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# Efficient Evaluation of the Material Response of Tissues Reinforced by Statistically Oriented Fibres

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## 1 Abstract

2 For several classes of soft biological tissues, modelling complexity is in part due to the arrangement  
3 of the collagen fibres. In general, the arrangement of the fibres can be described by defining, at  
4 each point in the tissue, the structure tensor (i.e., the tensor product of the unit vector of the local  
5 fibre arrangement by itself) and a probability distribution of orientation. In this approach, assuming  
6 that the fibres do not interact with each other, the overall contribution of the collagen fibres to  
7 a given mechanical property of the tissue can be estimated by means of an averaging integral of  
8 the constitutive function describing the mechanical property at study over the set of all possible  
9 directions in space. Except for the particular case of fibre constitutive functions that are polynomial  
10 in the transversely isotropic invariants of the deformation, the averaging integral cannot be evaluated  
11 directly, in a single calculation because, in general, the integrand depends both on deformation and  
12 on fibre orientation in a non-separable way. The problem is thus, in a sense, analogous to that of  
13 solving the integral of a function of two variables, which cannot be split up into the product of two  
14 functions, each depending only on one of the variables. Although numerical schemes can be used  
15 to evaluate the integral at each deformation increment, this is computationally expensive. With the  
16 purpose of containing computational costs, this work proposes approximation methods that are based  
17 on the direct integrability of polynomial functions and that do not require the step-by-step evaluation  
18 of the averaging integrals. Three different methods are proposed: *a*) a Taylor expansion of the fibre  
19 constitutive function in the transversely isotropic invariants of the deformation; *b*) a Taylor expansion  
20 of the fibre constitutive function in the structure tensor; *c*) for the case of a fibre constitutive function  
21 having a polynomial argument, an approximation in which the directional average of the constitutive  
22 function is replaced by the constitutive function evaluated at the directional average of the argument.  
23 Each of the proposed methods approximates the averaged constitutive function in such a way that it is  
24 multiplicatively decomposed into the product of a function of the deformation only and a function of  
25 the structure tensors only. In order to assess the accuracy of these methods, we evaluate the constitutive  
26 functions of the elastic potential and the Cauchy stress, for a biaxial test, under different conditions,  
27 i.e., different fibre distributions and different ratios of the nominal strains in the two directions. The  
28 results are then compared against those obtained for an averaging method available in the literature,  
29 as well as against the integration made at each increment of deformation.

30 **Keywords:** biological tissue, collagen, fibre-reinforced, structure tensor, fabric tensor, averaging, Finite  
31 Element Method, Continuum Mechanics, Elasticity

## 32 1. Introduction

33 Soft biological tissues can be seen as highly complex fibre-reinforced materials [1]. The solid phase  
 34 can be represented by a mixture of an isotropic matrix and transversely isotropic fibres. The spatial  
 35 arrangement of the fibres largely defines the anisotropy and inhomogeneity of the tissue (e.g., [2, 3]).  
 36 In some tissues, the fibres can be thought of as being arranged in a finite number of families, each  
 37 family being determined by the common direction of the fibres belonging to it. For instance, tissues  
 38 typically modelled with a single fibre family are ligaments and tendons [4], and tissues with two fibre  
 39 families are blood vessels [5, 6] and the atrium of the heart [7, 8].

40 However, the fibres usually have some dispersion with respect to the dominant direction(s) (e.g.,  
 41 [6]). Moreover, there are tissues in which the dominant direction changes with location within the tissue  
 42 or in which a dominant direction cannot be clearly defined. A prime example is articular cartilage,  
 43 in which the fibre orientation varies along the depth of the tissue, from parallel to the surface in  
 44 the superficial zone, to random in the middle zone (no dominant direction), to aligned to the depth  
 45 direction in the deep zone [9, 10]. Whenever one wishes to consider the dispersion about the dominant  
 46 direction(s) or tissues with more complex fibre orientations, it is necessary to describe the arrangement  
 47 of the fibres by means of an infinite number of statistically oriented fibres, which requires the use of  
 48 an orientation probability distribution.

49 Orientation probability distributions in Soft Tissue Biomechanics were first used by Lanir [11],  
 50 and later adopted by several researchers (e.g., [12, 13, 14, 6]). Similar techniques were independently  
 51 developed in the context of composite materials with inclusions [15, 16], and were subsequently trans-  
 52 ferred to biomechanical problems such as the determination of the overall elastic properties or the  
 53 overall permeability of soft tissues. These models were extended to the case of large deformations, at  
 54 first for the elasticity alone [17] and then for both elasticity and permeability [18]. Here, we shall use  
 55 the notation and concepts developed in these previous works.

56 For an extensive physical quantity, such as mass, momentum, energy, etc, the overall extent  $q$  of  
 57 the quantity associated with the mixture as a whole is obtained as the weighted sum

$$q = \sum_{\alpha} \phi_{\alpha} q_{\alpha}, \quad (1)$$

58 where  $q_{\alpha}$  is the value of the quantity in the constituent  $\alpha$  and  $\phi_{\alpha}$  is the volumetric fraction of the  
 59 constituent  $\alpha$ . For lack of better knowledge, this *rule-of-mixture* can be extended also to quantities,  
 60 such as the permeability, whose overall value may or may not be a linear combination as in Equation  
 61 (1). We are interested in mixtures including one or more fibre families, each having statistical orienta-  
 62 tion. The fibres in each family share the same properties but have different orientation, described by a  
 63 probability distribution. Therefore, we think of each fibre family as an infinity of fibres, and evaluate  
 64 its overall contribution by means of an integral over all directions in space. The overall contribution  
 65 of each fibre family to a certain physical quantity, given by the averaging integral of the quantity, is  
 66 called *fibre ensemble*.

67 The method proposed in [6], which we call GOH method (Gasser-Ogden-Holzapfel method),  
 68 accounts for the overall effect of each family of statistically oriented fibres by means of the directional  
 69 average of the material structure tensor (the tensor product of the unit vector representing the material  
 70 fibre direction by itself). In their approach to the overall elastic properties of the arterial wall, after  
 71 having defined a fibre elastic potential as a function of the structure tensor of a given direction, Gasser  
 72 et al. [6] replaced the structure tensor by its directional average. The rule-of-mixture method gives the  
 73 same results of the GOH method whenever the material property to be averaged is an affine function  
 74 (i.e., a constant plus a linear function) in the structure tensor [17]. The GOH method has the advantage  
 75 of requiring one single integration, directly. Indeed, once the probability distribution is known, the  
 76 directional average of the structure tensor is a given tensor that has to be evaluated only once, and  
 77 then used in all subsequent calculations. This makes the Finite Element (FE) implementation of the  
 78 GOH method quite straightforward and, indeed, the GOH method is available in the material libraries  
 79 of the commercially available software ABAQUS (Dassault Systèmes, Vélizy-Villacoublay, France).

80 In general, in the FE implementation of our rule-of-mixture method, the fibre ensemble (averaging  
 81 integral of a certain physical quantity) must be calculated at each increment of deformation [17, 19].  
 82 This is because of the coupled dependence of the integrand from *both* the structure tensor *and* the  
 83 deformation.

84 Although sometimes fairly expensive from the computational point of view, an efficient numerical  
 85 implementation of this method has been proposed [20], based on the use of spherical  $t$ -designs [21],  
 86 in which the surface of the unit sphere is discretised into a suitable set of points, and the integral is  
 87 evaluated as a summation on the discretised set of points. We recall that, since the oriented segment  
 88 joining the centre of the unit sphere with a given point on its surface defines univocally a direction  
 89 in space, the integration over the spherical surface can be made equivalent to integrating over all  
 90 directions in space. Other numerical methods for finding the integration points on the surface of the  
 91 sphere could be used (e.g., [22, 23, 24]), and a description of some of these methods can be found in  
 92 [25]. However, a single, direct integration is possible whenever the integrand is a *separable function*  
 93 of the deformation and the structure tensor, as is the case for *tensor-power polynomial* functions of  
 94 the structure tensor (the definition of *tensor-power polynomial* is given later, in Section 2.3). We note  
 95 that the GOH method is obtained in the instance of a tensor-power polynomial of degree one, which  
 96 is an affine function of the structure tensor.

97 In this work, based on the direct integrability of polynomial functions, we introduce and com-  
 98 pare three possible direct methods of approximation of the averaging integrals, with the purpose of  
 99 estimating their accuracy, and establishing the ranges within which they perform as alternative op-  
 100 tions to step-by-step integration criteria, while being computationally cheaper. We refer to these three  
 101 methods as:

- 102 1. INEX (*Invariant Expansion*): the function to be averaged is viewed as a function of the invariants  
 103 of the deformation that include the structure tensor, and then expanded in Taylor series about  
 104 the values of the invariants in the reference configuration; then, the resulting polynomial is  
 105 integrated;
- 106 2. STEX (*Structure Tensor Expansion*): the function to be averaged is expanded in Taylor series  
 107 about the structure tensor of a convenient direction, and the resulting (tensor-power) polynomial  
 108 is integrated;
- 109 3. PARG (*Polynomial Argument*): the function to be averaged is given by some function of an  
 110 argument that is a (tensor-power) polynomial in the structure tensor, and the average is taken  
 111 of the polynomial rather than of the whole function; in other words, the average is taken of  
 112 the “outermost” argument that can be written as a (tensor-power) polynomial in the structure  
 113 tensor.

114 These three methods are also compared with methods available in the literature:

- 115 4. GOH (*Gasser-Odgen-Holzappel*): the model proposed by Gasser et al. [6]; the GOH method can  
 116 be seen as the extreme of our PARG method, in which the “innermost” argument is averaged:  
 117 the structure tensor;
- 118 5. FESD (*Fibre Ensemble with Spherical Designs*): the step-by-step integration of the fibre ensemble  
 119 of a certain physical quantity performed with the method of the spherical  $t$ -designs [20, 26].

120 The comparison is made based on a benchmark test in the context of elasticity, namely a biaxial  
 121 tension test of a fibre-reinforced tissue sample, and the limitations of each methods are discussed.

## 122 2. Theoretical Background

123 We refer the Reader to the Appendix, where we briefly review the fairly standard Continuum Me-  
 124 chanics notation that we use (Appendix A), recall the definitions of the invariants of the deformation  
 125 for the cases of isotropy and transverse isotropy (Appendix B), as well as some basic relations in non-  
 126 linear hyperelasticity (Appendix C), which will serve as our example of application of the averaging  
 127 methods proposed in Section 3.

128 The notation follows that in a previous work [19], with a few small exceptions that allow for a  
 129 lighter notation. The reference configuration of a body is denoted  $\mathcal{B}$  (rather than  $\mathcal{B}_R$ ), the referential  
 130 volumetric fraction of constituent  $\alpha$  of a mixture is denoted  $\Phi_\alpha$  (rather than  $\phi_{\alpha R}$ ), and the referential  
 131 probability distribution of orientation of the fibres is denoted  $\Psi$  (rather than  $\psi$ ).

132 In this section, we first recall the volumetric-distortional decomposition of the deformation, which  
 133 we use for a purpose different than the usual one (quasi-incompressible materials), and introduce  
 134 some definitions that are useful for the objectives of this work. Then, we introduce some important  
 135 definitions in tensor algebra, and elucidate the averaging method based on the rule of mixtures that  
 136 we employ in this work, and that gives rise to what we call the fibre ensemble. Finally, we recall the  
 137 method by Gasser et al. [6] (GOH Method), to which we compare our results.

## 138 2.1. The Volumetric-Distortional Decomposition of the Deformation

139 The volumetric-distortional decomposition of the deformation gradient  $\mathbf{F}$  [27, 28, 29] is often employed  
 140 in the treatment of quasi-incompressible materials. However, we shall use it for a different purpose,  
 141 as outlined in Section 3.1. The deformation gradient tensor  $\mathbf{F}$  can be decomposed into its volumetric  
 142 and distortional (or isochoric) part,  $\mathbf{F} = J^{1/3}\bar{\mathbf{F}}$ . We refer to  $\bar{\mathbf{F}}$  as to the distortional (or isochoric)  
 143 part of  $\mathbf{F}$ , since, by construction, it is characterised by having a unitary determinant, i.e.,  $\det \bar{\mathbf{F}} = 1$ .  
 144 Consistently, we decompose the right Cauchy-Green deformation tensor as  $\mathbf{C} = J^{2/3}\bar{\mathbf{C}}$ , where the  
 145 isochoric part of  $\mathbf{C}$  is given by  $\bar{\mathbf{C}} = \bar{\mathbf{F}}^T \cdot \bar{\mathbf{F}}$  and satisfies the equality  $\det \bar{\mathbf{C}} = 1$ .

## 146 2.2. Some Important Definitions in Tensor Algebra

147 Here, we introduce some definitions for the case of material tensors but, naturally, these are analogous  
 148 for the case of spatial tensors. We indicate the full contraction of a material “contravariant” tensor  
 149  $\mathbb{T}$  and a material “covariant” tensor  $\mathbb{Z}$  of the same order  $r$  by means of the bra-ket notation  $\langle \mathbb{T} | \mathbb{Z} \rangle =$   
 150  $\mathbb{T}^{A_1 \dots A_r} \mathbb{Z}_{A_1 \dots A_r}$ . Note that the bra-ket notation can be used symmetrically, i.e.,  $\langle \mathbb{T} | \mathbb{Z} \rangle = \langle \mathbb{Z} | \mathbb{T} \rangle$ . For  
 151 the particular case of second-order tensors, we can alternatively write  $\mathbf{T} : \mathbf{Z} \equiv \langle \mathbf{T} | \mathbf{Z} \rangle = T^{AB} Z_{AB}$  and  
 152 we call  $\mathbf{T} : \mathbf{Z}$  the double contraction of  $\mathbf{T}$  and  $\mathbf{Z}$ .

153 Given any  $n$  material tensors  $\mathbb{A}_1, \dots, \mathbb{A}_n$  of the same “contravariant” order  $r$ , “covariant” order  $s$ ,  
 154 and overall order  $r + s$ , the *major-symmetric part* of the  $n(r + s)$ -th order tensor

$$\mathbb{T} = \mathbb{A}_1 \otimes \dots \otimes \mathbb{A}_n \quad (2)$$

155 is given by

$$\text{msym}(\mathbb{T}) = \frac{1}{n!} \sum_{\sigma \in \mathfrak{S}_n} \mathbb{A}_{\sigma_1} \otimes \dots \otimes \mathbb{A}_{\sigma_n}, \quad (3)$$

156 where each  $\sigma = \{\sigma_1, \dots, \sigma_n\}$  is one of all the  $n!$  possible permutations  $\mathfrak{S}_n$  of  $\{1, \dots, n\}$ . Note that,  
 157 if the tensors  $\mathbb{A}_i$  are of the first order (i.e., they are all vectors  $\mathbf{W}_i$  or all covectors  $\mathbf{\Pi}_i$ ), then the  
 158 major-symmetric part of  $\mathbb{T}$  coincides with its symmetric part.

159 For any material tensor  $\mathbb{A}$  (of any “contravariant” order  $r$ , “covariant” order  $s$ , and overall order  
 160  $r + s$ ), its  $n$ -th tensor power is defined as the  $n(r + s)$ -th order tensor

$$\mathbb{A}^{\otimes n} = \underbrace{\mathbb{A} \otimes \dots \otimes \mathbb{A}}_{n \text{ times}}, \quad (4)$$

161 and, by convention, we set  $\mathbb{A}^{\otimes 1} = \mathbb{A}$  and  $\mathbb{A}^{\otimes 0} = 1 \in \mathbb{R}$ . Given two tensors  $\mathbb{A}, \mathbb{B}$  (of the same “con-  
 162 travariant” order  $r$ , “covariant” order  $s$ , and overall order  $r + s$ ), the binomial tensor power  $(\mathbb{A} + \mathbb{B})^{\otimes n}$   
 163 is given by the generalised Newton’s formula

$$(\mathbb{A} + \mathbb{B})^{\otimes n} = \sum_{k=0}^n \left[ \binom{n}{k} \text{msym}(\mathbb{A}^{\otimes(n-k)} \otimes \mathbb{B}^{\otimes k}) \right], \quad (5)$$

164 where we recall that  $\binom{n}{k}$  is the binomial coefficient  $\binom{n}{k} = n! / (k!(n - k)!)$ .

