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Study of the Coefficients of Thermal Expansion of phases embedded in multiphase materials

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Abstract. The present paper describes a new approach developed in order to determine the Coefficients of Thermal Expansion (CTE) of phases embedded in two-phase materials. The procedure is based on the coupling of numerical simulations with scale transition models and experiments. A three scales one-site thermo-elastic self-consistent model following the formalism introduced by Kröner and Eshelby (KE) was extended to multiphase materials. Preliminary numerical computations justify to identify the CTE of a given phase embedded in a two-phase material with the CTE of the corresponding pure single-phase. This additional assumption was introduced in KE model. The implementation yields an explicit formulation for the unknown CTE of a phase embedded in a ($\alpha+\beta$) two-phases material. The application of this expression to the characterization of β thermal expansion properties implies the measurement or knowledge of several parameters. This approach was checked through the study of an Al-50%vol.-SiC-50%vol. MMC. The CTE of Silicon Carbide were determined as a function of the temperature. Simulated results obtained show a very good agreement with experimental values available in the literature.

Introduction.

The mechanical behaviour, performance [1] and durability [2] of materials are strongly related to the presence and the amount of residual stresses. During thermal processes, residual stresses appear under specific conditions. The magnitude and the sign of these stresses are strongly related to the Coefficients of Thermal Expansion of the grains and phases constituting the material. Numerous phases are very difficult to process under an acceptable non-porous single-phase state. It is the case of many advanced materials dedicated to industrial applications, as the β -phase of $\alpha+\beta$ Titanium alloys. But, this assertion is relevant for more common materials as carbide reinforced steels [3]. Thus, the characterization of the properties of phases embedded in multiphase material is a requirement for a more accurate prediction of the consequences of thermal loadings on the forming and service-life of the material. The CTE of a given phase, when it is embedded in a multiphase structure is studied in section 2-1. The crucial question of the deviation between the CTE of the single-phase and those of the same phase included in a heterogeneous matrix will be examined. According to a preceding study performed in pure elasticity [4], this groundwork will be used in order to implement the classical self-consistent thermo-elastic formalism. A solution will be proposed in order to characterize the CTE of a phase embedded in a multiphase material.

1. Self-Consistent estimates.

1.1 Constitutive equations. According to the original formalism introduced by Kröner in pure elasticity [5], Self-Consistent (SC) estimates are suitable to deal with the question of heterogeneous

materials such as polycrystals [6-7] or Metal Matrix Composites (MMC). The models are based on a statistical description of the material. SC approaches are particularly adapted to represent aggregates of crystalline grains exhibiting local variations of their properties due to the elastic and/or thermal heterogeneities occurring when the material is composed of two or more phases.

The material is split up into the classical three scales described in [8-9] :

- the macroscopic scale, denoted by the superscript ^I. The so-called Homogeneous Effective Medium (HEM) is assumed to have the average properties of the multiphase polycrystalline aggregate.
- the pseudo-macroscopic scale, denoted by the superscripts ^α, ^β and so on. It represents the average properties of the considered α (or β , ...) phase.
- The mesoscopic scale, denoted by the superscript ^{II}. The anisotropic properties of an inclusion of a given phase are exhibited at this level. The orientation of a crystallite is expressed by the letter Ω .

Let us consider a pure thermo-elastic sollicitation of the material. The thermal dilatation created by a temperature increment δT is considered as a stress-free strain. The stresses respectively induced in the grains (σ^{II}), the multiphase polycrystal (σ^I) and a particular phase e. g. α (σ^α) are linked to the strains (ϵ) as follows :

$$\sigma^{II} = \mathbf{c}^\alpha(\Omega) \cdot (\epsilon^{II} - \mathbf{m}^\alpha(\Omega) \delta T) \quad (1)$$

$$\sigma^I = \mathbf{C}^I \cdot (\epsilon^I - \mathbf{M}^I \delta T) \quad (2)$$

$$\sigma^\alpha = \mathbf{C}^\alpha \cdot (\epsilon^\alpha - \mathbf{M}^\alpha \delta T) \quad (3)$$

\mathbf{c}^α , \mathbf{C}^α and \mathbf{C}^I are the elastic stiffnesses, respectively at mesoscopic, pseudo-macroscopic and macroscopic scales, whereas \mathbf{m}^α , \mathbf{M}^α and \mathbf{M}^I denote the corresponding CTE tensors. $\mathbf{A} \cdot \mathbf{B}$ denotes the double scalar product $A_{ijkl} B_{klmn}$.

