsymbol{I} - f_B\boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA}\right)^{-1} + f_B\left(\boldsymbol{I} + f_A\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA}\right)^{-1}\right),\end{aligned} \tag{A1.1}$$

$$\boldsymbol{I} = f_A\left(\boldsymbol{I} - f_B\boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA}\right)^{-1} + f_B\left(\boldsymbol{I} + f_A\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA}\right)^{-1}, \tag{A1.2}$$

$$\left(\boldsymbol{I} + f_A\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA}\right) : \left(\boldsymbol{I} - f_B\boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA}\right) = f_A\left(\boldsymbol{I} + f_A\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA}\right) + f_B\left(\boldsymbol{I} - f_B\boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA}\right), \tag{A1.3}$$

$$\boldsymbol{I} + \left(f_A\boldsymbol{t}_A^V - f_B\boldsymbol{t}_B^V - f_Af_B\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_B^V\right) : \Delta\boldsymbol{C}_{BA} : \boldsymbol{I} + \left(f_A^2\boldsymbol{t}_A^V - f_B^2\boldsymbol{t}_B^V\right) : \Delta\boldsymbol{C}_{BA}, \tag{A1.4}$$

$$f_A\boldsymbol{t}_A^V - f_B\boldsymbol{t}_B^V - f_A^2\boldsymbol{t}_A^V + f_B^2\boldsymbol{t}_B^V = f_Af_B(\boldsymbol{t}_A^V - \boldsymbol{t}_B^V) = f_Af_B\boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_B^V, \tag{A1.5}$$

what yields the laminate property of Eq. (13) that $\boldsymbol{t}_A^V - \boldsymbol{t}_B^V = \boldsymbol{t}_A^V : \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_B^V$.

A2: for three phases A,B,D, assuming phases A and B to be continuous yields from Eq. (16b):

$$\Delta\boldsymbol{C}_{BA} = \left(\overline{\langle\boldsymbol{T}\rangle_{(B)}} + \boldsymbol{t}_B^V\right)^{-1} - \left(\overline{\langle\boldsymbol{T}\rangle_{(A)}} + \boldsymbol{t}_A^V\right)^{-1}, \tag{A2.1}$$

$$\left(\overline{\langle\boldsymbol{T}\rangle_{(B)}} + \boldsymbol{t}_B^V\right) : \Delta\boldsymbol{C}_{BA} : \left(\overline{\langle\boldsymbol{T}\rangle_{(A)}} + \boldsymbol{t}_A^V\right) = \left(\overline{\langle\boldsymbol{T}\rangle_{(A)}} + \boldsymbol{t}_A^V\right) - \left(\overline{\langle\boldsymbol{T}\rangle_{(B)}} + \boldsymbol{t}_B^V\right) = \boldsymbol{t}_A^V - \boldsymbol{t}_B^V + \overline{\langle\boldsymbol{T}\rangle_{(A)}} - \overline{\langle\boldsymbol{T}\rangle_{(B)}}, \tag{A2.2}$$

$$= \overline{\langle\boldsymbol{T}\rangle_{(B)}} : \Delta\boldsymbol{C}_{BA} : \overline{\langle\boldsymbol{T}\rangle_{(A)}} + \boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA} : \overline{\langle\boldsymbol{T}\rangle_{(A)}} + \overline{\langle\boldsymbol{T}\rangle_{(B)}} : \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_A^V + \boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_A^V, \tag{A2.3}$$

with over barred tensors standing for their inverse. This arrives at the equality of Eq. (17).