### 165 2.3. Materials with Statistically Oriented Fibres

166 Let  $\mathcal{F}$  be a generic physical quantity associated with a fibre-reinforced material, comprised of an  
 167 isotropic matrix and anisotropic statistically oriented fibres. The considered quantity may be either a  
 168 thermo-mechanical variable, such as stress, or a material property, such as stiffness or permeability.  
 169 The mixture of matrix and fibres is assumed to be constrained, i.e., the matrix and fibres attain the  
 170 same motion, with the same velocity  $\mathbf{v}$  and the same deformation gradient  $\mathbf{F}$ . For the sake of simplicity,  
 171 we limit ourselves to the case of a single family of statistically oriented fibres. The orientation of the  
 172 fibres is described by the probability  $\Psi(\mathbf{M})$  to find a fibre in a given referential direction  $\mathbf{M}$  in the  
 173 material unit sphere  $\mathbb{S}^2\mathcal{B} = \{\mathbf{M} : \|\mathbf{M}\| = 1\}$ . The probability density function  $\Psi$  is assumed to be  
 174 invariant under the transformation  $\mathbf{M} \mapsto -\mathbf{M}$ , and normalised to one over the sphere, i.e. [30, 31],

$$\Psi(-\mathbf{M}) = \Psi(\mathbf{M}), \quad \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) = 1. \quad (6)$$

175 Note that we shall omit writing the ‘‘area element’’ or, more properly, the *area two-form* [32, 33] ‘‘dS’’  
 176 in all surface integrals. If  $\Phi_0$  and  $\Phi_1$  are the referential volumetric fractions of the matrix and the  
 177 fibres, respectively, the physical quantity  $\mathcal{F}$  can be written, in the reference configuration, with the  
 178 rule-of-mixture expression

$$\mathcal{F} = \hat{\mathcal{F}}(\mathbf{C}, \Psi) = \Phi_0 \hat{\mathcal{F}}_0(\mathbf{C}) + \Phi_1 \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}), \quad (7)$$

179 where  $\mathbf{A} = \mathbf{M} \otimes \mathbf{M}$  is the structure tensor,  $\hat{\mathcal{F}}$  is the constitutive function of  $\mathcal{F}$ ,  $\hat{\mathcal{F}}_0$  is the isotropic  
 180 constitutive function of quantity  $\mathcal{F}_0$  in the matrix, and  $\hat{\mathcal{F}}_1$  is the anisotropic constitutive function of  
 181 quantity  $\mathcal{F}_1$  in the fibres. The integral

$$\mathcal{F}_e = \hat{\mathcal{F}}_e(\mathbf{C}, \Psi) = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}), \quad (8)$$

182 called the *fibre ensemble of  $\mathcal{F}_1$*  [18, 19], accounts for the effect of the fibres, and had initially been  
 183 introduced for the case of the elastic potential [17].

184 In general, it is not possible to factorise the deformation  $\mathbf{C}$  out of the integral, and therefore the  
 185 fibre ensemble cannot be calculated directly, but must be evaluated at each increment of deformation.  
 186 This has been done [20] by means of the method of the spherical  $t$ -designs [21, 34], i.e., a set of  $N$   
 187 points  $\{\mathbf{M}^{(1)}, \dots, \mathbf{M}^{(N)}\}$  in the material unit sphere  $\mathbb{S}^2\mathcal{B}$  such that, for polynomials  $\mathcal{P}$  of degree  $k \leq t$ ,

$$\int_{\mathbb{S}^2\mathcal{B}} \mathcal{P}(\mathbf{M}) = \frac{4\pi}{N} \sum_{r=1}^N \mathcal{P}(\mathbf{M}^{(r)}), \quad (9)$$

188 where  $4\pi$  is the (surface) measure of the unit sphere  $\mathbb{S}^2\mathcal{B}$ . As mentioned in the Introduction, we shall  
 189 denote the numerical integration of the rule-of-mixture expression of the fibre ensemble of Equation  
 190 (8), performed with the method of the spherical designs, by the acronym FESD.

191 It is crucial to remark that a single, direct integration is possible when the constitutive function  
 192  $\hat{\mathcal{F}}_1$  is a *separable* function of the structure tensor  $\mathbf{A}$  and the deformation  $\mathbf{C}$ . For a (scalar) constitutive  
 193 function  $\hat{\mathcal{F}}_1$ , the most common case of separable function is a *tensor-power polynomial* in the structure  
 194 tensor  $\mathbf{A}$  [19], of the type

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = a \left[ q_0(\mathbf{C}) + \sum_{p=1}^n \langle \mathbb{Q}_p(\mathbf{C}) | \mathbf{A}^{\otimes p} \rangle \right], \quad (10)$$

195 where  $a$  is a material constant with units of  $\mathcal{F}_1$ ,  $q_0(\mathbf{C})$  is a non-dimensional scalar function of the  
 196 deformation and the non-dimensional ‘‘covariant’’ tensor functions  $\mathbb{Q}_p$  are such that  $\mathbb{Q}_p(\mathbf{C})$  is a tensor  
 197 of order  $2p$ , which contracts with the ‘‘contravariant’’ tensor-power  $\mathbf{A}^{\otimes p}$ , which is the tensor of order  
 198  $2p$  defined by

$$\mathbf{A}^{\otimes p} = \underbrace{\mathbf{A} \otimes \dots \otimes \mathbf{A}}_{p \text{ times}}. \quad (11)$$

199 Note that the tensors  $\mathbb{Q}_p(\mathbf{C})$  and  $\mathbf{A}^{\otimes p}$  are fully “covariant” and fully “contravariant”, respectively, which  
 200 justifies the bra-ket notation,  $\langle \cdot | \cdot \rangle$ . For the constitutive function in Equation (10), it is possible to  
 201 exploit the linearity of the integration operator and to factorise the deformation out of each resulting  
 202 integral, so that the fibre ensemble of Equation (8) becomes [19]

$$\mathcal{F}_e = \hat{\mathcal{F}}_e(\mathbf{C}, \Psi) = a \left[ q_0(\mathbf{C}) + \sum_{p=1}^n \langle \mathbb{Q}_p(\mathbf{C}) | \mathbb{H}_p \rangle \right], \quad (12)$$

203 where we define the averaged structure tensor of order  $2p$  as [30, 31, 19, 35]

$$\mathbb{H}_p = \int_{\mathbb{S}^{2B}} \Psi(\mathbf{M}) \mathbf{A}^{\otimes p}. \quad (13)$$

204 Therefore, an analytical form of the fibre ensemble can be obtained as a function of the deformation  
 205  $\mathbf{C}$  and can be introduced directly into a Finite Element implementation, without the need to calculate  
 206 an integral at each increment of deformation.

207 We shall exploit the property of direct integrability of polynomial constitutive functions to pro-  
 208 pose our integration methods in Section 3.

209 **Remark.** We refer to the constitutive function of Equation (10) as to a tensor-power polynomial  
 210 because the *tensor power*  $\mathbf{A}^{\otimes p}$  is involved, rather than the regular power  $\mathbf{A}^p$ . Indeed, because of the  
 211 *idempotence* of  $\mathbf{A}$  [36, 37], the regular power would lead to the trivial result  $\mathbf{A}^p = \mathbf{A}$ , which means  
 212 that any (regular) polynomial of the  $N$ -th order in  $\mathbf{A}$  would reduce to an affine function in  $\mathbf{A}$ . The  
 213 idempotence of  $\mathbf{A}$  can be shown in components:

$$(\mathbf{A}^2)^{AD} = A^{AB} G_{BC} A^{CD} = M^A M^B G_{BC} M^C M^D = M^A M^D = A^{AD}. \quad (14)$$

214 Furthermore, we note that, for a second-order tensor, whereas the regular power is an *internal* oper-  
 215 ation, the tensor power is an *external* operation, in so far as its result is a tensor of different order  
 216 than the original one. A particularly interesting case occurs when the  $2p$ -th order tensor  $\mathbb{Q}_p(\mathbf{C})$  can be  
 217 written as the tensor product of  $p$  tensors of order two. An even more peculiar situation occurs when  
 218 it holds that  $\mathbb{Q}_p(\mathbf{C}) = q_p(\mathbf{C}) \mathbf{C}^{\otimes p}$ , for every  $p \in \{1, \dots, N\}$ , with  $q_p(\mathbf{C})$  being a suitable scalar-valued  
 219 function of  $\mathbf{C}$ , so that Equation (10) becomes

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = a \left[ q_0(\mathbf{C}) + \sum_{p=1}^n q_p(\mathbf{C}) \langle \mathbf{C}^{\otimes p} | \mathbf{A}^{\otimes p} \rangle \right]. \quad (15)$$

220 Because of the identity

$$I_4^p = (\mathbf{C} : \mathbf{A})^p = \langle \mathbf{C} | \mathbf{A} \rangle^p = \langle \mathbf{C}^{\otimes p} | \mathbf{A}^{\otimes p} \rangle, \quad (16)$$

221 the expression (15) becomes a polynomial of degree  $N$  in the fourth invariant  $I_4 = \langle \mathbf{C} | \mathbf{A} \rangle = \mathbf{C} : \mathbf{A}$ ,  
 222 i.e.,

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \check{\mathcal{F}}_1(I_4) = q_0(\mathbf{C}) + \sum_{p=1}^N q_p(\mathbf{C}) I_4^p. \quad (17)$$

#### 223 2.4. Averaged Structure Tensors $\mathbb{H}_p$ of Order $2p$

224 To the best of our knowledge, the generalised structure tensors that we denote  $\mathbb{H}_p$  in Equation (13)  
 225 were first introduced by Kanatani [30], who actually called “fabric tensors” the *deviatoric* parts of the  
 226  $\mathbb{H}_p$ , with some normalisation constants (cf., in [30], Equation (3.4), which corresponds exactly to the  
 227 definition of  $\mathbb{H}_p$ , and Equation (3.3), which defines Kanatani’s “fabric tensors”). Advani and Tucker  
 228 [31] noted that all averaged structure tensors of order smaller than  $p \geq 2$  can be found from  $\mathbb{H}_p$  by  
 229 contracting pairs of its indices (which, in our formalism, requires the use of the metric tensor), as it  
 230 can be shown, e.g., in components. Our group first employed the tensors  $\mathbb{H}_p$  only recently [19] and,  
 231 regretfully, we were unaware of the works by Kanatani [30] and Advani and Tucker [31] at that time.

232 For  $p = 1$ , the averaged tensor in Equation (13) coincides with the second-order tensor given by the  
 233 average of the structure tensor  $\mathbf{A}$ ,

$$\mathbf{H} = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathbf{A}, \quad (18)$$

234 which Gasser et al. [6] called ‘‘generalised structure tensor’’ and used as the basis of their averaging  
 235 method (see Section 2.5). In the following, we will generally use the identification  $\mathbb{H}_1 \equiv \mathbf{H}$ , except in  
 236 sums over  $p$  involving the tensors  $\mathbb{H}_p$  of order  $2p$ . To our knowledge, prior to this work, the fourth-order  
 237 averaged structure tensor  $\mathbb{H}_2$  was used in biomechanics by Vasta et al. [38] and Gizzi et al. [39], who  
 238 called it simply  $\mathbb{H}$ .

### 239 2.5. The Gasser-Ogden-Holzapfel Method (GOH)

240 The method proposed by Gasser et al. [6], thereby called GOH method, allows for a single, direct  
 241 integration, and we describe it here in our notation. Gasser et al. [6] proposed to evaluate the overall  
 242 effect of the fibres on a physical quantity  $\mathcal{F}$  by replacing the structure tensor  $\mathbf{A}$  in the fibre function  
 243  $\mathcal{F}_1 = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A})$  by means of its directional average  $\mathbf{H}$  introduced in Equation (18), to obtain

$$\mathcal{F}_{\text{GOH}} = \hat{\mathcal{F}}_{\text{GOH}}(\mathbf{C}, \Psi) = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{H}), \quad (19)$$

244 which they used in Equation (7) in place of our fibre ensemble  $\hat{\mathcal{F}}_e$  of Equation (8). When  $\hat{\mathcal{F}}_1$  is affine  
 245 in the structure tensor  $\mathbf{A}$ , i.e., it is a (tensor-power) polynomial of degree one in  $\mathbf{A}$ ,

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = a [q_0(\mathbf{C}) + \mathbf{Q}(\mathbf{C}) : \mathbf{A}], \quad (20)$$

246 where  $a$  is a constant,  $q_0$  is a scalar function of  $\mathbf{C}$ , and  $\mathbf{Q}$  is a second-order ‘‘covariant’’ tensor-valued  
 247 function of  $\mathbf{C}$ , we have

$$\begin{aligned} \hat{\mathcal{F}}_e(\mathbf{C}, \Psi) &= \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) a [q_0(\mathbf{C}) + \mathbf{Q}(\mathbf{C}) : \mathbf{A}] \\ &= a \left[ q_0(\mathbf{C}) + \mathbf{Q}(\mathbf{C}) : \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathbf{A} \right) \right] = a [q_0(\mathbf{C}) + \mathbf{Q}(\mathbf{C}) : \mathbf{H}] \\ &= \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{H}) = \hat{\mathcal{F}}_{\text{GOH}}(\mathbf{C}, \Psi), \end{aligned} \quad (21)$$

248 i.e., the rule-of-mixture method coincides with the GOH method [17].

## 249 3. Approximation of the Fibre Ensemble

250 Here we introduce three methods that provide analytical approximations of the fibre ensemble (8),  
 251 all based on the fact that, for a fibre function  $\hat{\mathcal{F}}_1$  that is a tensor-power polynomial in the structure  
 252 tensor, a single, direct integration is possible [19]. Two of the proposed methods are based on the  
 253 Taylor expansion of the fibre function  $\hat{\mathcal{F}}_1$ , in order to obtain polynomial functions in the structure  
 254 tensor  $\mathbf{A}$ . In the first method, we expand in the transversely isotropic invariants, which are linear  
 255 functions of  $\mathbf{A}$  (see Equation (72)). In the second method, we expand in the structure tensor  $\mathbf{A}$ . In  
 256 the third method, in a fashion similar to that of the GOH method [6], for the case of a fibre function  
 257  $\hat{\mathcal{F}}_1$  that is a function of a tensor-power polynomial  $\mathcal{P}(\mathbf{A})$ , we replace the fibre ensemble by the same  
 258 function evaluated at the directional average of  $\mathcal{P}(\mathbf{A})$ , which can be calculated directly.

### 259 3.1. Taylor Expansion in the Invariants (INEX)

260 Let  $\check{\mathcal{F}}_1(I_1, I_2, I_3, I_4, I_5) = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A})$  be the fibre constitutive function written as a function of the five  
 261 transversely isotropic invariants (when looking at a single direction  $\mathbf{M}$ , the symmetry is naturally  
 262 that of transverse isotropy). For the sake of a lighter notation, let us omit writing the three isotropic  
 263 invariants  $I_1, I_2, I_3$  among the arguments of  $\check{\mathcal{F}}_1$  and, for the sake of a simpler presentation, let us  
 264 assume that  $\check{\mathcal{F}}_1$  does not depend on the fifth invariant  $I_5$ . If  $I_5$  were included, the derivation would  
 265 be analogous, but lengthier, and the Taylor expansion formulae would require the introduction of the  
 266 multi-index notation. Furthermore, we write  $I_4 = J^{2/3} \bar{I}_4$ , i.e., we express  $I_4$  in terms of its purely

267 distortional counterpart  $\bar{I}_4$ . Therefore, let us write the fibre quantity  $\mathcal{F}_1$  as a constitutive function of  
 268  $J$  and  $\bar{I}_4$ , i.e.,

$$\mathcal{F}_1 = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \check{\mathcal{F}}_1(J, \bar{I}_4). \quad (22)$$

269 We remark that, although we omitted indicating explicitly the dependence of  $\check{\mathcal{F}}_1$  on  $I_3 = J^2$ , in the  
 270 sequel we express  $\check{\mathcal{F}}_1$  as a function of  $\bar{I}_4$  and  $J$  in order to emphasise that  $J$  is used to express  $I_4$  as  
 271  $J^{2/3}\bar{I}_4$ . We also remark that we are *not* decomposing  $I_4$  into its volumetric and distortional parts in  
 272 order to impose incompressibility, which is the most common case in which one uses the volumetric-  
 273 distortional decomposition, but because it serves our purpose of a Taylor expansion at a point of zero  
 274 (distortional) deformation, as it will be explained later.

275 For a given  $\mathbf{C} = J^{2/3}\bar{\mathbf{C}}$ , it is fairly straightforward to prove that the admissible values of  $I_4$   
 276 and  $\bar{I}_4$  belong to the closed intervals  $\Lambda(\mathbf{C}) = [\lambda_{\min}^2, \lambda_{\max}^2]$  and  $\Lambda(\bar{\mathbf{C}}) = [\bar{\lambda}_{\min}^2, \bar{\lambda}_{\max}^2]$ , respectively,  
 277 where  $\lambda_{\min}^2$  and  $\lambda_{\max}^2$  are the minimum and maximum eigenvalue of  $\mathbf{C}$ , and  $\bar{\lambda}_{\min}^2$  and  $\bar{\lambda}_{\max}^2$  are those  
 278 of  $\bar{\mathbf{C}}$  (see Appendix D for both an analytical and graphical proof). Note that, in the undeformed  
 279 configuration, for which  $\mathbf{C} = \bar{\mathbf{C}} = \mathbf{G}$  (where the metric tensor  $\mathbf{G}$  serves as the ‘‘covariant’’ identity  
 280 tensor), the intervals  $\Lambda(\mathbf{C})$  and  $\Lambda(\bar{\mathbf{C}})$  degenerate into the singleton  $\Lambda(\mathbf{G}) = \{1\}$ . We also remark  
 281 that the admissible intervals of  $I_5$  and  $\bar{I}_5$  have the same form of those of  $I_4$  and  $\bar{I}_4$ , except that the  
 282 exponents 2 of the maximum and minimum stretches have to be replaced by exponents 4.