SC simulation framework is based on a scale transition formalism. For ellipsoidal inclusions, mesoscopic stresses and strains are uniform in an inclusion and linearly related to the macroscopic stresses and strains through the well-known interaction equation [10] :

$$\sigma^{II} - \sigma^I = -\mathbf{C}^I \cdot \mathbf{R}^I \cdot (\epsilon^{II} - \epsilon^I) \quad (4)$$

Where the so-called reaction tensor \mathbf{R}^I will be easily obtained from the knowledge of the morphology of the crystallites and the average properties of the HEM :

$$\mathbf{R}^I = (\mathbf{I} - \mathbf{S}_{esh}) \cdot \mathbf{S}_{esh}^{-1} = (\mathbf{C}^{I^{-1}} - \mathbf{E}) \cdot \mathbf{E}^{-1} \quad (5)$$

Hill's tensor \mathbf{E} expresses the local interactions depending on the morphology of the crystallites [11]. It is linked to Eshelby's tensor \mathbf{S}_{esh} by $\mathbf{E} = \mathbf{C}^I \cdot \mathbf{S}_{esh}$. \mathbf{I} is the fourth order identity tensor.

In another main paper, Hill demonstrated the following useful average equations [12] :

$$\langle \sigma^{II} \rangle = \sigma^I \text{ and } \langle \epsilon^{II} \rangle = \epsilon^I \quad (6)$$

The pseudo-macroscopic scale has been defined by the average on the crystallites of the studied phase. This entails (6) to be rewritten at this level through an extension of Hill's formulation :

$$\langle \sigma^{II} \rangle_\alpha = \sigma^\alpha \text{ and } \langle \epsilon^{II} \rangle_\alpha = \epsilon^\alpha \quad (7)$$

The average elastic stiffness and CTE of the polycrystal may be deduced from the well-known equations [13] :

$$\mathbf{C}^I = \left\langle \mathbf{c}^i(\Omega) \cdot \left[\mathbf{I} + \mathbf{E} \cdot \left[\mathbf{c}^i(\Omega) - \mathbf{C}^I \right]^{-1} \right] \right\rangle_{i=1, N} \quad (8)$$

$$\mathbf{M}^I = \left\langle \left[\mathbf{c}^i(\Omega) + \mathbf{C}^I \cdot \mathbf{R}^I \right]^{-1} \cdot \mathbf{C}^I \right\rangle_{i=1, N}^{-1} \cdot \left\langle \left[\mathbf{c}^i(\Omega) + \mathbf{C}^I \cdot \mathbf{R}^I \right]^{-1} \cdot \mathbf{c}^i(\Omega) \cdot \mathbf{m}^i(\Omega) \right\rangle_{i=1, N}^{-1} \quad (9)$$

Where N is the number of grains in the HEM. It has been demonstrated in a previous work [4] that the pseudo-macroscopic stiffness of a given α -phase embedded in a multiphase polycrystal could be expressed by :

$$\mathbf{C}^\alpha = \left\langle \mathbf{c}^\alpha(\Omega) \cdot \mathbf{A}^\alpha \right\rangle_\alpha \cdot \left\langle \mathbf{A}^\alpha \right\rangle_\alpha^{-1} \text{ with } \mathbf{A}^\alpha = \left\langle \left[\mathbf{E} \cdot \left\{ \mathbf{c}^\alpha(\Omega) - \mathbf{C}^I \right\} + \mathbf{I} \right]^{-1} \right\rangle_\alpha \quad (10)$$

In consequence, the stiffness tensor at any scale will be determined independently from the thermal expansion properties of the material. Nevertheless, it should be remembered that, in metallic materials, the elastic stiffness often varies with the temperature.

In the following, only pure thermal loading will be considered so that, the material will be submitted to a stress-free macroscopic state. It is assumed that no macro or mesoscopic plastic deformation takes place during this process. Hence, from $\boldsymbol{\sigma}^I = \mathbf{0} = \mathbf{C}^I \cdot (\boldsymbol{\varepsilon}^I - \mathbf{M}^I \delta T)$, the macroscopic strain can be written as :

$$\boldsymbol{\varepsilon}^I = \mathbf{M}^I \delta T \quad (11)$$

Taking into account the condition (11) in (1) reduces the mesoscopic strains to :

$$\boldsymbol{\varepsilon}^II = \left[\mathbf{c}^\alpha(\Omega) + \mathbf{C}^I \cdot \mathbf{R}^I \right]^{-1} \cdot \left[\mathbf{C}^I \cdot \mathbf{R}^I \cdot \mathbf{M}^I + \mathbf{c}^\alpha(\Omega) \cdot \mathbf{m}^\alpha(\Omega) \right] \delta T \quad (12)$$

For each temperature increment δT , (8-9) will provide the required macroscopic elastic stiffness and Coefficients of Thermal Expansion from the knowledge of the single-crystal elastic and thermal properties. The remaining unknown interaction tensor is then deduced from \mathbf{C}^I according to (5). The mechanical state of each grain can be completely characterized for each δT through (12).