Next multiplying left by $\langle\boldsymbol{T}\rangle_{(B)}$ and right by $\langle\boldsymbol{T}\rangle_{(A)}$, it comes, with:

$$\left.\begin{aligned}\langle\boldsymbol{T}\rangle_{(A)} &= f_B\left((\boldsymbol{C}_A - \boldsymbol{C}_B)^{-1} - \boldsymbol{t}_A^V\right)^{-1} + f_D\left((\boldsymbol{C}_A - \boldsymbol{C}_D)^{-1} - \boldsymbol{t}_A^{V_D}\right)^{-1} = f_B\boldsymbol{T}_B^A + f_D\boldsymbol{T}_D^A \\ \langle\boldsymbol{T}\rangle_{(B)} &= f_A\left((\boldsymbol{C}_B - \boldsymbol{C}_A)^{-1} - \boldsymbol{t}_B^V\right)^{-1} + f_D\left((\boldsymbol{C}_B - \boldsymbol{C}_D)^{-1} - \boldsymbol{t}_B^{V_D}\right)^{-1} = f_A\boldsymbol{T}_A^B + f_D\boldsymbol{T}_D^B\end{aligned}\right\}, \tag{A2.4}$$

$$\Delta\boldsymbol{C}_{BA} + \langle\boldsymbol{T}\rangle_{(B)} : \boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA} + \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_A^V : \langle\boldsymbol{T}\rangle_{(A)} + \langle\boldsymbol{T}\rangle_{(A)} - \langle\boldsymbol{T}\rangle_{(B)} = 0. \tag{A2.5}$$

One arrives at Eq. (18), noting that $\boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA} = \boldsymbol{I} - \boldsymbol{t}_B^V : \overline{\boldsymbol{t}_A^V}$ and $\Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_A^V = \overline{\boldsymbol{t}_B^V} : \boldsymbol{t}_A^V - \boldsymbol{I}$, such that:

$$\langle\boldsymbol{T}\rangle_{(B)} : \boldsymbol{t}_B^V : \Delta\boldsymbol{C}_{BA} + \Delta\boldsymbol{C}_{BA} : \boldsymbol{t}_A^V : \langle\boldsymbol{T}\rangle_{(A)} = \langle\boldsymbol{T}\rangle_{(B)} : \left(\boldsymbol{I} - \boldsymbol{t}_B^V : \overline{\boldsymbol{t}_A^V}\right) + \left(\overline{\boldsymbol{t}_B^V} : \boldsymbol{t}_A^V - \boldsymbol{I}\right) : \langle\boldsymbol{T}\rangle_{(A)}. \tag{A2.6}$$

Appendix B. Elementary structure analysis for disordered two-phase isotropic-incompressible composites

In the isotropic incompressible (I-I) elastic case that reduces to a single (shear) modulus to estimate, all terms of the form $N^X - N^Y$ read, for the two description options considered in Section 4, $\frac{\mu^o - \widehat{\mu}^{*X}}{3\mu^o + 2\widehat{\mu}^{*X}} - \frac{\mu^o - \widehat{\mu}^{*Y}}{3\mu^o + 2\widehat{\mu}^{*Y}} = \frac{5\mu^o(\widehat{\mu}^{*Y} - \widehat{\mu}^{*X})}{(3\mu^o + 2\widehat{\mu}^{*X})(3\mu^o + 2\widehat{\mu}^{*Y})}$. When directly considering the reference matrix option 2 ($\widehat{\mu}^* = \mu^o$) which numerically proved to satisfy the remarkable contrast independency property, the system to solve from Eq. (27c), simplifies to[13]:

$$PP'\left(\frac{\mu_{[A,B]}^{OcLS} - \mu_{[A,B]}^{OmLS}}{3\mu^{[o]} + 2\mu_{[A,B]}^{OmLS}} - \left(\frac{\mu_{[A,B]}^{OcLS} - \mu^A}{3\mu^{[o]} + 2\mu^A} + \frac{\mu_{[A,B]}^{OcLS} - \mu^B}{3\mu^{[o]} + 2\mu^B}\right)\right) + P\frac{\mu_{[A,B]}^{OcLS} - \mu^A}{3\mu^{[o]} + 2\mu^A} + P'\frac{\mu_{[A,B]}^{OcLS} - \mu^B}{3\mu^{[o]} + 2\mu^B} = \frac{\mu_{[A,B]}^{OcLS} - \mu^{[o]}}{5\mu^{[o]}},$$