283 For our purposes, it is very important to note that it is *always* verified that  $\bar{I}_{40} = 1 \in \Lambda(\bar{\mathbf{C}}) =$   
 284  $[\bar{\lambda}_{\min}^2, \bar{\lambda}_{\max}^2]$ . Indeed, the condition  $\det \bar{\mathbf{C}} = 1$  implies that  $\bar{\lambda}_{\min}^2 < 1$  and  $\bar{\lambda}_{\max}^2 > 1$ . Therefore, if  $\check{\mathcal{F}}_1$   
 285 belongs to the space  $C^n(\mathring{\Lambda}(\bar{\mathbf{C}}))$  of continuously differentiable functions up to order  $n$  in the open set  
 286  $\mathring{\Lambda}(\bar{\mathbf{C}}) = ]\bar{\lambda}_{\min}^2, \bar{\lambda}_{\max}^2[$  of the interior points of  $\Lambda(\bar{\mathbf{C}})$ , it is possible to approximate  $\check{\mathcal{F}}_1$  by means of a  
 287 Taylor expansion in the variable  $\bar{I}_4$ , about the value  $\bar{I}_{40} = 1 \in \Lambda(\bar{\mathbf{C}})$ . For this purpose, we invoke the  
 288 Taylor’s expansion formula of order  $n$  for  $\check{\mathcal{F}}_1$ , which reads

$$\check{\mathcal{F}}_1(J, \bar{I}_4) = \check{\mathcal{T}}_n(J, \bar{I}_4) + \check{\mathcal{R}}_n(J, \bar{I}_4) = \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{\mathcal{F}}_1}{\partial \bar{I}_4^{(j)}}(J, 1) [\bar{I}_4 - 1]^j + \check{\mathcal{R}}_n(J, \bar{I}_4), \quad (23)$$

289 where  $\check{\mathcal{T}}_n(J, \bar{I}_4)$  is the Taylor polynomial of order  $n$  associated with  $\check{\mathcal{F}}_1$  at  $1 \in \mathring{\Lambda}(\bar{\mathbf{C}})$ . If  $\check{\mathcal{F}}_1$  is differen-  
 290 tiable  $n + 1$  times in  $\mathring{\Lambda}(\bar{\mathbf{C}}) \setminus \{1\}$ , the remainder  $\check{\mathcal{R}}_n(\bar{I}_4)$  can be given in Lagrange’s form as

$$\check{\mathcal{R}}_n(J, \bar{I}_4) = \frac{1}{(n+1)!} \frac{\partial^{(n+1)} \check{\mathcal{F}}_1}{\partial \bar{I}_4^{(n+1)}}(J, \xi_{n+1}) [\bar{I}_4 - 1]^{n+1}, \quad (24)$$

291 for some  $\xi_{n+1} \in \mathring{\Lambda}(\bar{\mathbf{C}})$  lying between 1 and  $\bar{I}_4$ , and depending on  $\bar{I}_4$  as well as on the order  $n$  of the  
 292 expansion.

293 We now exploit  $\bar{I}_4 = \bar{\mathbf{C}} : \mathbf{A} = J^{-2/3} \mathbf{C} : \mathbf{A}$ , and write the Taylor polynomial and the remainder  
 294 as explicit functions of the structure tensor  $\mathbf{A}$ , i.e.,

$$\check{\mathcal{T}}_n(J, \bar{I}_4) = \hat{\mathcal{T}}_n(\mathbf{C}, \mathbf{A}) = \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{\mathcal{F}}_1}{\partial \bar{I}_4^{(j)}}(J, 1) [J^{-2/3} \mathbf{C} : \mathbf{A} - 1]^j, \quad (25a)$$

$$\check{\mathcal{R}}_n(J, \bar{I}_4) = \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}) = \frac{1}{(n+1)!} \frac{\partial^{(n+1)} \check{\mathcal{F}}_1}{\partial \bar{I}_4^{(n+1)}}(J, \xi_{n+1}) [J^{-2/3} \mathbf{C} : \mathbf{A} - 1]^{n+1}, \quad (25b)$$

295 which give

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \hat{\mathcal{T}}_n(\mathbf{C}, \mathbf{A}) + \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}). \quad (26)$$

296 Then, we multiply both sides of Equation (26) by the probability distribution  $\Psi(\mathbf{M})$ , integrate over  
 297 the material sphere  $\mathbb{S}^2\mathcal{B}$ , and obtain

$$\mathcal{F}_e = \mathcal{G}_n + \mathcal{E}_n, \quad (27)$$

298 where  $\mathcal{F}_e$  is the fibre ensemble of Equation (8), and

$$\mathcal{G}_n = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{T}}_n(\mathbf{C}, \mathbf{A}), \quad (28a)$$

$$\mathcal{E}_n = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}), \quad (28b)$$

299 are the  $n$ -th order approximation of  $\mathcal{F}_e$  and the corresponding error  $\mathcal{E}_n$ , which is entirely defined by  
300 the difference  $\mathcal{E}_n := \mathcal{F}_e - \mathcal{G}_n$ . Equation (28a) defines the sequence  $\{\mathcal{G}_n\}_{n \in \mathbb{N}}$ , in which  $\mathcal{G}_n$  is given by

$$\begin{aligned} \mathcal{G}_n = \hat{\mathcal{G}}_n(\mathbf{C}, \Psi) &= \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{\mathcal{F}}_1}{\partial \bar{\mathbf{I}}_4^{(j)}}(J, 1) \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) [J^{-2/3} \mathbf{C} : \mathbf{A} - 1]^j \\ &= \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{\mathcal{F}}_1}{\partial \bar{\mathbf{I}}_4^{(j)}}(J, 1) \sum_{k=0}^j \binom{j}{k} (-1)^k (J^{-2/3})^{j-k} \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) [\mathbf{C} : \mathbf{A}]^{j-k}. \end{aligned} \quad (29)$$

301 The Cauchy-Green deformation tensor  $\mathbf{C}$  can be factorised out of the integral sign in (29) by using  
302 the identity (16), from which

$$\int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) [\mathbf{C} : \mathbf{A}]^{j-k} = \langle \mathbf{C}^{\otimes(j-k)} | \mathbb{H}_{j-k} \rangle. \quad (30)$$

303 By virtue of this result, the  $n$ -th order approximation of  $\mathcal{F}_e$  can be recast in the compact form

$$\mathcal{G}_n = \hat{\mathcal{G}}_n(\mathbf{C}, \Psi) = \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{\mathcal{F}}_1}{\partial \bar{\mathbf{I}}_4^{(j)}}(J, 1) \sum_{k=0}^j \binom{j}{k} (-1)^k (J^{-2/3})^{j-k} \langle \mathbf{C}^{\otimes(j-k)} | \mathbb{H}_{j-k} \rangle, \quad (31)$$

304 in which the deformation has been completely factorised with respect to directional averaging, the  
305 latter being accounted for by the averaged structure tensor of order  $2(j-k)$ ,  $\mathbb{H}_{j-k}$ . To estimate  
306 the error, let us consider for simplicity the case in which  $\mathcal{F}_1$  is a scalar constitutive function, so  
307 that its associated fibre ensemble,  $\mathcal{F}_e$ , and  $n$ -th order approximation,  $\mathcal{G}_n$ , are scalars too. If the error  
308  $\mathcal{E}_n = \mathcal{F}_e - \mathcal{G}_n$  vanishes as  $n$  goes towards infinity,  $\mathcal{F}_e$  can be represented exactly by the limit  $\lim_{n \rightarrow \infty} \mathcal{G}_n$ ,  
309 in which case it holds that

$$\mathcal{F}_e = \lim_{n \rightarrow \infty} \mathcal{G}_n. \quad (32)$$

310 To estimate  $\mathcal{E}_n$ , we follow the theory of Taylor expansion formulae, and we infer that, if there exist  
311 positive constants  $L$  and  $Q$ , such that

$$\left| \frac{\partial^{n+1} \check{\mathcal{F}}_1}{\partial \bar{\mathbf{I}}_4^{n+1}}(J, \bar{\mathbf{I}}_4) \right| \leq L Q^{n+1}, \quad \forall \bar{\mathbf{I}}_4 \in \mathring{\Delta}(\bar{\mathbf{C}}), \quad (33)$$

312 then it holds that

$$|\mathcal{E}_n| \leq \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \left| \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}) \right| \leq L \frac{Q^{n+1}}{(n+1)!} \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) |\bar{\mathbf{I}}_4 - 1|^{n+1} \leq L \frac{Q^{n+1} (\bar{\lambda}_{\max}^2 - \bar{\lambda}_{\min}^2)^{n+1}}{(n+1)!}. \quad (34)$$

313 Note that, in the case in which  $\mathcal{F}_1$  is a tensor-valued constitutive quantity, the estimates (33) and (34)  
314 must be generalised by replacing the absolute value with an appropriate norm.

315 For a sufficiently high order  $n$  of the Taylor's expansion (23), we enforce the approximation

$$\mathcal{F}_e \simeq \mathcal{G}_n, \quad (35)$$

316 the accuracy of which increases when the absolute value of the error,  $|\mathcal{E}_n|$ , tends towards zero. For  
317 example, this is the case when  $\bar{\lambda}_{\max}^2$  and  $\bar{\lambda}_{\min}^2$  tend to be equal to each other.

318 Equations (27) and (35) constitute the INEX (Invariant Expansion) method, and provide a  
319 polynomial approximation of the fibre ensemble  $\hat{\mathcal{F}}_e$ , *regardless* of the form of the orientation probability  
320 distribution  $\Psi$ . This is achieved by expanding the fibre constitutive function  $\check{\mathcal{F}}_1(J, \cdot)$  about  $\bar{\mathbf{I}}_4 = 1$ ,  
321 which rules out any dependence on a "privileged" direction  $\mathbf{M}_0$ . In order to clearly show this, let  
322  $\mathbf{M}_0$  be any direction, with the associated structure tensor  $\mathbf{A}_0 = \mathbf{M}_0 \otimes \mathbf{M}_0$ . Since  $\bar{\mathbf{I}}_4 = \bar{\mathbf{C}} : \mathbf{A}_0$ ,

323 when  $\bar{\mathbf{C}} = \mathbf{G}$ , we have that  $\bar{I}_{40} = 1$ , for every  $\mathbf{M}_0$ . Therefore, the INEX approximation is valid for  
 324 any orientation distribution  $\Psi$ . This is in contrast with the STEEX method presented in Section 3.2,  
 325 which is based on the expansion about  $\mathbf{A}_0 = \mathbf{M}_0 \otimes \mathbf{M}_0$ , and is thus accurate *only* for orientation  
 326 distributions  $\Psi$  with small dispersions about  $\mathbf{M}_0$ . We observed that the INEX method gave the best  
 327 results for even orders of expansion.

### 328 3.2. Taylor Expansion in the Structure Tensor (STEEX)

329 Given a fibre function  $\mathcal{F}_1 = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A})$  and a structure tensor  $\mathbf{A}_0 = \mathbf{M}_0 \otimes \mathbf{M}_0$ , if  $\hat{\mathcal{F}}_1$  is of class  $C^n$  in  
 330 a neighbourhood of  $\mathbf{A}_0$ , it is possible to use Taylor's expansion formula in  $\mathbf{A}$  about  $\mathbf{A}_0$ ,

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \hat{\mathcal{T}}_n(\mathbf{C}, \mathbf{A}) + \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}) = \sum_{j=0}^n \frac{1}{j!} \left\langle \frac{\partial^{(j)} \hat{\mathcal{F}}_1}{\partial \mathbf{A}^{(j)}}(\mathbf{C}, \mathbf{A}_0) \middle| (\mathbf{A} - \mathbf{A}_0)^{\otimes j} \right\rangle + \hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A}), \quad (36)$$

331 where, similarly to the case of the INEX method,  $\hat{\mathcal{T}}_n(\mathbf{C}, \mathbf{A})$  is the Taylor polynomial of order  $n$ , and  
 332 if  $\hat{\mathcal{F}}_1$  is of class  $C^{n+1}$  in  $\mathbf{A}$ , the remainder  $\hat{\mathcal{R}}_n(\mathbf{C}, \mathbf{A})$  can be expressed in Lagrange's form (we omit  
 333 the details).

334 Multiplying both sides of Equation (36) by the probability density  $\Psi$  and then integrating over  
 335 the material sphere  $\mathbb{S}^2\mathcal{B}$ , we obtain

$$\mathcal{F}_e = \mathcal{G}_n + \mathcal{E}_n, \quad (37)$$

336 where, analogously to the case of the INEX method,  $\mathcal{F}_e$  is the fibre ensemble of Equation (8),  $\mathcal{G}_n$  is  
 337 the  $n$ -th order approximation of  $\mathcal{F}_e$  and  $\mathcal{E}_n$  is the  $n$ -th order error, defined formally as in Equations  
 338 (28a) and (28b). The term  $\mathcal{G}_n$  of the sequence  $\{\mathcal{G}_n\}_{n \in \mathbb{N}}$  is given by

$$\begin{aligned} \mathcal{G}_n = \hat{\mathcal{G}}_n(\mathbf{C}, \Psi) &= \int_{\mathbb{S}^2\mathcal{B}} \left[ \Psi(\mathbf{M}) \sum_{j=0}^n \frac{1}{j!} \left\langle \frac{\partial^{(j)} \hat{\mathcal{F}}_1}{\partial \mathbf{A}^{(j)}}(\mathbf{C}, \mathbf{A}_0) \middle| (\mathbf{A} - \mathbf{A}_0)^{\otimes j} \right\rangle \right] \\ &= \sum_{j=0}^n \frac{1}{j!} \left\langle \frac{\partial^{(j)} \hat{\mathcal{F}}_1}{\partial \mathbf{A}^{(j)}}(\mathbf{C}, \mathbf{A}_0) \middle| \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) (\mathbf{A} - \mathbf{A}_0)^{\otimes j} \right\rangle. \end{aligned} \quad (38)$$

339 Note that the integrals on the right-hand side of the bra-ket are independent of the deformation  $\mathbf{C}$ .  
 340 Using the expression (5) of the binomial tensor power and the linearity of the integral operation, it is  
 341 possible to write Equation (38) in the form

$$\mathcal{F}_e \simeq \mathcal{G}_n = \hat{\mathcal{G}}_n(\mathbf{C}, \Psi) = \sum_{j=0}^n \frac{1}{j!} \left\langle \frac{\partial^{(j)} \hat{\mathcal{F}}_1}{\partial \mathbf{A}^{(j)}}(\mathbf{C}, \mathbf{A}_0) \middle| \sum_{k=0}^j [(-1)^k \binom{j}{k} \text{msym}(\mathbb{H}_{j-k} \otimes \mathbf{A}_0^{\otimes k})] \right\rangle, \quad (39)$$

342 which features the averaged structure tensors  $\mathbb{H}_p$  of Equation (13). Equations (37) and (39) yield  
 343 an analytical approximation of the fibre potential  $\mathcal{F}_e$  as a function of the deformation  $\mathbf{C}$ . It seems  
 344 natural to expand about the structure tensor  $\mathbf{A}_0 = \mathbf{M}_0 \otimes \mathbf{M}_0$  relative to the dominant direction  $\mathbf{M}_0$   
 345 of the fibres, in which case the best results are obtained when the dispersion of the fibres about that  
 346 direction is relatively small. We note that Vasta et al. [38] have in fact implemented what here we  
 347 would call the STEEX method of order 2, i.e., involving only  $\mathbb{H}_1 \equiv \mathbf{H}$  and  $\mathbb{H}_2$ . Also in this case, the  
 348 best results were obtained for even orders of expansion.