1.2 Derivation of SC modeling for the simulation of pseudo-macroscopic CTE. Our purpose is to find an expression for the CTE of a phase embedded in a multiphase material, according to the classical SC model introduced in section 1-1. Let us consider the case of a ($\alpha+\beta$) two-phases material. α -phase elastic and thermal properties will be supposed to be perfectly identified, whereas β -phase CTE are the unknowns. In this paper, the pseudo-macroscopic elastic stiffness of the β -phase will be assumed to be known from the literature or previously determined according to the method given by Fréour et al. in [4].

Introducing the volume fractions f^α and f^β , the average stress relations (6-7) may be rewritten as a function of the pseudo-macroscopic values. Then, taking account of (3) and (11) yields the relation :

$$\boldsymbol{\sigma}^I = \left\langle \boldsymbol{\sigma}^II \right\rangle_\alpha + \left\langle \boldsymbol{\sigma}^II \right\rangle_\beta = f^\alpha \boldsymbol{\sigma}^\alpha + f^\beta \boldsymbol{\sigma}^\beta = f^\alpha \mathbf{C}^\alpha \cdot (\boldsymbol{\varepsilon}^\alpha - \mathbf{M}^\alpha \delta T) + f^\beta \mathbf{C}^\beta \cdot (\boldsymbol{\varepsilon}^\beta - \mathbf{M}^\beta \delta T) = 0 \quad (13)$$

Thus, β CTE tensor satisfies :

$$\mathbf{M}^\beta = \frac{f^\alpha}{f^\beta \delta T} \mathbf{C}^{\beta-1} \cdot \mathbf{C}^\alpha \cdot \boldsymbol{\varepsilon}^\alpha - \frac{f^\alpha}{f^\beta} \mathbf{C}^{\beta-1} \cdot \mathbf{C}^\alpha \cdot \mathbf{M}^\alpha + \frac{1}{\delta T} \boldsymbol{\varepsilon}^\beta \quad (14)$$

While β CTE remain unknown, the main difficulty is that the pseudo-macroscopic strain in β could not be evaluated from the classical method described in the second part and involving the application of (7) consecutively to (12). Our basic hypothesis on the macroscopic stress raises more convenient simplifications. Let us integrate (11) in the strain average relation (7). ε^α and ε^β are linked through the following formula : $f^\alpha \varepsilon^\alpha + f^\beta \varepsilon^\beta = \varepsilon^I = \mathbf{M}^I \delta T$.

The expression obtained for ε^β yields the following simplified form for \mathbf{M}^β :

$$\mathbf{M}^\beta = \frac{f^\alpha}{f^\beta} \left[\mathbf{C}^{\beta-1} \dots \mathbf{C}^\alpha - \mathbf{I} \right] \cdot \left(\frac{1}{\delta T} \varepsilon^\alpha \right) + \frac{1}{f^\beta} \mathbf{M}^I - \frac{f^\alpha}{f^\beta} \mathbf{C}^{\beta-1} \dots \mathbf{C}^\alpha \dots \mathbf{M}^\alpha \quad (15)$$

The values taken by \mathbf{M}^β components should not depend on any state variable as the pseudo-macroscopic strain or the temperature. It will be demonstrated in the following that (15) satisfies this criterion. Let us develop the mesoscopic strain in a given α -grain (12). The pseudo-macroscopic strain in α results from the average relation (7), so that the ratio $\frac{1}{\delta T} \varepsilon^\alpha$ will be expressed as follows :

$$\frac{1}{\delta T} \varepsilon^\alpha = \left\langle \left[\mathbf{c}^\alpha(\Omega) + \mathbf{C}^I \dots \mathbf{R}^I \right]^{-1} \right\rangle \cdot \left(\mathbf{C}^I + \mathbf{C}^I \dots \mathbf{R}^I \right) \cdot \mathbf{M}^I + \left\langle \left[\mathbf{c}^\alpha(\Omega) + \mathbf{C}^I \dots \mathbf{R}^I \right]^{-1} \right\rangle \cdot \left[\mathbf{c}^\alpha(\Omega) \cdot \mathbf{m}^\alpha(\Omega) - \mathbf{C}^I \dots \mathbf{M}^I \right] \quad (16)$$