$$PP'\left(\frac{\mu_{]A,B[}^{OcLS} - \mu_{]A,B[}^{OmLS}}{3\mu^{]o[} + 2\mu_{]A,B[}^{OmLS}} - \left(\frac{\mu_{]A,B[}^{OcLS} - \mu^A}{3\mu^{]o[} + 2\mu^A} + \frac{\mu_{]A,B[}^{OcLS} - \mu^B}{3\mu^{]o[} + 2\mu^B}\right)\right) + P\frac{\mu_{]A,B[}^{OcLS} - \mu^B}{3\mu^{]o[} + 2\mu^B} + P'\frac{\mu_{]A,B[}^{OcLS} - \mu^A}{3\mu^{]o[} + 2\mu^A} = \frac{\mu_{]A,B[}^{OcLS} - \mu^{]o[}}{5\mu^{]o[}}.$$

All terms of each these equations were multiplied by $\frac{3\mu^{[o]} + 2\mu_{[A,B]}^{OcLS}}{5\mu^{[o]}}$ and $\frac{3\mu^{]o[} + 2\mu_{]A,B[}^{OcLS}}{5\mu^{]o[}}$ respectively and use is made of $\widehat{\mu}_{[A,B]}^{*HSA} = \mu_{[A,B]}^{OHSA} = \mu^A$, $\widehat{\mu}_{[A,B]}^{*HSB} = \mu_{[A,B]}^{OHSB} = \mu^B$, $\langle N\rangle_{[A,B]}^{[o]} = 0$, $\{N\}_{]A,B[}^{]o[} = 0$.

Further multiplying all (scalar) coefficients by respectively $\langle\mu\rangle$ and $\{\mu\}$, yields, without modifying the Equation system to solve in Eq. (27c), for the two left hand sides:

$$PP'\left(\frac{\mu^A\mu^B - \{\mu\}\langle\mu\rangle}{\left(3\mu^{[o]} + 2\{\mu\}\right)} - \left(\frac{\mu^A\left(\mu^B - \langle\mu\rangle\right)}{\left(3\mu^{[o]} + 2\mu^A\right)} + \frac{\mu^B\left(\mu^A - \langle\mu\rangle\right)}{\left(3\mu^{[o]} + 2\mu^B\right)}\right)\right) + P\frac{\mu^A\left(\mu^B - \langle\mu\rangle\right)}{\left(3\mu^{[o]} + 2\mu^A\right)} + P'\frac{\mu^B\left(\mu^A - \langle\mu\rangle\right)}{\left(3\mu^{[o]} + 2\mu^B\right)},$$

[13] The notation $[A, B]$ (resp. $]A, B[$) stands for the material with f_A fraction of phase A (resp. B) and f_B of phase B (resp. A), while $[o]$ (resp. $]o[$) stands for the matrix of the related *SC* estimate, by definition of which both quantities $\langle N\rangle_{[A,B]}^{[o]}$ and $\{N\}_{]A,B[}^{]o[}$ are simultaneously zero.

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$$PP'\left(\frac{\mu^A\mu^B-\langle\mu\rangle\{\mu\}}{\left(3\mu^{]o[}+2\langle\mu\rangle\right)}-\left(\frac{\mu^A\left(\mu^B-\{\mu\}\right)}{\left(3\mu^{]o[}+2\mu^A\right)}+\frac{\mu^B\left(\mu^A-\{\mu\}\right)}{\left(3\mu^{]o[}+2\mu^B\right)}\right)\right)+P\frac{\mu^B\left(\mu^A-\{\mu\}\right)}{\left(3\mu^{]o[}+2\mu^B\right)}+P'\frac{\mu^A\left(\mu^B-\{\mu\}\right)}{\left(3\mu^{]o[}+2\mu^A\right)},$$

to be respectively equal to the related right hand sides $\frac{\mu^A\mu^B-\mu^{[o]}\langle\mu\rangle}{5\mu^{[o]}}$ and $\frac{\mu^A\mu^B-\mu^{]o[}\{\mu\}}{5\mu^{]o[}}$.