### 349 3.3. Polynomial Argument Method (PARG)

350 In this section, we consider a fibre function of the type

$$\mathcal{F}_1 = \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A})), \quad (40)$$

351 where  $\mathfrak{f}$  describes the physical quantity that has to be modelled (e.g., the elastic potential) and  $\mathcal{P}(\mathbf{C}, \mathbf{A})$   
 352 is defined by the  $N$ -th degree *tensor-power polynomial*

$$\mathcal{P}(\mathbf{C}, \mathbf{A}) := q_0(\mathbf{C}) + \sum_{p=1}^N \langle \mathbb{Q}_p(\mathbf{C}) | \mathbf{A}^{\otimes p} \rangle, \quad (41)$$

353 in which, as in Equation (10),  $q_0$  and  $\mathbb{Q}_p$  are, respectively, a non-dimensional scalar-valued func-  
 354 tion and a non-dimensional ‘‘covariant’’ tensor-valued function of order  $2p$  of the right Cauchy-Green  
 355 deformation tensor. We propose to approximate the fibre ensemble as

$$\mathcal{F}_e = \hat{\mathcal{F}}_e(\mathbf{C}, \Psi) = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A})) \simeq \mathfrak{f} \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right). \quad (42)$$

356 This approximation becomes exact if the operation of directional averaging commutes with the function  
 357  $\mathfrak{f}$ . This holds true, for example, when  $\mathfrak{f}$  is a polynomial of degree  $M$  in  $\mathcal{P}(\mathbf{C}, \mathbf{A})$ , and  $\mathcal{P}(\mathbf{C}, \mathbf{A})$  is  
 358 expressed as the  $\hat{\mathcal{F}}_1$  of Equation (15), with  $\mathbb{Q}_p(\mathbf{C}) = q_p(\mathbf{C}) \mathbf{C}^{\otimes p}$ , and  $q_p(\mathbf{C})$  scalar functions of  $\mathbf{C}$ , so  
 359 that  $\mathcal{P}(\mathbf{C}, \mathbf{A})$  can be written as a polynomial in  $I_4$ :

$$\mathcal{P}(\mathbf{C}, \mathbf{A}) = \check{\mathcal{P}}(I_4) = q_0(\mathbf{C}) + \sum_{p=1}^N q_p(\mathbf{C}) I_4^p. \quad (43)$$

360 With these assumptions,  $\mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A}))$  can be reformulated as a polynomial of degree  $MN$  in  $I_4$ , i.e.,

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A})) = \mathfrak{f}(\check{\mathcal{P}}(I_4)) = a_0(\mathbf{C}) + \sum_{h=1}^{MN} a_h(\mathbf{C}) I_4^h = a_0(\mathbf{C}) + \sum_{h=1}^{MN} a_h(\mathbf{C}) \langle \mathbf{C} | \mathbf{A} \rangle^h, \quad (44)$$

361 where each function  $a_h$ , with  $h \in \{0, \dots, MN\}$ , is obtained by combining the functions  $q_p$  of (17)  
 362 with the coefficients of the polynomial expressing  $\mathfrak{f}$ . By using the identity (16), which leads to

$$\int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \langle \mathbf{C} | \mathbf{A} \rangle^h = \left\langle \mathbf{C}^{\otimes h} \left| \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathbf{A}^{\otimes h} \right. \right\rangle = \langle \mathbf{C}^{\otimes h} | \mathbb{H}_h \rangle, \quad (45)$$

363 the fibre ensemble can be expressed exactly in terms of the averaged generalised structure tensors  $\mathbb{H}_h$ ,  
 364 i.e.,

$$\mathcal{F}_e = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = a_0(\mathbf{C}) + \sum_{h=1}^{MN} a_h(\mathbf{C}) \langle \mathbf{C}^{\otimes h} | \mathbb{H}_h \rangle. \quad (46)$$

365 In general, however, for arbitrary functions  $\mathfrak{f}$ , the approximation (42) is exact in the limit  $\mathbf{C} \rightarrow \mathbf{G}$ .  
 366 Indeed, at  $\mathbf{C} = \mathbf{G}$ , one obtains  $I_4 = I_{40} = \langle \mathbf{G} | \mathbf{A} \rangle = 1$  and the polynomial

$$\mathcal{P}(\mathbf{G}, \mathbf{A}) = \check{\mathcal{P}}(1) = q_0(\mathbf{G}) + \sum_{p=1}^N q_p(\mathbf{G}) \quad (47)$$

367 becomes constant with respect to the structure tensor, thereby rendering the approximation (42) an  
 368 identity. Nevertheless, the reliability of (42) deteriorates when  $\mathbf{C}$  deviates from  $\mathbf{G}$ .

369 To highlight the loss of accuracy of (42) when  $\mathbf{C} \neq \mathbf{G}$ , let us consider a physically relevant example.  
 370 We set  $N = 2$ ,  $q_0(\mathbf{C}) = 1$ ,  $q_1(\mathbf{C}) = -2$ , and  $q_2(\mathbf{C}) = 1$ , so that  $\mathcal{P}(\mathbf{C}, \mathbf{A})$  takes the form

$$\mathcal{P}(\mathbf{C}, \mathbf{A}) = 1 - 2\langle \mathbf{C} | \mathbf{A} \rangle + \langle \mathbf{C}^{\otimes 2} | \mathbf{A}^{\otimes 2} \rangle = (\langle \mathbf{C} | \mathbf{A} \rangle - 1)^2, \quad (48)$$

371 and we assume that the fibre function  $\mathcal{F}_1$  represents the anisotropic elastic potential of the Holzapfel-  
 372 Gasser-Ogden [5] type

$$\hat{\mathcal{F}}_1(\mathbf{C}, \mathbf{A}) = \hat{W}_{1a}(\mathbf{C}, \mathbf{A}) = \frac{1}{2} c_{1a} [\exp(\langle \mathbf{C} | \mathbf{A} \rangle - 1)^2 - 1]. \quad (49)$$

373 In this case, after introducing the auxiliary variable  $\eta = (\langle \mathbf{C} | \mathbf{A} \rangle - 1)^2$ , the function  $\mathfrak{f}$  is identified with

$$\mathfrak{f}(\eta) = \frac{1}{2} c_{1a} [\exp(\eta) - 1]. \quad (50)$$

374 Since  $\mathfrak{f}$  can be expanded in Taylor series about  $\eta = 0$ , we obtain

$$\mathfrak{f}(\eta) = \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{\eta^n}{n!}. \quad (51)$$

375 Thus, substituting (51) into (42) yields (for brevity, we omit the dependence of functions on their own  
376 arguments)

$$\mathcal{F}_e = \int_{\mathbb{S}^2\mathcal{B}} \Psi \mathfrak{f} = \frac{1}{2} c_{1a} \int_{\mathbb{S}^2\mathcal{B}} \Psi \sum_{n=1}^{+\infty} \frac{\eta^n}{n!} = \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta^n. \quad (52)$$

377 At each order  $n \geq 1$ , we write the integral  $\int_{\mathbb{S}^2\mathcal{B}} \Psi \eta^n$  as

$$\int_{\mathbb{S}^2\mathcal{B}} \Psi \eta^n = \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta \right)^n + \mathfrak{R}_n, \quad (53)$$

378 where we refer to  $\mathfrak{R}_n$  as to the  $n$ -th order residuum of the approximation. Consequently, (52) can be  
379 rewritten as

$$\mathcal{F}_e = \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta \right)^n + \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \mathfrak{R}_n. \quad (54)$$

380 Since the first term on right-hand-side of (54) is the exponential of the mean value of  $\eta$ , we obtain

$$\begin{aligned} \mathcal{F}_e &= \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta \right)^n + \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \mathfrak{R}_n \\ &= \frac{1}{2} c_{1a} \left[ \exp \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta \right) - 1 \right] + \frac{1}{2} c_{1a} \sum_{n=1}^{+\infty} \frac{1}{n!} \mathfrak{R}_n \end{aligned} \quad (55)$$

381 We remark that the residuum  $\mathfrak{R}_n$  can be computed exactly at any order. Indeed, it holds true that

$$\begin{aligned} \mathfrak{R}_n &= \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta^n - \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi \eta \right)^n \\ &= \sum_{j=0}^{2n} \binom{2n}{j} (-1)^j \langle \mathbf{C}^{\otimes (2n-j)} | \mathbb{H}_{2n-j} \rangle - \left( \langle \mathbf{C}^{\otimes 2} | \mathbb{H}_2 \rangle - 2 \langle \mathbf{C} | \mathbf{H} \rangle + 1 \right)^n. \end{aligned} \quad (56)$$

382 It can be shown, however, that even in the case of an equi-biaxial test (performed on an incompressible  
383 material characterised by diagonal matrix representation of  $\mathbf{C}$ ,  $[\mathbf{C}] = \text{diag}\{\lambda^2, \lambda^2, \lambda^{-4}\}$ ), the residuals  
384 may not tend to zero sufficiently fast, even for values of  $\lambda$  sufficiently close to unity. This behaviour  
385 contributes to corrupt the reliability of the PARG method and to deteriorate its agreement with the  
386 FESD method.

387 We note that, as it happens for the whole  $\hat{\mathcal{F}}_1$  in the general case of Equation (8), if  $\mathcal{P}$  in Equation  
388 (42) is an affine function, i.e., a polynomial of degree one, the PARG method reduces to the GOH  
389 method proposed in [6]. The main difference between the PARG method and the GOH method is the  
390 level at which the fibre ensemble is approximated. While in the GOH method the averaging integral  
391 is performed on the innermost argument, the structure tensor  $\mathbf{A}$ , in the PARG method of Equation  
392 (42), we take the average of the outermost argument,  $\mathcal{P}(\mathbf{C}, \mathbf{A})$ , that can be written as a tensor-power  
393 polynomial in  $\mathbf{A}$ . We remark that, while the GOH method is applicable to any constitutive function,  
394 the PARG method is only applicable when the constitutive function is expressible as a function of a  
395 tensor-power polynomial in  $\mathbf{A}$ .

## 396 4. Application to Elasticity

397 As an example of application of the integration methods presented in Section 3, we look at the averaged  
398 physical quantities that are most often sought for in the mechanics of fibre-reinforced materials and  
399 biomechanics of soft tissue: elastic potential and stress. Therefore, our physical quantity  $\mathcal{F}$  takes the  
400 meaning of elastic potential  $W$  in Equation (7), and we write

$$W = \hat{W}(\mathbf{C}, \Psi) = \Phi_0 \hat{W}_0(\mathbf{C}) + \Phi_1 \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_1(\mathbf{C}, \mathbf{A}). \quad (57)$$

401 The averaging integral of the fibre potential  $W_1$  is called the *fibre ensemble potential* [17]:

$$W_e = \hat{W}_e(\mathbf{C}, \Psi) = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_1(\mathbf{C}, \mathbf{A}). \quad (58)$$

402 In general, it is possible to attribute some “bulk” isotropic stiffness to the fibres, e.g., by using a fibre  
 403 potential  $\hat{W}_1$  given by the sum of an isotropic term and a term depending solely on the anisotropic  
 404 invariants  $I_4$  and  $I_5$  [18]. The fibre potential  $\hat{W}_1$  could therefore be written, as a function  $\check{W}_1$  of the  
 405 invariants, as

$$\check{W}_1(I_1, I_2, I_3, I_4, I_5) = \check{W}_{1i}(I_1, I_2, I_3) + \check{W}_{1a}(I_4, I_5). \quad (59)$$

406 Furthermore, in those cases in which the contribution of a fibre in direction  $\mathbf{M}$  is to be ruled out if  
 407 the direction undergoes contraction, i.e., if  $I_4 = \mathbf{C} : \mathbf{A} < 1$ , it is possible to use the Heaviside step  
 408 function  $\mathcal{H}$  evaluated at  $I_4 - 1$ , and write

$$\check{W}_{1a}(I_4, I_5) = \mathcal{H}(I_4 - 1) \check{W}_{1b}(I_4, I_5), \quad (60)$$

409 where  $\check{W}_{1b}$  describes the anisotropic behaviour in extension and is called “base” potential. We remark  
 410 that, in order to be able to employ the integration methods presented in Section 3, we must renounce  
 411 discriminating between fibres in extension (which are unaffected by the Heaviside step function) and  
 412 fibres in contraction (which are “killed” by the Heaviside step to reflect the fact that they do not  
 413 bear load). Indeed, if we were to use the Heaviside step in the fibre potential as in Equation (59), all  
 414 approximating potentials presented in Section 3 would have to be multiplied by the Heaviside step  
 415 as well. The Heaviside step with argument  $I_4 - 1 = \mathbf{C} : \mathbf{A} - 1$  would rule out the possibility of a  
 416 single, direct integration. There are two reasons for this: *a*) it would be in general impossible to know  
 417 which fibres undergo contraction *a priori*, and one would have to evaluate this at each increment  
 418 of deformation; *b*) the hypotheses of continuity and differentiability necessary for expandability of  
 419 functions in Taylor series would be, in general, violated. Therefore, an integration at each increment  
 420 of deformation would remain the only available solution method, thus defeating the purpose of the  
 421 proposed approximation methods.

422 This means that, in terms of range of applicability to the evaluation of the overall elastic behav-  
 423 iour, the methods presented in Section 3 are limited to those cases in which all fibres, or at least  
 424 most of the fibres, are in extension. This can be safely said for tissues with fibres lying mostly on a  
 425 plane and subjected to tensile plane stress. A typical example is that of blood vessels, which work as  
 426 inflated-extended tubes under physiological conditions. Schematically, blood vessels can be represented  
 427 as having, at every point, two dominant fibre directions (with some dispersion) mostly contained in  
 428 the tangent plane at that point (see, e.g., Figure 1 in [5]).

429 **Remark.** We are aware of the existence of mathematical models in which the collagen fibres contribute  
 430 to the tissue’s overall compressive stiffness. It has been recently reported [40] that this is the case, for  
 431 example, in aged or diseased intervertebral discs, and it was assumed that the fibres’ contribution to  
 432 compressive loads increases with increasing strain magnitude and is influenced by the orientation of  
 433 the fibres. Still, to the best of our knowledge and understanding, in articular cartilage (the tissue which  
 434 motivated our current study) no correlation of compressive stiffness with collagen content has been  
 435 observed [41]. For this reason, we decided to exclude all fibres that are not stretched. Even though  
 436 this modelling assumption may turn out to be far from reality in some circumstances, we do not make  
 437 it with the purpose of simplifying the calculations. On the contrary, the necessary introduction of the  
 438 Heaviside step in the evaluation of the fibre ensemble makes it highly non-linear in a non-differentiable  
 439 way, thereby excluding *a priori* the possibility of applying the methods proposed in this work.

440 For our illustrative purposes, let us choose simple forms of the matrix potential  $\hat{W}_0$ , isotropic  
 441 fibre potential  $\hat{W}_{1i}$  and (base) anisotropic fibre potential  $\hat{W}_{1b}$  (such that  $\hat{W}_{1a} = \mathcal{H}(I_4 - 1) \hat{W}_{1b}$ ),

$$\hat{W}_0(\mathbf{C}) = \frac{1}{2} c_0 (I_1(\mathbf{C}) - 3), \quad (61a)$$

$$\hat{W}_{1i}(\mathbf{C}) = \frac{1}{2} c_{1i} (I_1(\mathbf{C}) - 3), \quad (61b)$$

$$\hat{W}_{1b}(\mathbf{C}, \mathbf{A}) = \frac{1}{2} c_{1a} [\exp((\mathbf{C} : \mathbf{A} - 1)^2) - 1], \quad (61c)$$

442 in which  $c_0$ ,  $c_{1i}$  and  $c_{1a}$  are material parameters, and we assume referential volumetric fractions  
 443  $\Phi_0 = \Phi_1 = 0.5$ . The exponential form of the base anisotropic potential in Equation (61c) has been  
 444 chosen because it predicts well the characteristic stress response of soft tissues with collagen fibres  
 445 being undulated in the undeformed configuration, with a toe region and a region of increased stiffness  
 446 [42]. Moreover, since it consists of the exponential of a polynomial in  $I_4 = \mathbf{C} : \mathbf{A}$ , it also allows the  
 447 use of the PARG method proposed in Section 3.3. Note that, although the invariant  $I_5$  should also  
 448 be included in order to obtain a complete transversely isotropic representation (and avoid unphysical  
 449 results, see, e.g., [43]), very often  $I_5$  is left out, in order to limit the number of material parameters,  
 450 and therefore of experimental tests, needed to characterise the material. In passing, we note that the  
 451 form chosen for  $\hat{W}_1$  makes it a particular case of exponential Fung potential [44, 45, 46, 47], which is  
 452 the exponential of a quadratic form in the Green-Lagrange strain. Indeed, by using the definition of  
 453 Green-Lagrange strain  $\mathbf{E} = \frac{1}{2}(\mathbf{C} - \mathbf{G})$ , we can write the argument of the exponential in (61c) as a  
 454 quadratic form in  $\mathbf{E}$ :

$$(\mathbf{C} : \mathbf{A} - 1)^2 = (\mathbf{C} : \mathbf{A} - \mathbf{G} : \mathbf{A})^2 = (2\mathbf{E} : \mathbf{A})^2 = 4 [\mathbf{E} : (\mathbf{A} \otimes \mathbf{A}) : \mathbf{E}]. \quad (62)$$

455 In a Cartesian (material) reference frame with axes  $\mathbf{E}_1$ ,  $\mathbf{E}_2$ ,  $\mathbf{E}_3$ , we consider a sample of incom-  
 456 pressible soft tissue, which undergoes a biaxial tension test in directions  $\mathbf{E}_1$  and  $\mathbf{E}_2$ , with a prescribed  
 457 ratio of the nominal strain in direction 2 to the nominal strain in direction 1, i.e.,

$$\zeta = \frac{\lambda_2 - 1}{\lambda_1 - 1}. \quad (63)$$

458 In an isochoric ( $J = \det \mathbf{F} = 1$ ) biaxial test in directions  $\mathbf{E}_1$  and  $\mathbf{E}_2$ , with nominal strain ratio  $\zeta$ , the  
 459 matrix representations of the deformation gradient  $\mathbf{F}$  and the right Cauchy-Green deformation  $\mathbf{C}$  are

$$[\mathbf{F}] = \text{diag} \left[ \lambda, \zeta(\lambda - 1) + 1, \frac{1}{\lambda(\zeta(\lambda - 1) + 1)} \right], \quad (64)$$

$$[\mathbf{C}] = \text{diag} \left[ \lambda^2, (\zeta(\lambda - 1) + 1)^2, \frac{1}{\lambda^2(\zeta(\lambda - 1) + 1)^2} \right], \quad (65)$$

460 so that  $\zeta = 1$  describes an equi-biaxial test, for which  $[\mathbf{F}] = \text{diag}[\lambda, \lambda, \lambda^{-2}]$  and  $[\mathbf{C}] = \text{diag}[\lambda^2, \lambda^2, \lambda^{-4}]$ ,  
 461  $0 < \zeta < 1$  means that direction  $\mathbf{E}_1$  is being stretched more than direction  $\mathbf{E}_2$ , and  $\zeta > 1$  vice versa.