This ratio only depends on the constants describing the thermo-mechanical properties of the material. Thus, according to (16) any state variable is excluded from expression (15) obtained for the pseudo-macroscopic CTE of β . Once f^α , f^β , \mathbf{c}^α , \mathbf{C}^α , \mathbf{C}^β , \mathbf{C}^I , \mathbf{M}^I , \mathbf{m}^α and \mathbf{M}^α known, e.g. from measurements or calculation, one will easily calculate the pseudo-macroscopic CTE of any given β -phase owing to (15) and (16). In consequence, in spite of the assumptions concerning the macroscopic sollicitation considered for the needs of the demonstration, these forms are general and will be satisfied whatever the mechanical state of the system.

Now, the case of the pseudo-macroscopic elastic stiffnesses and CTE should be carefully examined. In theory, (10) demonstrate the potential existence of a deviation between the elastic stiffness of a phase embedded in a multiphase material and the corresponding single-phase stiffness. This point has been developed in [4]. In fact, in most cases, it was numerically justified that pseudo-macroscopic elastic stiffnesses could be identified with the single phase elasticity constants. However, a priori, it is not possible to predict anything about the question of pseudo-macroscopic CTE. Thus, we will discuss about this point in the following section.

2. Application of SC model to the study of pseudo-macroscopic CTE

2.1 Analysis of the deviation between pseudo-macroscopic and single-phase CTE. In order to quantify the deviation between pure single-phases CTE and the corresponding values for the same phase when it is embedded in a multiphase material, it is first necessary to express the relation ruling thermo-elastic interactions existing between a phase and the polycrystalline matrix. Considering the case of α -phase, and according to (4), it comes :

$$\sigma^\alpha - \sigma^I = -\mathbf{C}^I \dots \mathbf{R}^I \cdot (\varepsilon^\alpha - \varepsilon^I) \quad (17)$$

In the case when the material is not subjected to any macroscopic stress, the integration of the conditions (2, 3, 11) in (17), followed by basic tensorial calculations, lead to a simplified form for the CTE of a given α -phase embedded in a multiphase material :

$$\mathbf{M}^\alpha = \frac{1}{\delta T} \varepsilon^\alpha + \frac{1}{\delta T} \mathbf{C}^{\alpha-1} \dots \mathbf{C}^I \dots \mathbf{R}^I \cdot \varepsilon^\alpha - \mathbf{C}^{\alpha-1} \dots \mathbf{C}^I \dots \mathbf{R}^I \cdot \mathbf{M}^I \quad (18)$$

It should be remembered that equation (16) yields the simplification of the state variables, so that M^α only depends on the thermal and mechanical constants of the material. SC formalism demonstrates that the average CTE of a given phase should not be the same in a pure single-phase material and in a multiphase polycrystal, due to the interphase interactions revealed by the terms C^I , R^I and M^I which depend on the thermo-elastic properties of the others phases constituting the structure.

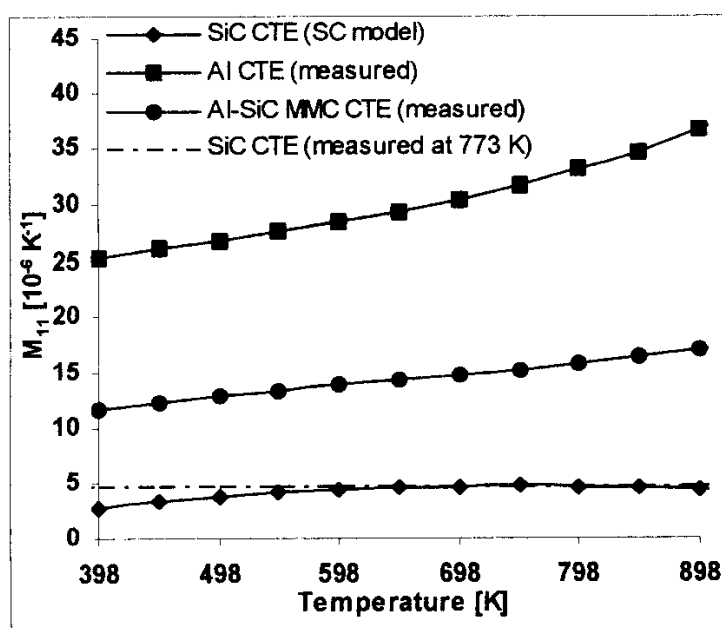
Equation (18) was applied to several two-phases material chosen to evaluate the influence on pseudo-macroscopic CTE of the listed parameters : single crystal elastic anisotropy, single crystal thermal expansion anisotropy, heterogeneities of single-phase elastic stiffnesses, and heterogeneities of single-phase CTE. Results are detailed in table 1. In most cases, pseudo-macroscopic CTE deviates from the single-phases values from less than 1%. In consequence, in spite of the expression demonstrated in (18), numerical computations justifies the identification of pseudo-macroscopic CTE to single-phase CTE.