Use was here made of the definitions $\mu^{OmLS}_{[A,B]}=\{\mu\}$, $\mu^{OcLS}_{[A,B]}=\overline{\{\mu\}}=\frac{\mu^A\mu^B}{\langle\mu\rangle}$, $\mu^{OmLS}_{]A,B[}=\langle\mu\rangle$, $\mu^{OcLS}_{]A,B[}=\overline{\langle\mu\rangle}=\frac{\mu^A\mu^B}{\{\mu\}}$ for the reference matrices of the two Laminate System schemes related to the two $[A, B]$ and $]A, B[$ dual two-phase I-I composites. Additional rearrangements using relations such as $\mu^A\mu^B-\langle\mu\rangle\{\mu\}=-f_Af_B(\mu^A-\mu^B)^2$; $\mu^A(\mu^B-\langle\mu\rangle)=f_A\mu^A(\mu^B-\mu^A)$; $\mu^B(\mu^A-\langle\mu\rangle)=f_B\mu^B(\mu^A-\mu^B)$; $\mu^A(\mu^B-\{\mu\})=f_B\mu^A(\mu^B-\mu^A)$; $\mu^B(\mu^A-\{\mu\})=f_A\mu^B(\mu^A-\mu^B)$ and also introducing the reduced notations $\tilde\mu^I=\mu^I/\mu^A$ with the contrast $r=\tilde\mu^B=\mu^B/\mu^A$, yields finally (after having divided by the common term $r-1$) to the left hand side forms:

$$PP'\left(-\frac{f_Af_B(r-1)}{3\tilde\mu^{[o]}+2\{\tilde\mu\}}-\frac{f_A}{3\tilde\mu^{[o]}+2}+\frac{f_Br}{3\tilde\mu^{[o]}+2r}\right)+P\frac{f_A}{3\tilde\mu^{[o]}+2}-P'\frac{f_Br}{3\tilde\mu^{[o]}+2r},$$

$$PP'\left(-\frac{f_Af_B(r-1)}{3\tilde\mu^{]o[}+2\langle\tilde\mu\rangle}-\frac{f_B}{3\tilde\mu^{]o[}+2}+\frac{f_Ar}{3\tilde\mu^{]o[}+2r}\right)-P\frac{f_Ar}{3\tilde\mu^{]o[}+2r}+P'\frac{f_B}{3\tilde\mu^{]o[}+2},$$

to be respectively equaled with $\frac{f_A(r-\tilde\mu^{[o]})+f_Br(1-\tilde\mu^{[o]})}{5\tilde\mu^{[o]}(r-1)}$ and $\frac{f_B(r-\tilde\mu^{]o[})+f_Ar(1-\tilde\mu^{]o[})}{5\tilde\mu^{]o[}(r-1)}$.

This is still the system of Eq. (27c) with simplified coefficients which can be shown related as:

$$\frac{3\tilde\mu^{[o]}+2\{\tilde\mu\}}{f_Af_B}=\frac{3\tilde\mu^{[o]}+2}{f_A}+\frac{3\tilde\mu^{[o]}+2r}{f_B}=\frac{1}{[b]}+\frac{r}{[d]}=>[a]=[d]-[b]-\frac{(r-1)[b][d]}{[d]+r[b]}$$

$$\frac{3\tilde\mu^{]o[}+2\langle\tilde\mu\rangle}{f_Af_B}=\frac{3\tilde\mu^{]o[}+2}{f_B}+\frac{3\tilde\mu^{]o[}+2r}{f_A}=\frac{1}{]b[}+\frac{r}{]d[}=>]a[=]d[-]b[-\frac{(r-1)]b[]d[}{]d[+r]b[}$$

and with also $[a]=\frac{[d]^2-r[b]^2}{[d]+r[b]}$ and $]a[=\frac{]d[^2-r]b[^2}{]d[+r]b[}$.