462 We assume that the fibres are oriented according to a transversely isotropic von Mises distribution  
 463 (see, e.g., [6, 20, 48]),

$$\varrho(\Theta) = \frac{1}{\pi} \sqrt{\frac{b}{2\pi}} \frac{\exp[b(\cos(2\Theta) + 1)]}{\text{erfi}(\sqrt{2b})}, \quad (66)$$

464 where  $\Theta$  is the angle between the generic direction  $\mathbf{M}$  and the axis of transverse isotropy  $\mathbf{M}_0$ ,  $\text{erf}(x)$   
 465 and  $\text{erfi}(x) = -i \text{erf}(ix)$  denote the error function at  $x$  and the imaginary error function at  $x$ , respec-  
 466 tively [49], and  $b$  is called concentration parameter. In the form reported in Equation (66), the von  
 467 Mises distribution can accommodate both positive and negative values of the concentration parameter  
 468 [50, 48, 19]. The limit  $b \rightarrow +\infty$  describes fibres all aligned in the direction  $\mathbf{M}_0$  of the axis of symmetry,  
 469 the limit  $b \rightarrow 0$  represents isotropy, and the limit  $b \rightarrow -\infty$  describes fibres all lying on the transverse  
 470 plane, which is, by definition, orthogonal to the direction of the axis of symmetry  $\mathbf{M}_0$ . For simplicity,  
 471 we assume that the axis of symmetry  $\mathbf{M}_0$  coincides with the direction  $\mathbf{E}_1$  of axis 1 of the biaxial test  
 472 (Figure 1).

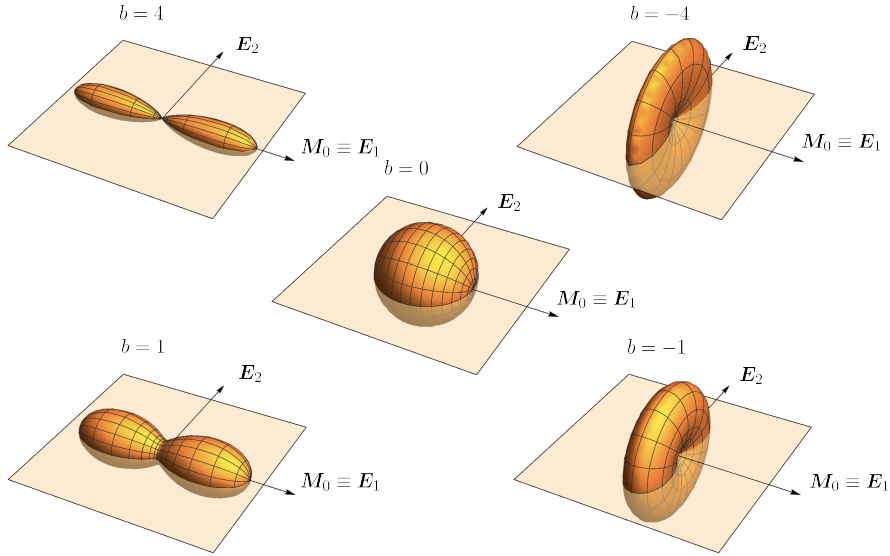


FIGURE 1. Fibre arrangement for the samples undergoing biaxial test in the plane of directions  $\mathbf{E}_1$  and  $\mathbf{E}_2$ . The orientation of the fibres follows a von Mises distribution with axis of symmetry  $\mathbf{M}_0$  parallel to  $\mathbf{E}_1$ . The cases of  $b = 4$  (fibres mostly aligned in the direction of symmetry),  $b = 1$ ,  $b = 0$  (fibres isotropically distributed),  $b = -1$  and  $b = -4$  (fibres mostly lying on the transverse plane) are shown as an example.

473 The approximated integration methods proposed in Section 3 are applied to the calculation of  
 474 the ensemble potential  $\hat{W}_e$  with the provision that, even if the fibres are modelled as extension-only,  
 475 i.e.,  $\hat{W}_{1a} = \mathcal{H}(I_4 - 1)\hat{W}_{1b}$ , the approximation is made with  $\hat{W}_{1a} \equiv \hat{W}_{1b}$ . Indeed, as noted above, we  
 476 *must* renounce to excluding the fibres in contraction when employing our approximation methods.  
 477 The three proposed methods are implemented with the assumptions outlined below:

- 478 1. INEX: the expansion is performed about  $\bar{I}_{40} = 1$  as outlined in Section 3.1, and is truncated at  
 479 order 6, which, in contrast with what happens with the structure tensor expansion STEX, is still  
 480 computationally manageable;
- 481 2. STEX: the expansion is performed about the structure tensor  $\mathbf{A}_0 = \mathbf{M}_0 \otimes \mathbf{M}_0$  of the direction  
 482  $\mathbf{M}_0 \equiv \mathbf{E}_1$  of the axis of symmetry of the potential; the expansion is truncated at order 4, which  
 483 is the maximum order of expansion that the computational resources in our hands allowed;
- 484 3. PARG: the outermost argument of polynomial form in the fibre potential  $\hat{W}_1$  of Equation (61c)  
 485 is given by  $(\mathbf{C} : \mathbf{A} - 1)^2$ , the directional average of which is evaluated.

486 The three proposed methods are compared against:

- 487 4. GOH: replacement of the structure tensor  $\mathbf{A}$  in Equation (61c) with the directional average  $\mathbf{H}$   
 488 of Equation (18) [6];
- 489 5. FESD: integration of the fibre ensemble, at each increment of deformation, by means of the  
 490 method of the spherical  $t$ -designs; note that all fibres, in extension and contraction, are taken  
 491 into account, i.e., as in the three proposed methods, we consider  $\hat{W}_{1a} \equiv \hat{W}_{1b}$ ;
- 492 6. FESDH: integration of the fibre ensemble at the each increment of deformation, as originally  
 493 introduced in [20] for the elastic properties, i.e., with the fibre potential  $\hat{W}_{1a} = \mathcal{H}(I_4 - 1)\hat{W}_{1b}$ ,  
 494 that “kills” the fibres in contraction; this is done to verify under which conditions “sparing” the  
 495 fibres in contraction is an acceptable approximation.

496 The method for the evaluation of the stresses is provided in Appendix E. The values of the elastic  
 497 potential  $W$  and the total Cauchy stresses  $\sigma^{11}$  and  $\sigma^{22}$  are plotted as a function of the stretch  $\lambda$  under  
 498 the deformation described by Equation (65), and are normalised with respect to the material parameter

499  $c_0$  of Equation (61a), while  $c_{1i}$  and  $c_{1a}$  are assumed to have the values  $0.5 c_0$  and  $5 c_0$  respectively. At  
 500 a given value of the strain ratio  $\zeta$ , a set of three plots ( $W$ ,  $\sigma^{11}$  and  $\sigma^{22}$ ) is produced for each value  
 501 of the concentration parameter  $b$  equal to 4 (strong alignment in the direction of symmetry of the  
 502 probability distribution), 1 (weak alignment), 0 (isotropic distribution),  $-1$  (weak alignment on the  
 503 transverse plane),  $-4$  (strong alignment). Figure 2 reports the plots obtained for  $\zeta = 1$  (equi-biaxial  
 504 test), Figure 3 for  $\zeta = 0.5$  (direction  $\mathbf{E}_1$  stretched more than direction  $\mathbf{E}_2$ ) and Figure 4 for  $\zeta = 2$   
 505 (direction  $\mathbf{E}_2$  stretched more than direction  $\mathbf{E}_1$ ). All calculations were performed with *Mathematica*  
 506 (Wolfram Research, Champaign, Illinois, USA).

507 We note (Figure 1) that the fibre distribution with negative values of the concentration parameter  
 508  $b$  is quite unrealistic: in a quasi-two-dimensional sample of a real soft tissue, very few fibres would  
 509 be oriented out-of-plane. We chose to keep this distribution, particularly for the quite extreme case  
 510 of  $b = -4$ , because most of the fibres are oriented out-of-plane, and therefore undergo contraction.  
 511 This offers a way to verify what discrepancy the fibres in contraction cause between the results of the  
 512 FESD calculation that does not exclude them and those of the FESDH calculation that does exclude  
 513 them.

## 514 5. Results

515 The FESDH method, including the Heaviside function in order to “kill” the fibres in contraction, is  
 516 regarded as the “correct” computation, in so far as it rigorously follows the rule of mixtures as in  
 517 Equations (7) and (57). For the tested values of the concentration parameter  $b$  and the strain ratio  
 518  $\zeta$ , the FESD that does not discriminate between fibres in extension and contraction gives very close  
 519 results to the “correct” FESDH method, except for some discrepancy, mainly in the potential, for the  
 520 case of large negative  $b$ . A discrepancy between FESD and FESDH is expected as, for large negative  
 521  $b$ , the orientation of a quite large fraction of the fibres is close to the  $\mathbf{E}_3$  (out of plane) direction, and  
 522 these fibres are therefore in contraction. However, the discrepancy is much smaller than expected (see,  
 523 e.g., the plots for  $b = -1$  and  $b = -4$  in Figure 3).

524 Among all tested methods, the INEX method is systematically the one that gives the results  
 525 closest to those of FESDH/FESD for all values of  $b$  in the equi-biaxial case (Figure 2), almost always  
 526 in the case of  $\zeta = 0.5$ , except in a few cases in which it is slightly outperformed by the PARG method  
 527 and the GOH method (e.g., potential and stresses for  $b = 4$ , Figure 3). For  $\zeta = 2$ , while the INEX  
 528 method is generally the second closest to the spherical designs method (after the STEX method, as  
 529 mentioned below), the fit is not as good as in the cases of  $\zeta = 1$  and  $\zeta = 0.5$ .

530 The STEX method is by far the most inappropriate. As expected, it works best when the proba-  
 531 bility  $\Psi$  is peaked around the direction  $\mathbf{M}_0$  about which the expansion is performed. For the considered  
 532 von Mises probability, this situation corresponds to values of the concentration parameter  $b$  greater  
 533 than zero. Indeed, for the fairly large value  $b = 4$ , it is very close to the FESDH/FESD method.  
 534 However, even for  $b = 4$ , it fails to describe a physically correct behaviour for the stress in direction  
 535 2, when  $\zeta = 0.5$  (Figure 3). The results become generally disastrous for lower values of  $b$ , with several  
 536 occurrences of unphysical behaviour (i.e., decreasing stress in direction 2 for increasing strain), al-  
 537 though in some cases (e.g., particularly for  $\zeta = 2$ , Figure 4) the STEX method evaluates the potential  
 538 very accurately, even for small or negative  $b$ .

539 The PARG method turned out to be a fairly reasonable approximation of the FESDH/FESD  
 540 method. For the equi-biaxial test (Figure 2) and for  $\zeta = 2$  (Figure 4), it is more accurate for positive  
 541 values of  $b$ . However, this trend is reversed for  $\zeta = 0.5$  (Figure 3), i.e., the PARG works better for  
 542 negative values of  $b$ . In general, for given  $b$  and  $\zeta$ , the values of the potential and the stresses yielded by  
 543 the PARG method lie between those of the INEX and the GOH methods, with a few exceptions (e.g.,  
 544  $b = 4$  in the equi-biaxial test and  $b = 4, -4$  for  $\zeta = 0.5$ ) where the PARG method is the closest to the  
 545 FESDH/FESD method. For all tested conditions, the PARG method is closer to the FESDH/FESD  
 546 method than the GOH method is.

547 The GOH method has a good agreement with the FESDH/FESD method for large positive values  
 548 of the concentration parameter  $b$ . However, for  $b = 0$  (isotropic distribution) and negative values of  
 549  $b$ , the behaviour of the GOH method deviates quite substantially from that of the FESDH/FESD  
 550 method. For the tested values of  $b$  and  $\zeta$ , the behaviour of the GOH method is easily predictable, in  
 551 the sense that, for a given  $\zeta$ , a higher value of  $b$  necessarily means a behaviour closer to FESDH/FESD,  
 552 and there seems to be no exceptions.

553 To give an idea about the computational time for each method, we show in Table 1 the time  
 554 required to produce the curves for the equi-biaxial test reported in Figure 2 for  $b = 4$ . In order to  
 555 examine quantitatively the accuracy of the proposed methods, we provide in Figures 5a and 5b the  
 556 curves describing, for two different values of the concentration parameter  $b$ , the absolute error of the  
 557 elastic potential  $W$ , computed for  $\lambda \in [1.0, 1.6]$  by regarding the FESDH method as the reference  
 558 one, i.e.,  $\mathcal{E}_M := |W_M - W_{\text{FESDH}}|$ , with  $M \in \{\text{STEX, INEX, PARG, GOH, FESD}\}$ . The thin, black  
 559 lines corresponding to the values of the absolute error 0.05 for  $b = 4$ , and 0.1 for  $b = -4$  define a  
 560 threshold that identifies, for each value of the concentration parameter, a maximal range of validity,  
 561 i.e., the maximal subset of the stretch interval  $[1.0, 1.6]$  within which the absolute error is assumed  
 562 to be acceptable. Furthermore, for a given value of  $\lambda$  belonging to this range, i.e.,  $\lambda = 1.3$ , Table 2  
 563 and 3 report the values of the relative error of the elastic potential and the stress  $\sigma^{11}$  for varying  
 564 concentration parameter  $b$ . In doing this, we take the FESDH method as the term of comparison.

565 Clearly, the results obtained by using the FESD approach are by far the closest to the ones  
 566 determined by FESDH. This is because the two procedures differ from each other only by the presence  
 567 of the Heaviside step function. Thus, for situations in which almost all fibres are stretched, there is  
 568 virtually no difference between FESD and FESDH. In contrast, when there is a substantial fraction of  
 569 fibres that are not stretched, the results obtained by employing the FESD deviate from those predicted  
 570 by the FESDH. Specifically, both the amplitude and the sense of the deviations depend on the stretch  
 571  $\lambda$ , concentration parameter  $b$ , and deformation mode  $\zeta$ . For example, the FESD overestimates the  
 572 values of  $W/c_0$  for  $\zeta = 1$  and  $b = -4$  (cf. Figure 2), while it underestimates them for  $\zeta = 2$  and  $b = -4$   
 573 (cf. Figure 4). Looking at Table 2, we also notice that, in contrast to what happens for all other  
 574 methods, the relative error pertaining to INEX decreases with decreasing  $b$ , i.e., when the fibres tend  
 575 to lie transversely to the symmetry axis. We argue that this result is related to the fact that the INEX  
 576 method does not select any particular structure tensor for the Taylor expansion formula approximating  
 577 the elastic potential. On the contrary, since the STEX method necessitates to specify the structure  
 578 tensor around which the Taylor expansion formula is constructed, it produces a comparatively small  
 579 absolute error (cf. Figure 5a) when the fibres are concentrated around a given direction ( $b = 4$ ), while  
 580 its accuracy deteriorates for decreasing  $b$ , i.e., when the fibres tend to deviate from that direction.

581 We notice that, for  $b = 4$ , the INEX and PARG approximations are the closest to FESDH/FESD.  
 582 For the case of PARG, this may be due to the fact that this method does not substitute  $\hat{\mathcal{F}}_1$  with its  
 583 Taylor polynomial but, rather, it calculates an *exact* average of the polynomial argument of the fibre  
 584 constitutive function. Thus, the more the fibres are peaked around a given direction, the more accurate  
 585 the PARG method becomes. Looking at the columns of Tables 2 and 3 relative to the INEX and PARG  
 586 methods, we notice that the choice of the “optimal” approximation criterion is quite problem-dependent  
 587 (i.e., it depends on  $b$ ). Consequently, there could be cases (e.g., in inhomogeneous problems, or if  $b$   
 588 changes in time due to some sort of tissue remodelling) in which the approximation method has to  
 589 be chosen adaptively, thereby switching from one to the other in order to minimise the error. For  
 590 completeness, we mention that the relative errors associated with the stress  $\sigma^{11}$  are not monotonic  
 591 functions of  $b$  for the FESD and the PARG methods. A plausible explanation for this behaviour could  
 592 be their capability of resolving the fibre orientation with increasing dispersion (i.e., with  $b \rightarrow -\infty$ ).

593 All the methods belonging to the class of approximations not calling for step-by-step integrations  
 594 (such as the algorithms based on the spherical designs) fail to be accurate after some “threshold” value  
 595 of the stretch that depends on the deformation mode (biaxial, equi-biaxial, etc.) as well as on the  
 596 concentration parameter associated with the chosen probability density distribution.

597 As is visible in the plots of the components of Cauchy stress, the main influence on the mono-  
 598 tonicity and convexity of the curves is given by the interplay between the concentration parameter,  $b$ ,  
 599 which characterises the von Mises distribution, and the parameter  $\zeta$ , which defines the deformation  
 600 mode. In particular, for  $\zeta = 1$  and  $\zeta = 0.5$ , the stress curves lose convexity with decreasing  $b$ . Indeed,  
 601 when the deformation along the symmetry axis is greater than, or equal to, the deformation in the  
 602 transverse plane, on which the fibres tend to lie for decreasing  $b$ , the STEX method is the one that  
 603 deviates the most from the FESDH predictions, thereby introducing unphysical stiffnesses (cf. e.g.,  
 604 Figures 2 and 3).