Material	Considered phase	Single-crystal Elastic Anisotropy	Single-crystal CTE anisotropy	Single-phase elastic stiffness heterogeneity	Single-phase CTE heterogeneity	Maximum deviation between single-phase CTE and the corresponding pseudo-macroscopic CTE
Zr-Ti	α -Zr	Weak	Medium	Weak	Strong	1.2%
	α -Ti	Weak	Weak			<< 1%
Fe-Cu	α -Fe	Medium	None	Medium	Weak	<< 1%
	Cu	Strong	None			<< 1%
Al-Zn	Al	Weak	None	Strong	Strong	<< 1%
	Zn	Strong	Strong			2.4%

Table 1 : Qualitative and quantitative study of the deviation between pseudo-macroscopic and single-phase CTE.

This additional assumption implies to neglect interphase interactions. As a matter of fact, it has two major consequences. Firstly, the model reduces to the very classical “one inclusion per phase” SC model. Secondly this hypothesis provides the knowledge of the remaining unknowns in (15). Hence, this assumption should be considered as an enhancement of the model, because it ensures the possibility to characterize the thermal properties of a phase embedded in a two-phase material through (15). Nevertheless, the validity of the additional hypothesis should be checked a posteriori.

2.2 Determination of SiC CTE from experiments performed on a two-phases MMC. Silicon Carbide (SiC) is extremely difficult to obtain in satisfying single-phase samples.



Sintered samples include porosities which could be considered as a strongly heterogeneous secondary phase, due to its infinite compliance. Several papers demonstrate the main effects of the porosity on the mechanical behaviour of an extensive variety materials : concrete, metals and so on [15-17]. In terms of thermal expansion, it is almost sure that the average behaviour of a hypothetical bulk SiC polycrystal would not be the same than the one of a mixture SiC-porosities. In consequence, the study of SiC CTE should be preferably achieved on a non-porous two-phase sample, according to the formalism previously developed in this work.

Figure 2 : Predicted and experimental [14] evolution of the CTE in a 50 vol.% SiC_p/Al MMC.

Analysis of the evolution of the CTE of Al_{50% vol}-SiC_{50% vol} and pure aluminium polycrystals are available in the literature [14]. The required data has been introduced in SC simulation scheme as detailed in the previous section. Figure 2 illustrates the curves obtained at macroscopic and pseudo-macroscopic scales in the composite. A horizontal line corresponding to SiC CTE measured around 773 K is given as a comparison [18].

SC model predicts values in good accordance with the experiments in a wide temperature range [648 K-848 K]. Moreover, the results agrees with some conventional properties attributed to ceramics e. g. : a weak variation of their CTE with the temperature.

This study confirms the validity of approaches coupling experiments to self-consistent models in order to characterize the thermal expansion of phases embedded in two-phases polycrystals.

Conclusions

Thermo-elastic self-consistent scale transition formalism was extended to the case of two-phases materials in a more convenient way. The solution developed takes into account the pseudo-macroscopic scale which figures the average properties of the different phases constituting the material. An additional assumption was considered. Theory predicts that pseudo-macroscopic Coefficients of Thermal Expansions may deviate from the single-phase corresponding values. Numerical computations justifies, however, the identification of these tensors. This hypothesis provides an original method for the characterization of the CTE of a phase embedded in a two-phase material. This method implies the coupling of experimental techniques and SC scale transition modeling. It was checked through the determination of the CTE of the Silicon Carbide included in aluminium based matrix. The simulations agree with values previously published in the literature. This work raises several perspectives. In further studies, SC model will be formulated in order to express the CTE of a phase directly from the pseudo-macroscopic strains measured on the diffracting volume of the material. XRD being extremely sensitive to texture effects, this alternative approach should be more pertinent to treat the case of rolled samples, for example.

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