The effective shear moduli $\tilde\mu^{[o]}$ and $\tilde\mu^{]o[}$ from the *SC* estimate forms in each equation are solution of the relations which read (once reduced accordingly to the stepwise coefficient modifications and wit $p=\frac{3}{5}f_A-\frac{2}{5}f_B=f_A-\frac{2}{5}$ and $q=\frac{3}{5}f_B-\frac{2}{5}f_A=f_B-\frac{2}{5}$):

$$\frac{3}{5}\tilde\mu^{[o]2}-\tilde\mu^{[o]}(p+qr)-\frac{2}{5}r=0 \text{ and } \frac{3}{5}\tilde\mu^{]o[2}-\tilde\mu^{]o[}(q+pr)-\frac{2}{5}r=0, \tag{B.1}$$

such that:

$$r=\frac{3\tilde\mu^{[o]2}-\tilde\mu^{[o]}p}{\tilde\mu^{[o]}q+2}=\tilde\mu^{[o]}\frac{3\tilde\mu^{[o]}-p}{\tilde\mu^{[o]}q+2};\ r-1=\frac{\left(\tilde\mu^{[o]}-1\right)\left(3\tilde\mu^{[o]}+2\right)}{\tilde\mu^{[o]}q+2};\ r-\tilde\mu^{[o]}=\frac{5f_A\tilde\mu^{[o]}\left(\tilde\mu^{[o]}-1\right)}{\tilde\mu^{[o]}q+2}$$

(resp. $r=\tilde\mu^{]o[}\frac{3\tilde\mu^{]o[}-q}{\tilde\mu^{]o[}p+2}$); $r-1=\frac{(\tilde\mu^{]o[}-1)(3\tilde\mu^{]o[}+2)}{\tilde\mu^{]o[}p+2}$; $r-\tilde\mu^{]o[}=\frac{5f_B\tilde\mu^{]o[}(\tilde\mu^{]o[}-1)}{\tilde\mu^{]o[}p+2}$).

Then, the coefficients in the Equation system to solve can be simply written:

$$[d]=\frac{f_Br}{3\tilde\mu^{[o]}+2r}=\frac{f_Br\left(\tilde\mu^{[o]}-1\right)}{(q+2)\tilde\mu^{[o]}(r-1)}=\frac{r\left(\tilde\mu^{[o]}-1\right)}{5\tilde\mu^{[o]}(r-1)},$$

$$[b]=\frac{f_A}{3\tilde\mu^{[o]}+2}=\frac{f_A\left(\tilde\mu^{[o]}-1\right)}{\left(\tilde\mu^{[o]}q+2\right)(r-1)}=\frac{\left(r-\tilde\mu^{[o]}\right)}{5\tilde\mu^{[o]}(r-1)},$$

$$[d]+r[b]=\frac{r\left(\tilde\mu^{[o]}-1\right)}{5\tilde\mu^{[o]}(r-1)}+r\frac{\left(r-\tilde\mu^{[o]}\right)}{5\tilde\mu^{[o]}(r-1)}=\frac{r}{5\tilde\mu^{[o]}};\ [d]^2-r[b]^2=\frac{\left(\tilde\mu^{[o]2}-r\right)r}{25\tilde\mu^{[o]2}(r-1)},$$

$$[a]=\frac{[d]^2-r[b]^2}{[d]+r[b]}=\frac{\tilde\mu^{[o]2}-r}{5\tilde\mu^{[o]}(r-1)};\ [k]=\frac{f_A\left(r-\tilde\mu^{[o]}\right)+f_Br\left(1-\tilde\mu^{[o]}\right)}{5\tilde\mu^{[o]}(r-1)}=f_A[b]-f_B[d].$$