605 Moreover, a computation of the stress, e.g.,  $\sigma^{11}$ , shows that the summand of  $\sigma^{11}$  responsible  
 606 for the concavity in the stress curves is given by the Lagrange multiplier introduced to account for  
 607 the incompressibility constraint. To show that this is actually the case, we take as example the stress  
 608 approximated by means of the INEX method. Hereafter, for ease of demonstration, we write its  
 609 expression only for the Taylor expansion of the elastic potential up to the second order. In the figures,  
 610 however, we show also the stress for the case of an expansion up to the sixth order. By using the  
 611 elastic potential (57), along with (61a)–(61c) and arresting the Taylor expansion of  $\hat{W}_1(\mathbf{C}, \mathbf{A})$  at the  
 612 order  $n = 2$ , the approximated expression of the constitutive part of the second Piola-Kirchhoff stress  
 613 tensor reads

$$\mathbf{S}_c^{\text{app}} := 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}) = \Phi_0 c_0 \mathbf{G}^{-1} + \Phi_1 c_{1i} \mathbf{G}^{-1} + \mathbf{S}_{1a}^{\text{app}}, \quad (67)$$

614 where (cf. (98))

$$\mathbf{S}_{1a}^{\text{app}} = 2\Phi_1 c_{1a} [\mathbb{H}_2 : \mathbf{C} - \mathbb{H}_1]. \quad (68)$$

615 Because of the imposed incompressibility, the overall second Piola-Kirchhoff stress tensor is given by  
 616  $\mathbf{S}^{\text{app}} = -p\mathbf{C}^{-1} + \mathbf{S}_c^{\text{app}}$ , where  $p$  is the Lagrange multiplier (*not* coinciding with the pressure in the  
 617 present treatment) associated with the incompressibility constraint. Accordingly, for an equi-biaxial  
 618 test (i.e., when  $[\mathbf{C}] = \text{diag}\{\lambda^2, \lambda^2, \lambda^{-4}\}$ ), the component  $\sigma^{11}$  of the Cauchy stress tensor becomes

$$\sigma^{11} = -p + (\Phi_0 c_0 + \Phi_1 c_{1i}) \lambda^4 + \sigma_{1a}^{11}, \quad (69)$$

619 with  $\sigma_c^{11} := (\Phi_0 c_0 + \Phi_1 c_{1i}) \lambda^4 + \sigma_{1a}^{11}$  being the constitutive part of  $\sigma^{11}$  and

$$\sigma_{1a}^{11} = 2\Phi_1 c_{1a} [(\mathbb{H}_2)^{1111} \lambda^4 + (\mathbb{H}_2)^{1122} \lambda^4 + (\mathbb{H}_2)^{1133} \frac{1}{\lambda^2} - (\mathbb{H}_1)^{11} \lambda^2]. \quad (70)$$

620 Plotting  $\sigma^{11}$  versus  $\lambda$  shows that  $\sigma_c^{11}$  is a convex function of  $\lambda$ , whereas the negative of the Lagrange  
 621 multiplier,  $-p$ , is a concave function  $\lambda$ . Since  $\sigma_c^{11}$  grows almost linearly for values of  $\lambda$  close to unity,  
 622 the composition  $\sigma^{11} = -p + \sigma_c^{11}$  turns out to be non-convex. This is depicted in Figures 6a and 6b,  
 623 where the effect of raising the order of the approximation is testified by the increasing curvature, for  
 large enough values of  $\lambda$  of the constitutive part of stress.

TABLE 1. Computational time [s] for graphs at  $\zeta = 1$  and  $b = 4$  and stretch range  $\lambda \in [1.0, 1.6]$ ; time increment in FESD and FESDH is 40 ms.

Quantity	STEX	INEX	PARG	GOH	FESD	FESDH
elastic potential $W$	0.66	0.06	0.05	0.06	1.66	2.81
stress $\sigma^{11}$	0.78	0.44	0.08	0.14	25.20	38.80
stress $\sigma^{22}$	0.19	0.55	0.08	0.06	25.34	38.17

624

## 625 6. Summary and Discussion

626 In a biological tissue (or industrial material) with a statistical distribution of reinforcing fibres, the  
 627 effect of the fibres on the overall constitutive function  $\hat{\mathcal{F}}$  of a given physical quantity can be obtained

TABLE 2. Relative error [%] for the elastic potential  $W$ , in the equi-biaxial test ( $\zeta = 1$ ) and at  $\lambda = 1.3$ .

$b$	FESD	INEX	STEX	PARG	GOH
4	0.06	0.04	1.30	1.13	4.85
1	3.87	4.63	45.35	1.26	26.92
0	9.20	10.26	102.85	6.44	14.86
-1	14.99	16.45	163.98	12.49	29.02
-4	23.02	25.99	250.77	21.95	39.35

TABLE 3. Relative error [%] for the stress  $\sigma^{11}$ , in the equi-biaxial test ( $\zeta = 1$ ) and at  $\lambda = 1.3$ .

$b$	FESD	INEX	STEX	PARG	GOH
4	0.0007	0.7728	3.5272	2.9521	6.9328
1	0.7915	1.2773	181.0450	6.5662	34.0420
0	3.0387	3.8109	552.9186	4.2988	41.0398
-1	7.6397	8.7854	1205.4554	2.3712	35.4135
-4	20.4896	22.8115	2664.3860	19.3050	13.5467

628 by integrating the fibre constitutive function  $\hat{\mathcal{F}}_1$ , weighted by an orientation probability distribution,  
629 over the set of all directions in space (cf. Equation (7)). The resulting integral, called fibre ensemble  
630  $\hat{\mathcal{F}}_e$  in this work (cf. Equation (8)), can in general only be evaluated numerically at each increment  
631 of deformation, since the deformation (usually represented by the right Cauchy-Green deformation  
632 tensor  $\mathbf{C}$ ) cannot be factorised out of the integral sign, except in the case in which  $\hat{\mathcal{F}}_1$  is expressed as a  
633 tensor-power polynomial in the structure tensor  $\mathbf{A}$  [19]. Even though the numerical integration of  $\hat{\mathcal{F}}_e$  is  
634 flexible and can be made very accurate, it is sometimes computationally expensive. Indeed, especially  
635 in time-dependent nonlinear problems, it has to be “called” at each time-step and at each iteration of  
636 some nonlinear solver, thereby increasing computational costs. With the aim of containing these costs,  
637 we exploited polynomials to achieve a single, direct integration of a given fibre constitutive function  
638  $\hat{\mathcal{F}}_1$ , and thus an approximation of the corresponding fibre ensemble  $\hat{\mathcal{F}}_e$ . We elaborated three methods:  
639 a Taylor expansion in the transversely isotropic invariants (INEX method), which we presented in the  
640 case of functions of the fourth invariant  $I_4$  alone, but which can be seamlessly extended to functions  
641 including also the fifth invariant,  $I_5$ ; a Taylor expansion in the structure tensor  $\mathbf{A}$  about a given  
642 value  $\mathbf{A}_0$  corresponding to a direction  $\mathbf{M}_0$  (STEX method); and, for the case of fibre constitutive  
643 functions  $\hat{\mathcal{F}}_1$  expressed as some function of a polynomial  $\mathcal{P}(\mathbf{C}, \mathbf{A})$ , the replacement of  $\mathcal{P}(\mathbf{C}, \mathbf{A})$  with  
644 its directional average (PARG method). The latter method is similar to the GOH method proposed  
645 in [6]. We emphasise that our methods are not meant to replace the step-by-step integration, which  
646 is considered to be the most accurate method to represent a constitutive function expressed by the  
647 rule of mixtures, and was regarded as term of comparison to test the accuracy of our approximations.  
648 Rather, our methods aim to offer alternative options to step-by-step integration schemes, such as the  
649 FESD and the FESDH, since the direct integration of constitutive functions can be performed *before*  
650 discretising the system in time and before starting any iterative scheme for solving nonlinear problems.

651 We chose to test the proposed methods for the case of the elastic potential and the associated  
652 stress. We compared the proposed methods to the “exact” integration, performed at each increment  
653 of deformation by means of the method of the spherical designs [21, 34, 20], which we have called here  
654 FESD method, as well as to the GOH method [6]. A calculation including the Heaviside function was  
655 made with the method of the spherical designs (FESDH method) in order to eliminate the contribution  
656 of the fibres in contraction and to estimate in which conditions counting also the fibres undergoing  
657 contraction is acceptable.

658 As mentioned in Section 5, for most of the tested conditions, the INEX method (expansion in the  
 659 invariants) was the closest to the “rigorous” integration performed with FESDH/FESD method (fibre  
 660 ensemble evaluated by means of the method of the spherical designs, with or without the Heaviside  
 661 function to eliminate the fibres in contraction). What really distinguishes the INEX method from the  
 662 other ones is that its accuracy is weakly dependent on the distribution of the fibres (concentration  
 663 parameter  $b$ ). Moreover, one could improve the accuracy of the approximation by simply computing a  
 664 higher-order expansion. In contrast, the accuracy of the other tested methods shows a clear dependence  
 665 on the distribution of the fibres, i.e., their accuracy is higher for high values of  $b$  and decreases, often  
 666 sensibly, as  $b$  becomes negative.

667 In conclusion, when implementing the fibre ensemble (Equation (8)) arising from the rule of  
 668 mixtures into Finite Elements, the method of Taylor expansion in the invariants (INEX) constitutes a  
 669 valid, computationally inexpensive, direct integration method, alternative to programming a complex  
 670 user subroutine that employs the method of the spherical designs to perform the directional averages  
 671 at each increment of deformation. The integrals  $\mathbb{H}_p$  needed in the INEX method (Equations (27) and  
 672 (35)) can be evaluated directly (Equation (13)) with a commercially available calculation package  
 673 such as *Mathematica* (Wolfram Research, Champaign, Illinois, USA), and then exported into a much  
 674 simpler user subroutine to be used in the Finite Element code. In fact, once the highest order  $2n$  of  
 675 the expansion is set, one can simply calculate the corresponding tensor  $\mathbb{H}_n$ , and then obtain all tensors  
 676  $\mathbb{H}_p$  of lower order  $2 \leq 2p < 2n$  by contracting any  $n - p$  pairs of indices [31]. Moreover, for the case  
 677 of the von Mises distribution, which is determined univocally by the concentration parameter  $b$ , the  
 678 tensors  $\mathbb{H}_p$  can be exported as functions of  $b$ , which has the obvious advantage of providing a function  
 679 rather than an array of values. It is in our future plans to develop similar methods for fibre-reinforced  
 680 biological tissues seen as higher-gradient materials (see, e.g., [51, 52, 53]).

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 688 Excellence in Research) [AG].

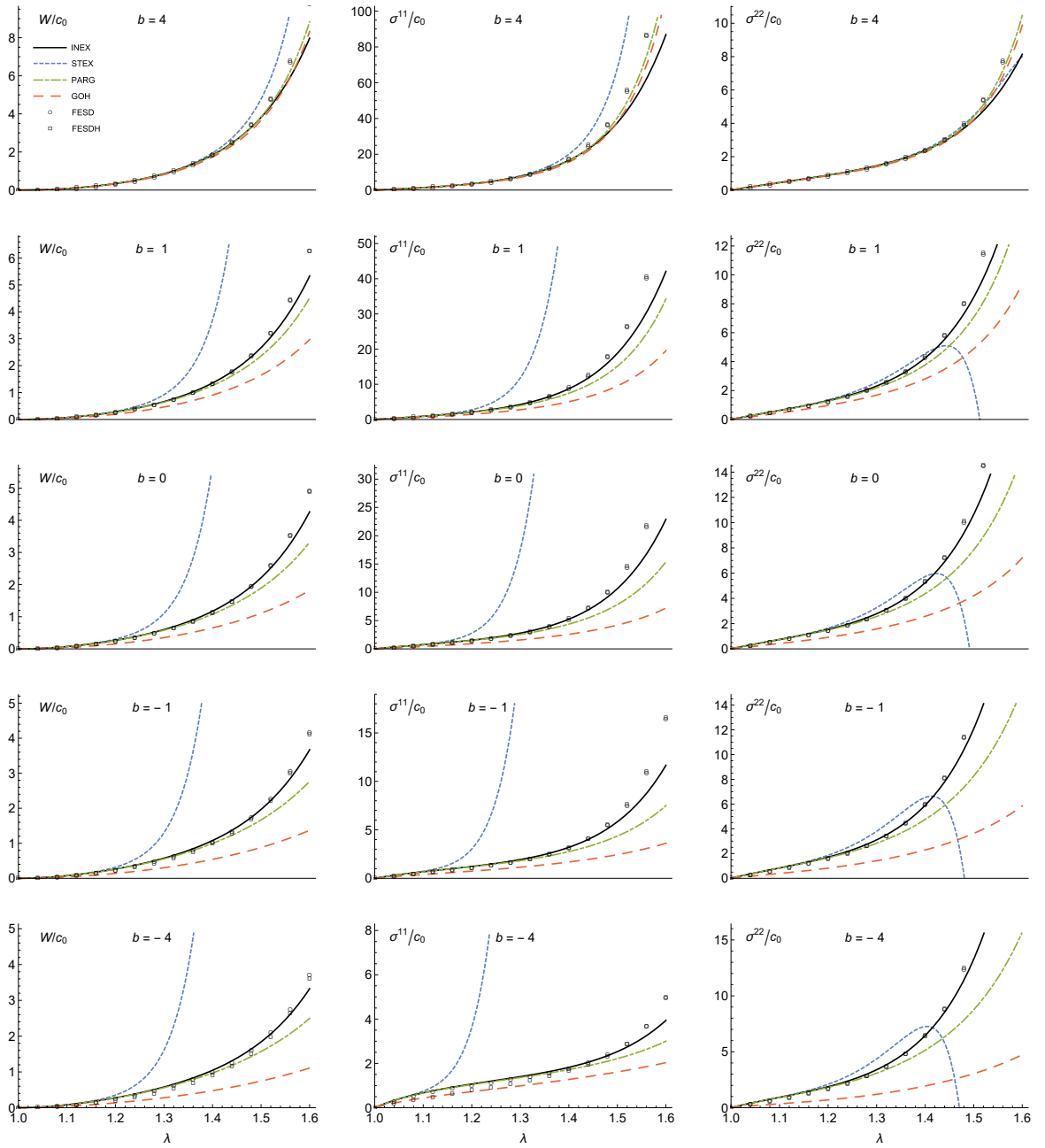


FIGURE 2. Elastic potential and stress for the equi-biaxial test ( $\zeta = 1$ )

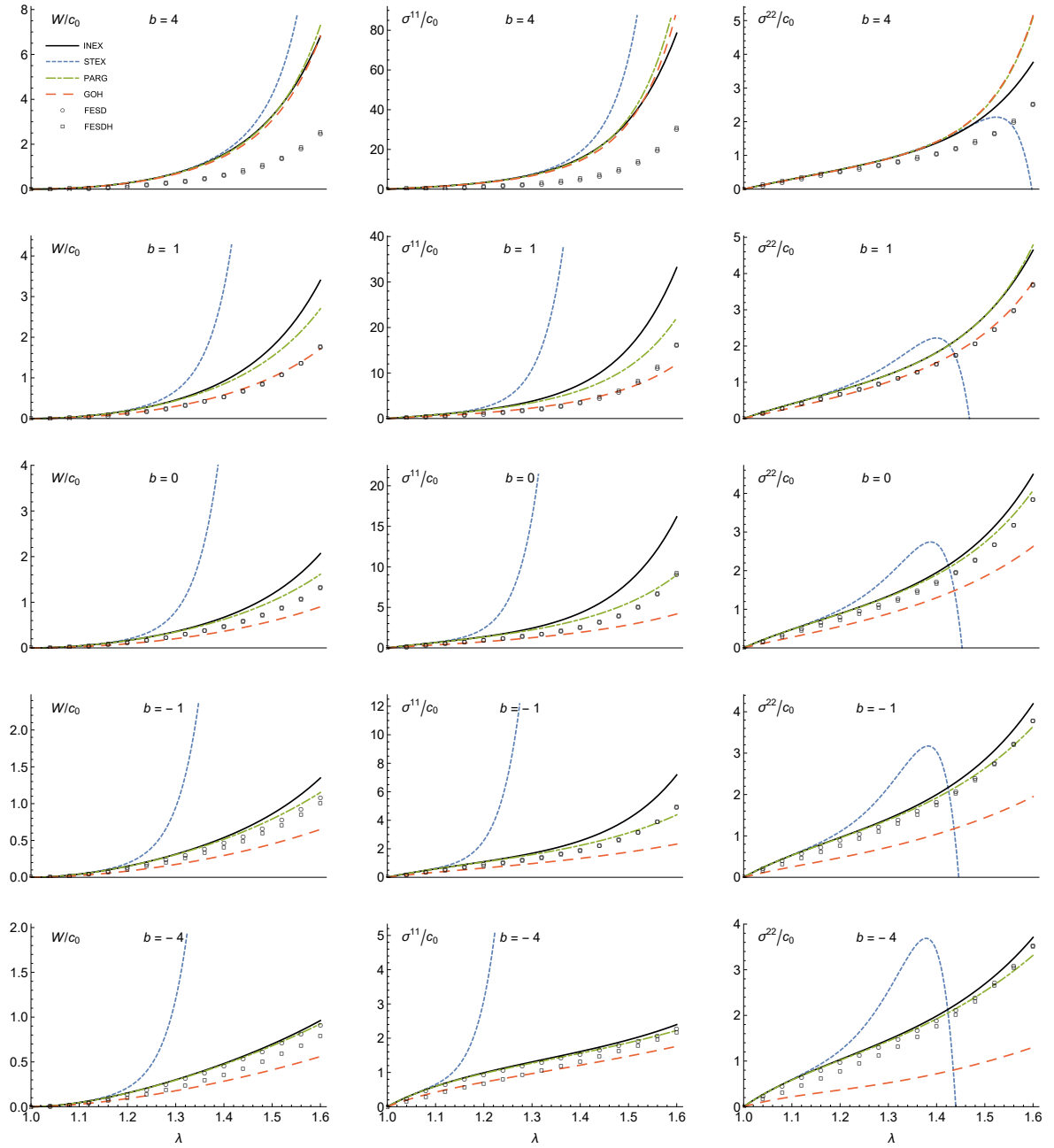


FIGURE 3. Elastic potential and stress for a biaxial test with  $\zeta = 0.5$

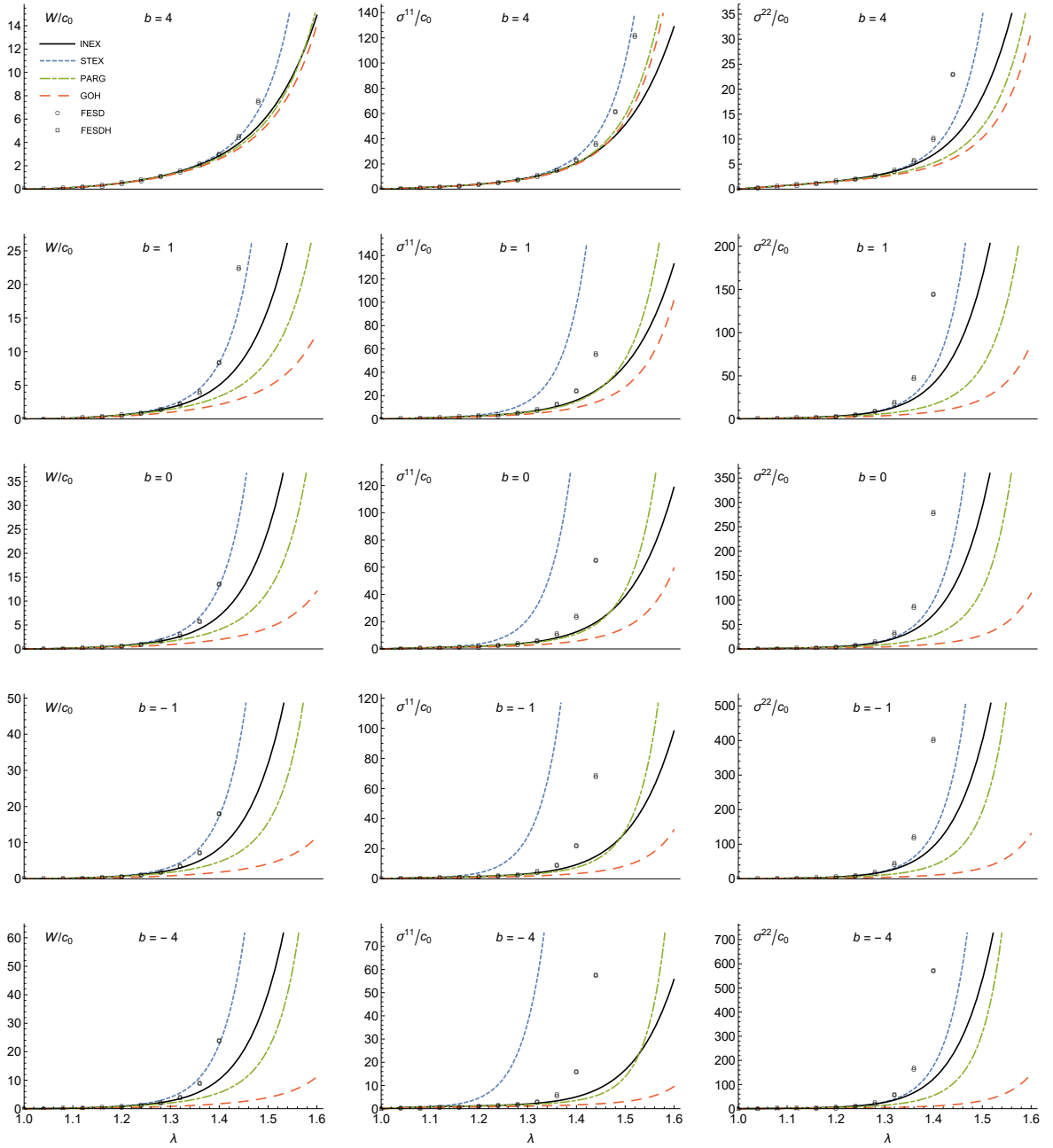


FIGURE 4. Elastic potential and stress for a biaxial test with  $\zeta = 2$

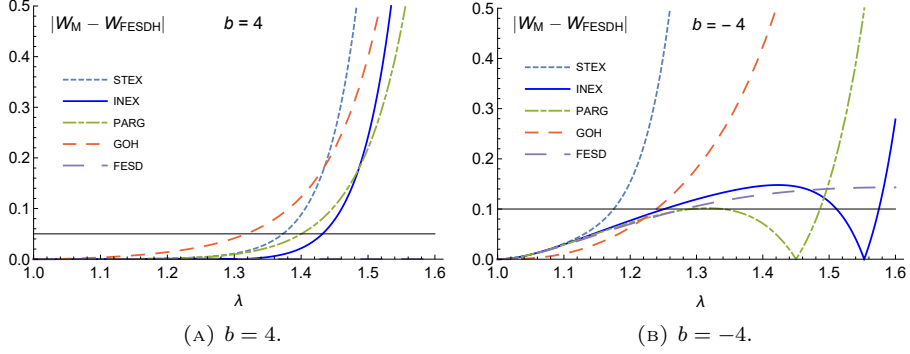


FIGURE 5. Absolute error  $|W_M - W_{\text{FESDH}}|$  of the elastic potential  $W$ , with  $M \in \{\text{STEX, INEX, PARG, GOH, FESD}\}$ , for two different values of the concentration parameter,  $b = 4$  and  $b = -4$ .

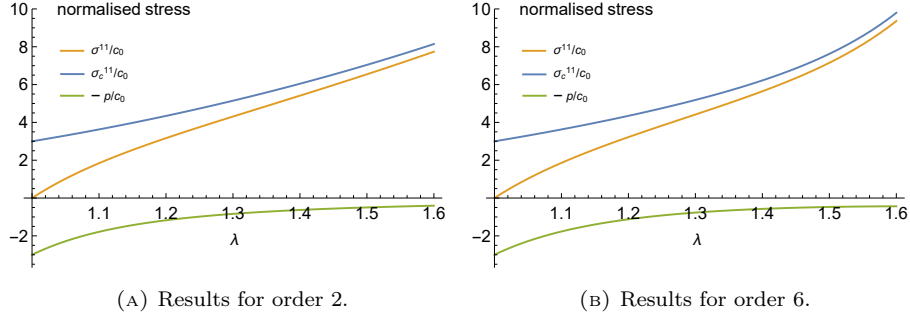


FIGURE 6. Normalised stresses and pressure for the INEX method (orders 2 and 6).

## 689 Appendix A. Continuum Mechanics Notation and Definitions

690 The deformation  $\chi$  maps material points  $X = (X^1, X^2, X^3)$  in the reference configuration  $\mathcal{B}$  into  
691 spatial points  $x = (x^1, x^2, x^3)$  in the physical space  $\mathcal{S}$ . The deformation gradient  $\mathbf{F}$  has components  
692  $F^a_A = \chi^a_{,A}$  and pushes-forward material vectors  $\mathbf{W}$  with components  $W^A$  into spatial vectors  $\mathbf{FW}$   
693 with components  $F^a_A W^A$ . The inverse  $\mathbf{F}^{-1}$  pulls-back spatial vectors  $\mathbf{w}$  with components  $w^a$  into  
694 material vectors  $\mathbf{F}^{-1}\mathbf{w}$  with components  $(\mathbf{F}^{-1})^A_a w^a$ . The transpose  $\mathbf{F}^T$  pulls-back spatial covectors  $\boldsymbol{\pi}$   
695 with components  $\pi_a$  into material covectors  $\mathbf{F}^T\boldsymbol{\pi}$  with components  $(\mathbf{F}^T)_A^a \pi_a = F^a_A \pi_a$ . The inverse  
696 transpose  $\mathbf{F}^{-T}$  pushes-forward material covectors  $\boldsymbol{\Pi}$  with components  $\Pi_A$  into spatial covectors  $\mathbf{F}^{-T}\boldsymbol{\Pi}$   
697 with components  $(\mathbf{F}^{-T})_a^A \Pi_A = (\mathbf{F}^{-1})^A_a \Pi_A$ . The determinant  $J = \det \mathbf{F}$  is called volume ratio and  
698 measures volumetric deformation.

699 The reference configuration  $\mathcal{B}$  and the physical space  $\mathcal{S}$  are equipped with metric tensors  $\mathbf{G}$   
700 and  $\mathbf{g}$ , respectively, which define the scalar products of material and spatial vectors as  $\langle \mathbf{W}, \mathbf{Y} \rangle =$   
701  $\mathbf{W} \cdot \mathbf{Y} = \mathbf{W} \mathbf{G} \mathbf{Y} = W^A G_{AB} Y^B$  and  $\langle \mathbf{w}, \mathbf{y} \rangle = \mathbf{w} \cdot \mathbf{y} = \mathbf{w} \mathbf{g} \mathbf{y} = w^a g_{ab} y^b$ , respectively. The pull-back of  
702 the spatial metric  $\mathbf{g}$  is the right Cauchy-Green deformation tensor  $\mathbf{C} = \mathbf{F}^T \mathbf{g} \mathbf{F} = \mathbf{F}^T \cdot \mathbf{F}$ , with com-  
703 ponents  $(\mathbf{F}^T)_A^a g_{ab} F^b_B = F^a_A g_{ab} F^b_B$ . The pull-back of the inverse spatial metric  $\mathbf{g}^{-1}$  is the Piola  
704 deformation tensor  $\mathbf{B} = \mathbf{F}^{-1} \mathbf{g}^{-1} \mathbf{F}^{-T} = \mathbf{F}^{-1} \cdot \mathbf{F}^{-T} = \mathbf{C}^{-1}$ , with components  $(\mathbf{F}^{-1})^A_a g^{ab} (\mathbf{F}^{-T})_b^B =$   
705  $(\mathbf{F}^{-1})^A_a g^{ab} (\mathbf{F}^{-1})^B_b$ . The difference between the *pulled-back material metric*  $\mathbf{C}$  and the natural ma-  
706 terial metric  $\mathbf{G}$ , normalised by the coefficient  $1/2$ , is the Green-Lagrange strain  $\mathbf{E} = \frac{1}{2}(\mathbf{C} - \mathbf{G})$ .

## 707 Appendix B. Invariants of the Deformation

708 Isotropy is the material symmetry defined as the invariance of a given physical quantity with respect  
709 to the whole group of rotations [54]. For an isotropic material, the three scalar invariants of the  
710 deformation are

$$I_1 = \text{tr}(\mathbf{C}) = \mathbf{G}^{-1} : \mathbf{C}, \quad (71a)$$

$$I_2 = \frac{1}{2} [(\text{tr}(\mathbf{C}))^2 - \text{tr}(\mathbf{C}^2)], \quad (71b)$$

$$I_3 = \det(\mathbf{C}). \quad (71c)$$

711 Given a vector  $\mathbf{M}$ , belonging to the material (or referential) unit sphere  $\mathbb{S}^2\mathcal{B} = \{\mathbf{M} : \|\mathbf{M}\| = 1\}$ ,  
712 transverse isotropy with respect to the direction  $\mathbf{M}$  is defined as the invariance under arbitrary  
713 rotations about  $\mathbf{M}$ . When the material properties do not depend on the sense of  $\mathbf{M}$ , it is possible to  
714 introduce the structure tensor  $\mathbf{A} = \mathbf{M} \otimes \mathbf{M}$ , which is invariant for inversions of  $\mathbf{M}$ , i.e., transformations  
715 of the type  $\mathbf{M} \mapsto -\mathbf{M}$ . For the case of transverse isotropy, two additional invariants are defined as a  
716 function of the structure tensor  $\mathbf{A}$  [55]:

$$I_4 = \mathbf{C} : \mathbf{A} = \mathbf{M} \mathbf{C} \mathbf{M} = (\mathbf{F} \mathbf{M}) \cdot (\mathbf{F} \mathbf{M}) = \lambda_{\mathbf{M}}^2, \quad (72a)$$

$$I_5 = \mathbf{C}^2 : \mathbf{A}, \quad (72b)$$

717 where  $\lambda_{\mathbf{M}}^2$  is the square of the stretch in direction  $\mathbf{M}$ . By enforcing the volumetric-distortional decom-  
718 position of  $\mathbf{C}$ , i.e.,  $\mathbf{C} = J^{2/3} \bar{\mathbf{C}}$  (see Section 2.1), the invariants introduced in Equations (71a)–(72b)  
719 can be rewritten as  $I_1 = J^{2/3} \bar{I}_1$ ,  $I_2 = J^{4/3} \bar{I}_2$ ,  $I_3 = J^2 \bar{I}_3$ ,  $I_4 = J^{2/3} \bar{I}_4$ , and  $I_5 = J^{4/3} \bar{I}_5$ , where  
720 the generic  $\bar{I}_q$ , with  $q = 1, \dots, 5$ , is obtained by substituting  $\mathbf{C}$  with  $\bar{\mathbf{C}}$  in the expression of the  
721 corresponding invariant  $I_q$ . Clearly, it holds that  $\bar{I}_3 = \det \bar{\mathbf{C}} = 1$ .

## 722 Appendix C. Hyperelasticity

723 An elastic material is called hyperelastic if the stress can be obtained by differentiation of a function,  
724 called elastic potential or elastic strain energy density, with respect to the conjugated measure of  
725 strain/deformation. If the potential is given as a function  $W = \hat{W}(\mathbf{C})$  of the right Cauchy-Green  
726 deformation  $\mathbf{C}$ , the second Piola-Kirchhoff stress is obtained as

$$\mathbf{S} = 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}). \quad (73)$$

727 The Cauchy stress is obtained by means of the forward Piola transformation

$$\boldsymbol{\sigma} = J^{-1} \mathbf{F} \mathbf{S} \mathbf{F}^T = J^{-1} \mathbf{F} \left[ 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T. \quad (74)$$

728 If the material is incompressible, the kinematical constraint  $J = 1$  of isochoric (i.e., volume-preserving)  
729 motion must be enforced by means of the Lagrange multiplier  $p$  (which does *not* have the physical  
730 meaning of hydrostatic pressure in this treatment), and the second Piola-Kirchhoff stress tensor is  
731 given by

$$\mathbf{S} = -J p \mathbf{B} + 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}), \quad (75)$$

732 where  $\mathbf{B} = \mathbf{C}^{-1}$  is the Piola deformation tensor. To obtain the Cauchy stress  $\boldsymbol{\sigma}$ , a forward Piola  
733 transformation is performed on  $\mathbf{S}$ , i.e.,

$$\boldsymbol{\sigma} = -p \mathbf{g}^{-1} + J^{-1} \mathbf{F} \left[ 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T, \quad (76)$$

734 where  $\mathbf{g}^{-1}$  is the inverse spatial metric tensor, which plays the role of the “contravariant” identity  
735 tensor.

736 **Appendix D. Admissible Interval of  $I_4$  or  $\bar{I}_4$**

737 We want to prove that, under a deformation  $\mathbf{C}$ , the admissible values of  $I_4 = \mathbf{C} : \mathbf{A}$  belong to the  
 738 interval  $[\lambda_{\min}^2, \lambda_{\max}^2]$ , where  $\lambda_{\min}^2$  and  $\lambda_{\max}^2$  are the minimum and maximum eigenvalues of  $\mathbf{C}$ , for  
 739 every  $\mathbf{A} = \mathbf{M} \otimes \mathbf{M}$ . The same holds for the case of the distortional part  $\bar{\mathbf{C}}$  of the deformation, i.e.,  
 740  $\bar{I}_4 \in [\bar{\lambda}_{\min}^2, \bar{\lambda}_{\max}^2]$ , where  $\bar{\lambda}_{\min}^2$  and  $\bar{\lambda}_{\max}^2$  are the minimum and maximum eigenvalues of  $\bar{\mathbf{C}}$ .