Similarly (note that compared with the series $[x]$, f_A and p interchange with f_B and q):

$$]d[=\frac{f_Ar}{3\tilde\mu^{]o[}+2r}=\frac{f_Ar\left(\tilde\mu^{]o[}-1\right)}{(p+2)\tilde\mu^{]o[}(r-1)}=\frac{r\left(\tilde\mu^{]o[}-1\right)}{5\tilde\mu^{]o[}(r-1)},$$

$$]b[=\frac{f_B}{3\tilde\mu^{]o[}+2}=\frac{f_B\left(\tilde\mu^{]o[}-1\right)}{\left(\tilde\mu^{]o[}p+2\right)(r-1)}=\frac{\left(r-\tilde\mu^{]o[}\right)}{5\tilde\mu^{]o[}(r-1)},$$

$$]d[+r]b[=\frac{r\left(\tilde\mu^{]o[}-1\right)}{5\tilde\mu^{]o[}(r-1)}+r\frac{\left(r-\tilde\mu^{]o[}\right)}{5\tilde\mu^{]o[}(r-1)}=\frac{r}{5\tilde\mu^{]o[}};\ ]d[^2-r]b[^2=\frac{\left(\tilde\mu^{]o[2}-r\right)r}{25\tilde\mu^{]o[2}(r-1)},$$

$$]a[=\frac{]d[^2-r]b[^2}{]d[+r]b[}=\frac{\tilde\mu^{]o[2}-r}{5\tilde\mu^{]o[}(r-1)};\ ]k[=\frac{f_B\left(r-\tilde\mu^{]o[}\right)+f_Ar\left(1-\tilde\mu^{]o[}\right)}{5\tilde\mu^{]o[}(r-1)}=f_B]b[-f_A]d[.$$

Suppression of the common denominator in each equation finally yields the coefficient set:

$$[a] = \tilde{\mu}^{[o]2} - r,\ [b] = r - \tilde{\mu}^{[o]},\ [d] = r\left(\tilde{\mu}^{[o]} - 1\right),\ [k] = f_A[b] - f_B[d], \tag{B.2a}$$

$$]a[= \tilde{\mu}^{]o[2} - r,\ ]b[= r - \tilde{\mu}^{]o[},\ ]d[= r\left(\tilde{\mu}^{]o[} - 1\right),\ ]k[= f_B]b[- f_A]d[, \tag{B.2b}$$

from which it comes (for $[d]]a[+ [a]]b[\neq 0$):

$$\beta = \frac{[b]]a[+ [a]]d[}{[d]]a[+ [a]]b[} = \frac{\left(r - \tilde{\mu}^{[o]}\right)\left(\tilde{\mu}^{]o[2} - r\right) + r\left(\tilde{\mu}^{]o[} - 1\right)\left(\tilde{\mu}^{[o]2} - r\right)}{r\left(\tilde{\mu}^{[o]} - 1\right)\left(\tilde{\mu}^{]o[2} - r\right) + \left(r - \tilde{\mu}^{]o[}\right)\left(\tilde{\mu}^{[o]2} - r\right)}, \tag{B.3a}$$

$$\alpha = \frac{[a]]k[- [k]]a[}{[d]]a[+ [a]]b[} = f_B\frac{[a]]b[+ [d]]a[}{[d]]a[+ [a]]b[} - f_A\frac{[a]]d[+ [b]]a[}{[d]]a[+ [a]]b[} = -f_A\beta + f_B. \tag{B.3b}$$

The calculation of the coefficient β suffices. The Eq. (B.1) also yield:

$$\tilde{\mu}^{[o]2} - r = 2\tilde{\mu}^{[o]}P + \frac{r}{3}\left(\tilde{\mu}^{[o]}q - 1\right) = \frac{1}{3}\left(p(\tilde{\mu}^{[o]} - r) + qr(\tilde{\mu}^{[o]} - 1)\right), \tag{B.4a}$$

$$\tilde{\mu}^{]o[2} - r = 2\tilde{\mu}^{]o[}Q + \frac{r}{3}\left(\tilde{\mu}^{]o[}p - 1\right) = \frac{1}{3}\left(q(\tilde{\mu}^{]o[} - r) + pr(\tilde{\mu}^{]o[} - 1)\right), \tag{B.4b}$$

such that:

$$\begin{aligned}
\beta &= \frac{[b]]a[+ [a]]d[}{[d]]a[+ [a]]b[} = \frac{\left(r - \tilde{\mu}^{[o]}\right)\left(\tilde{\mu}^{]o[2} - r\right) + r\left(\tilde{\mu}^{]o[} - 1\right)\left(\tilde{\mu}^{[o]2} - r\right)}{r\left(\tilde{\mu}^{[o]} - 1\right)\left(\tilde{\mu}^{]o[2} - r\right) + \left(r - \tilde{\mu}^{]o[}\right)\left(\tilde{\mu}^{[o]2} - r\right)} \\
&= \frac{\left(r - \tilde{\mu}^{[o]}\right)\left(q(\tilde{\mu}^{]o[} - r) + pr(\tilde{\mu}^{]o[} - 1)\right) + r\left(\tilde{\mu}^{]o[} - 1\right)\left(p(\tilde{\mu}^{[o]} - r) + qr(\tilde{\mu}^{[o]} - 1)\right)}{r\left(\tilde{\mu}^{[o]} - 1\right)\left(q(\tilde{\mu}^{]o[} - r) + pr(\tilde{\mu}^{]o[} - 1)\right) + \left(r - \tilde{\mu}^{]o[}\right)\left(p(\tilde{\mu}^{[o]} - r) + qr(\tilde{\mu}^{[o]} - 1)\right)} \\
&= \frac{(r - O)(q(X - r) + pr(X - 1)) + r(X - 1)(p(O - r) + qr(O - 1))}{r(O - 1)(q(X - r) + pr(X - 1)) + (r - X)(p(O - r) + qr(O - 1))} \\
&= \frac{q\left(r^2(X - 1)(O - 1) - (X - r)(O - r)\right)}{p\left(r^2(X - 1)(O - 1) - (X - r)(O - r)\right)} = \frac{q}{p} = \frac{3f_B - 2f_A}{3f_A - 2f_B}.
\end{aligned} \tag{B.5}$$

From β, it comes $\alpha = -f_A\frac{3f_B - 2f_A}{3f_A - 2f_B} + f_B\frac{3f_A - 2f_B}{3f_A - 2f_B} = \frac{2({f_A}^2 - {f_B}^2)}{3f_A - 2f_B} = \frac{2(f_A - f_B)}{3f_A - 2f_B}$.

The analytical resolution for P, P' then follows from $P(\alpha + \beta C)[a] + P[b] - (\alpha + \beta P)[d] = [k]$, what simply yields $P = \frac{-([b] + \alpha[a] - \beta[d]) \pm \sqrt{([b] + \alpha[a] - \beta[d])^2 + 4\beta[a](\alpha[d] + [k])}}{2\beta[a]}$.

Everything solved without difficulty to be reported, one finds:

$$\text{(1) For } f_A < 3/5,\ P = -\frac{1}{2}\left(\frac{4f_A - 5}{3 - 5f_A} + \sqrt{\left(\frac{4f_A - 5}{3 - 5f_A}\right)^2 - 4\frac{3f_A}{3 - 5f_A}\left(\frac{3 - 5f_A}{3 - 5f_A}\right)}\right), \tag{B.6a}$$

$$\text{(2) For } f_A > 3/5,\ P = -\frac{1}{2}\left(\frac{4f_A - 5}{3 - 5f_A} - \sqrt{\left(\frac{4f_A - 5}{3 - 5f_A}\right)^2 - 4\frac{3f_A}{3 - 5f_A}\left(\frac{3 - 5f_A}{3 - 5f_A}\right)}\right), \tag{B.6b}$$

$$\text{and } P' = \alpha + \beta P = \frac{2(f_A - f_B)}{3f_A - 2f_B} + \frac{3f_B - 2f_A}{3f_A - 2f_B}P = \frac{(2 - 4f_B) + (5f_B - 2)P}{3 - 5f_B}, \tag{B.7}$$

which are all shear contrast-independent quantities.

For $[d]]a[+ [a]]b[= 0$ (at $f_A = 0.4$), the solution comes from $([a]]k[- [k]]a[) + C([b]]a[+ [a]]d[) = 0$ say from Eq. (B.7), $P_{f_A=0.4} = (4f_B - 2)/(5f_B - 2) = 0.4 = P'_{f_B=0.4}$, and $P'_{f_A=0.4} = ([k] - 0.4[b])/(0.4[a] - [d])_{f_A=0.4} = 9/13 \approx 0.692 = P_{f_B=0.4}$, in the continuity of the plots of Fig. 7. All equations from the beginning still hold when the coefficient 2/5, as it appears in particular in p and q, is substituted with coefficient w. Thus, the solution for two-phase I-I elastic-like composites also holds for isotropic dielectric-like ones, in which case $w = 1/3$.

Appendix C. The Self-Consistent estimate for a two-phase I-C composite

Using Eq. (7) for the shear μ and bulk $\theta = \frac{3\lambda + 2\mu}{3} = \frac{2\mu}{3}\left(\frac{1+\nu}{1-2\nu}\right)$ moduli eigenvalues yields the two scalar coupled relations:

$$\mu^{*SC}_{[AB]} = \langle\mu\rangle + \left(\mu^{*SC}_{[AB]} - \mu^A\right) : t^{*ISC}_{\mu} : \left(\mu^{*SC}_{[AB]} - \mu^B\right);\ \theta^{*SC}_{[AB]} = \langle\theta\rangle + \left(\theta^{*SC}_{[AB]} - \theta^A\right) : t^{*ISC}_{\theta} : \left(\theta^{*SC}_{[AB]} - \theta^B\right). \tag{C.1}$$

With the two involved Green operator terms recalled in Eq. (32), they take the forms:

$$\mu^{*SC} = \langle\mu\rangle + \left(\mu^{*SC} - \mu_A\right)\left(\frac{2}{5\mu^{*SC}}\frac{6\mu^{*SC} + 3\theta^{*SC}}{4\mu^{*SC} + 3\theta^{*SC}}\right)\left(\mu^{*SC} - \mu_B\right), \tag{C.2a}$$

$$\theta^{*SC} = \langle\theta\rangle + \left(\theta^{*SC} - \theta_A\right)\left(\frac{3}{4\mu^{*SC} + 3\theta^{*SC}}\right)\left(\theta^{*SC} - \theta_B\right). \tag{C.2b}$$

From the bulk equation one obtains the relation:

$$\theta^{*SC} = \frac{4\langle\theta\rangle\mu^{*SC} + 3\theta_A\theta_B}{4\mu^{*SC} + 3\{\theta\}} = \langle\theta\rangle\frac{4\mu^{*SC} + 3\overline{\{\bar{\theta}\}}}{4\mu^{*SC} + 3\{\theta\}}, \tag{C.3}$$

that allows to first solving the shear equation of rank four in μ^{*SC} once θ^{*SC} is eliminated in it and then the bulk equation. Thanks to this I-C elastic *SC* solution that we do not need explicitly here, one can admit the ratio $z^*_{SC} = \theta^{*SC}/\mu^{*SC}$ be definite such as to write the two-equation system as:

$$\mu^{*SC} = \langle\mu\rangle + \left(\mu^{*SC} - \mu_A\right)\frac{w}{\mu^{*SC}}\left(\mu^{*SC} - \mu_B\right) \text{ with } w = \frac{2}{5}\frac{6 + 3z^*_{SC}}{4 + 3z^*_{SC}}, \tag{C.4a}$$

$$\theta^{*SC} = \langle\theta\rangle + \left(\theta^{*SC} - \theta_A\right)\frac{w'}{\theta^{*SC}}\left(\theta^{*SC} - \theta_B\right) \text{ with } w' = \frac{3z^*_{SC}}{4 + 3z^*_{SC}}. \tag{C.4b}$$

In the I-I case that corresponds to an infinite z^*_{SC} ratio, $w' \to 1$ but the bulk equation becomes undetermined and $w \to 2/5$, what results in the one-modulus I-I problem solved in Appendix B.

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