741 Let us consider the representation of the deformation  $\mathbf{C}$  in terms of its eigenvalues,

$$[\mathbf{C}] = [C_{AB}] = \text{diag}[\lambda_1^2, \lambda_2^2, \lambda_3^2], \quad (77)$$

742 Note that we can write the fourth invariant as

$$I_4 = \mathbf{C} : \mathbf{A} = \mathbf{C} : (\mathbf{M} \otimes \mathbf{M}) = \mathbf{M} \mathbf{C} \mathbf{M} = M^A C_{AB} M^B, \quad (78)$$

743 from which we obtain the equation of an ellipsoid, with matrix  $[\frac{1}{I_4} C_{AB}] = \text{diag}[\frac{\lambda_1^2}{I_4}, \frac{\lambda_2^2}{I_4}, \frac{\lambda_3^2}{I_4}]$ , i.e.,

$$M^A \left[ \frac{1}{I_4} C_{AB} \right] M^B = 1, \quad \Rightarrow \quad \frac{(M^1)^2}{I_4/\lambda_1^2} + \frac{(M^2)^2}{I_4/\lambda_2^2} + \frac{(M^3)^2}{I_4/\lambda_3^2} = 1, \quad (79)$$

744 and semi-axes given by  $\sqrt{I_4}/\lambda_\alpha$ . If we also impose the fact that  $\mathbf{M}$  is a unit vector, we obtain

$$\|\mathbf{M}\|^2 = \mathbf{M} \cdot \mathbf{M} = \mathbf{M} \mathbf{G} \mathbf{M} = M^A G_{AB} M^B = 1. \quad (80)$$

745 Assuming Cartesian coordinates for simplicity of representation, we have that the matrix of the metric  
 746 tensor  $\mathbf{G}$  reduces to the identity, i.e.,  $G_{AB} = \delta_{AB}$ , and the equation above reduces to the equation of  
 747 the unit sphere

$$M^A \delta_{AB} M^B = 1, \quad \Rightarrow \quad (M^1)^2 + (M^2)^2 + (M^3)^2 = 1 \quad (81)$$

748 The admissible values of  $I_4$  are those for which the ellipsoid and the sphere intersect, i.e., the  
 749 system of equations given by (79) and (81) admits a solution. Evidently, the minimum value of  $I_4$  is  
 750 attained when the *major* semi-axis of the ellipsoid ( $\sqrt{I_4}/\lambda_{\min}$ ) equals the radius of the sphere, i.e.,  
 751  $I_4 = \lambda_{\min}^2$  (Figure 7a), and the maximum value of  $I_4$  is attained when the *minor* semi-axis of the  
 752 ellipsoid ( $\sqrt{I_4}/\lambda_{\max}$ ) equals the radius of the sphere, i.e.,  $I_4 = \lambda_{\max}^2$  (Figure 7c). For  $I_4 \in \mathring{\Lambda}(\mathbf{C}) =$   
 753  $]\lambda_{\min}^2, \lambda_{\max}^2[$ , the intersection of the the ellipsoid and the sphere is given by two symmetric curves  
 754 (Figure 7b).

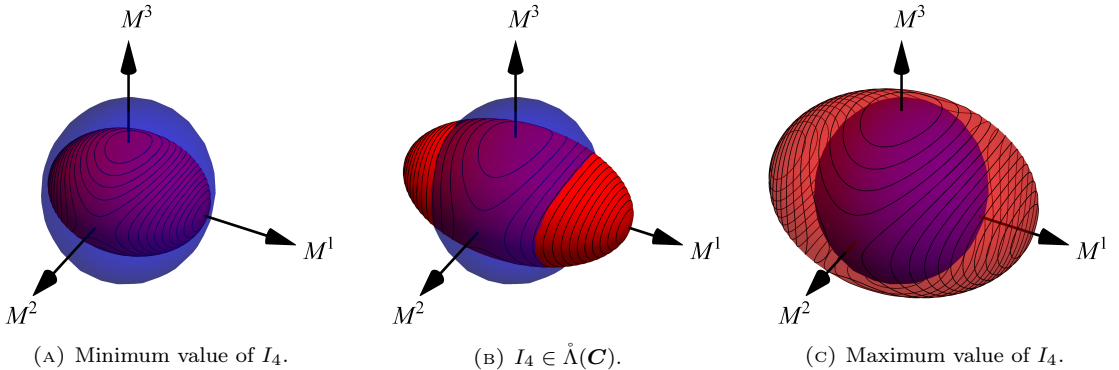


FIGURE 7. Graphical representation of the admissible values of  $I_4$ .

## 755 Appendix E. Example of Evaluation of the Stress

756 In the INEX and STEx methods, our strategy for the evaluation of the stress was to first expand the  
 757 ensemble potential and then differentiate the Taylor-expanded potential with respect to the deformation.  
 758 This was aimed at minimising the number of integrals to be performed. As an example, let us  
 759 look at the evaluation of the stress for the INEX method, in which, if the incompressibility constraint  
 760  $J = 1$  is enforced, we have

$$W_e \simeq \mathcal{G}_n = \hat{\mathcal{G}}_n(\mathbf{C}, \Psi) = \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \hat{W}_1}{\partial \bar{I}_4^{(j)}}(1, 1) \sum_{k=0}^j \binom{j}{k} (-1)^k \langle \mathbf{C}^{\otimes(j-k)} | \mathbb{H}_{j-k} \rangle. \quad (82)$$

761 The Cauchy stress is computed according to Equation (76), in which  $J = 1$  can be set, i.e.,

$$\boldsymbol{\sigma} = \mathbf{F} \mathbf{S} \mathbf{F}^T = -p \mathbf{g}^{-1} + \mathbf{F} \left[ 2 \frac{\partial \hat{W}}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T = -p \mathbf{g}^{-1} + \boldsymbol{\sigma}_c, \quad (83)$$

762 where  $p$  is the Lagrange multiplier associated with the condition  $J = 1$ , and  $\boldsymbol{\sigma}_c$  is the constitutive part  
 763 of  $\boldsymbol{\sigma}$  (here,  $p$  is not the hydrostatic pressure, because  $\boldsymbol{\sigma}_c$  need not be deviatoric in this formulation of  
 764 incompressible hyperelasticity). By using the elastic potential

$$\hat{W}(\mathbf{C}) = \Phi_0 \hat{W}_0(\mathbf{C}) + \Phi_1 \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \hat{W}_1(\mathbf{C}, \mathbf{A}), \quad (84)$$

765  $\boldsymbol{\sigma}_c$  can be written as

$$\begin{aligned} \boldsymbol{\sigma}_c &= \mathbf{F} \left[ 2\Phi_0 \frac{\partial \hat{W}_0}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T + \mathbf{F} \left[ 2\Phi_1 \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \frac{\partial \hat{W}_1}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}) \right] \mathbf{F}^T \\ &= \mathbf{F} \left[ 2\Phi_0 \frac{\partial \hat{W}_0}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T + \mathbf{F} \left[ 2\Phi_1 \frac{\partial \hat{W}_e}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T, \end{aligned} \quad (85)$$

766 where  $\hat{W}_e(\mathbf{C}) = \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \hat{W}_1(\mathbf{C}, \mathbf{A})$  is the fibre ensemble elastic potential. Next,  $\hat{W}_1(\mathbf{C}, \mathbf{A})$  is written  
 767 as the sum of an isotropic and an anisotropic contribution, i.e.,

$$\hat{W}_1(\mathbf{C}, \mathbf{A}) = \hat{W}_{1i}(\mathbf{C}) + \hat{W}_{1a}(\mathbf{C}, \mathbf{A}), \quad (86)$$

768 and the fibre ensemble potential becomes

$$\hat{W}_e(\mathbf{C}) = \hat{W}_{1i}(\mathbf{C}) + \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \hat{W}_{1a}(\mathbf{C}, \mathbf{A}), \quad (87)$$

769 so that  $\boldsymbol{\sigma}_c$  takes on the form

$$\boldsymbol{\sigma}_c = \mathbf{F} \left[ 2\Phi_0 \frac{\partial \hat{W}_0}{\partial \mathbf{C}}(\mathbf{C}) + 2\Phi_1 \frac{\partial \hat{W}_{1i}}{\partial \mathbf{C}}(\mathbf{C}) \right] \mathbf{F}^T + \mathbf{F} \left[ 2\Phi_1 \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \frac{\partial \hat{W}_{1a}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}) \right] \mathbf{F}^T. \quad (88)$$

770 The general formula (88) should now be specialised according to the approximation method that is  
 771 adopted. Since both  $\hat{W}_0(\mathbf{C})$  and  $\hat{W}_{1i}(\mathbf{C})$  contribute to  $\boldsymbol{\sigma}_c$  in the same way for all methods (indeed,  
 772 they are independent of the direction of the fibres, and thus need not be approximated by any of our  
 773 methods), we can restrict our calculations by focusing on the anisotropic stress contribution of the  
 774 fibres only, i.e.,

$$\boldsymbol{\sigma}_{1a} := \mathbf{F} \left[ 2\Phi_1 \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \frac{\partial \hat{W}_{1a}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}) \right] \mathbf{F}^T. \quad (89)$$

775 Moreover, since the averaging integral in (89) pertains only to the partial second Piola-Kirchhoff stress  
 776 tensor

$$\mathbf{S}_{1a} := 2\Phi_1 \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \frac{\partial \hat{W}_{1a}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}), \quad (90)$$

777 it suffices for our purposes to provide, for each of the four proposed approximation methods, the  
 778 corresponding approximated expression of  $\mathbf{S}_{1a}$ , which we denote by  $\mathbf{S}_{1a}^{\text{app}}$ . The stress  $\mathbf{S}_{1a}$  computed  
 779 according to the FESDH method shall be regarded as “exact”.

780 We recall that, for the FESDH method,  $\hat{W}_{1a}(\mathbf{C}, \mathbf{A}) = \mathcal{H}(\mathbf{C} : \mathbf{A} - 1)\hat{W}_{1b}(\mathbf{C}, \mathbf{A})$ , and  $\mathbf{S}_{1a}$  is given  
 781 by

$$\mathbf{S}_{1a} = 2\Phi_1 \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M})\mathcal{H}(\mathbf{C} : \mathbf{A} - 1) \frac{\partial \hat{W}_{1b}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}). \quad (91)$$

782 In the FESD, INEX, PARG, and STEx methods, we do not premultiply  $\hat{W}_{1b}$  by the Heaviside function,  
 783 so that  $\hat{W}_{1a}(\mathbf{C}, \mathbf{A}) \equiv \hat{W}_{1b}(\mathbf{C}, \mathbf{A})$  holds true. Thus, with reference to the FESD approximation,  $\mathbf{S}_{1a}^{\text{app}}$   
 784 is given by

$$\mathbf{S}_{1a}^{\text{app}} = 2\Phi_1 \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \frac{\partial \hat{W}_{1a}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}) = 2\Phi_1 \frac{\partial}{\partial \mathbf{C}} \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_{1a}(\mathbf{C}, \mathbf{A}). \quad (92)$$

785 For the INEX method, we approximate  $\int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_{1a}(\mathbf{C}, \mathbf{A})$  as

$$\int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_{1a}(\mathbf{C}, \mathbf{A}) \simeq \sum_{j=0}^n \frac{1}{j!} \frac{\partial^{(j)} \check{W}_{1a}}{\partial \bar{I}_4^{(j)}}(1, 1) \sum_{k=0}^j \binom{j}{k} (-1)^k \langle \mathbf{C}^{\otimes(j-k)} | \mathbb{H}_{j-k} \rangle. \quad (93)$$

786 Consequently,  $\mathbf{S}_{1a}^{\text{app}}$  reads

$$\mathbf{S}_{1a}^{\text{app}} = 2\Phi_1 \sum_{j=1}^n \frac{1}{j!} \frac{\partial^{(j)} \check{W}_{1a}}{\partial \bar{I}_4^{(j)}}(1, 1) \sum_{k=0}^{j-1} \binom{j}{k} (-1)^k \frac{\partial}{\partial \mathbf{C}} \langle \mathbf{C}^{\otimes(j-k)} | \mathbb{H}_{j-k} \rangle, \quad (94)$$

787 and one has to compute the derivative

$$\mathbf{T} : \left( \frac{\partial}{\partial \mathbf{C}} \langle \mathbf{C}^{\otimes \ell} | \mathbb{H}_\ell \rangle \right) = \ell \langle \mathbb{H}_\ell | \mathbf{T} \otimes \mathbf{C}^{\otimes(\ell-1)} \rangle, \quad \ell \in \mathbb{N}, \ell \geq 1, n \geq 1, \quad (95)$$

788 where  $\mathbf{T}$  is an arbitrary “covariant” second-order tensor. This result can be proven by invoking the  
 789 fact that, at least in the case of transverse isotropy,  $\mathbb{H}_\ell$  is fully symmetric for every  $\ell$ , and noticing  
 790 that (we show the explicit index calculation only for  $\ell = 1, 2$ ):

$$\frac{\partial}{\partial C_{RS}} \langle \mathbf{C}^{\otimes 1} | \mathbb{H}_1 \rangle = \frac{\partial}{\partial C_{RS}} (C_{MN}(\mathbb{H}_1)^{MN}) = (\mathbb{I}^T)_{MN}{}^{RS} (\mathbb{H}_1)^{MN} = (\mathbb{H}_1)^{RS}, \quad (96)$$

$$\begin{aligned} \frac{\partial}{\partial C_{RS}} \langle \mathbf{C}^{\otimes 2} | \mathbb{H}_2 \rangle &= \frac{\partial}{\partial C_{RS}} (C_{MN} C_{PQ}(\mathbb{H}_2)^{MNPQ}) \\ &= (\mathbb{I}^T)_{MN}{}^{RS} C_{PQ}(\mathbb{H}_2)^{MNPQ} + C_{MN} (\mathbb{I}^T)_{PQ}{}^{RS} (\mathbb{H}_2)^{MNPQ} \\ &= 2(\mathbb{H}_2)^{RSAB} C_{AB}. \end{aligned} \quad (97)$$

791 Therefore, we obtain (again with the help of an arbitrary “covariant” second-order tensor  $\mathbf{T}$ )

$$\mathbf{T} : \mathbf{S}_{1a}^{\text{app}} = 2\Phi_1 \sum_{j=1}^n \frac{1}{j!} \frac{\partial^{(j)} \check{W}_{1a}}{\partial \bar{I}_4^{(j)}}(1, 1) \sum_{k=0}^{j-1} \binom{j}{k} (-1)^k (j-k) \langle \mathbb{H}_{j-k} | \mathbf{T} \otimes \mathbf{C}^{\otimes(j-k-1)} \rangle. \quad (98)$$

792 For the PARG method, we write  $\hat{W}_{1a}(\mathbf{C}, \mathbf{A}) = \mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A}))$ , where  $\mathfrak{f}$  is any differentiable function of  
 793 its argument, and  $\mathcal{P}(\mathbf{C}, \mathbf{A})$  is a tensor-power polynomial. Then, we enforce the approximation

$$\int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \hat{W}_{1a}(\mathbf{C}, \mathbf{A}) = \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathfrak{f}(\mathcal{P}(\mathbf{C}, \mathbf{A})) \simeq \mathfrak{f} \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right). \quad (99)$$

794 and  $\mathbf{S}_{1a}^{\text{app}}$  becomes

$$\mathbf{S}_{1a}^{\text{app}} = 2\Phi_1 \mathfrak{f}' \left( \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right) \left( \frac{\partial}{\partial \mathbf{C}} \int_{\mathbb{S}^2\mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right). \quad (100)$$

795 In the specific case in which

$$f(\mathcal{P}(\mathbf{C}, \mathbf{A})) = \frac{1}{2}c_{1a} [\exp(\mathcal{P}(\mathbf{C}, \mathbf{A})) - 1] \text{ and } \mathcal{P}(\mathbf{C}, \mathbf{A}) = (\langle \mathbf{C} | \mathbf{A} \rangle - 1)^2,$$

796 so that

$$\int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) = \langle \mathbf{C}^{\otimes 2} | \mathbb{H}_2 \rangle - 2\langle \mathbf{C} | \mathbb{H}_1 \rangle + 1, \quad (101)$$

797 we obtain

$$\begin{aligned} \mathbf{S}_{1a}^{\text{app}} &= \Phi_1 c_{1a} \exp \left( \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right) \left( \frac{\partial}{\partial \mathbf{C}} \int_{\mathbb{S}^2 \mathcal{B}} \Psi(\mathbf{M}) \mathcal{P}(\mathbf{C}, \mathbf{A}) \right) \\ &= \Phi_1 c_{1a} \exp \left( \langle \mathbf{C}^{\otimes 2} | \mathbb{H}_2 \rangle - 2\langle \mathbf{C} | \mathbb{H}_1 \rangle + 1 \right) (2\mathbb{H}_2 : \mathbf{C} - 2\mathbb{H}_1). \end{aligned} \quad (102)$$

798 Finally, for the STEx method, if  $\mathbf{T}$  is an arbitrary ‘‘covariant’’ second-order tensor, we have

$$\begin{aligned} \mathbf{T} : \mathbf{S}_{1a}^{\text{app}} &= \mathbf{T} : 2\Phi_1 \frac{\partial \hat{W}_{1a}}{\partial \mathbf{C}}(\mathbf{C}, \mathbf{A}) \\ &+ 2\Phi_1 \sum_{j=1}^n \left\langle \frac{1}{j!} \frac{\partial^{(j+1)} \hat{W}_{1a}}{\partial \mathbf{A}^{(j)} \partial \mathbf{C}}(\mathbf{C}, \mathbf{A}_0) \left| \mathbf{T} \otimes \sum_{k=0}^j \left[ (-1)^k \binom{j}{k} \text{msym}(\mathbb{H}_{j-k} \otimes \mathbf{A}_0^{\otimes k}) \right] \right. \right\rangle. \end{aligned} \quad (103)$$

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