A comparison of the Hart-Smith model with the Arruda-Boyce and Gent formulations for rubber elasticity

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Equivalence of the Hart-Smith model with Arruda-Boyce and Gent formulations for rubber elasticity

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Abstract

The present paper demonstrates the equivalence between the Hart-Smith constitutive model with the more recent Arruda and Boyce eight chains and Gent constitutive models. The ability of these three models to predict both small and large strain responses of rubbers is highlighted and equations that relate their material parameters are established.

Key words: Rubber material, Finite strain, Constitutive behaviour.

Introduction

Elastomers exhibit a very complex mechanical behavior, that includes large strain, time-dependent response, hysteresis and accommodation (Mullins effect). In the last few years, there was a significant effort to propose realistic constitutive equations for these materials [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. These models focus on the whole behavior of elastomers or on only one of the previously mentioned phenomena. Nevertheless, all constitutive equations are based on the general theory of hyperelasticity [11]. So, consider now the non-linear elastic response of rubber like material. It is characterized by the existence of a strain energy potential W. Assuming that elastomers are both isotropic and incompressible, this strain energy function only depends on the two first invariants of the left Cauchy-Green stretch tensor $\mathbf{B}$:

$$W = W(I_1, I_2) \tag{1}$$

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with:

$$I_1 = tr(\mathbf{B}) \qquad and \qquad I_2 = \left[I_1^2 - tr(\mathbf{B^2})\right] \tag{2}$$

The true stress tensor is defined by the differentiation of W with respect to $\mathbf{B}$:

$$\sigma = -p\mathbf{I} + 2\mathbf{B}\frac{\partial W}{\partial \mathbf{B}} = -p\mathbf{I} + 2\left(\frac{\partial W}{\partial I_1} + I_1\frac{\partial W}{\partial I_2}\right)\mathbf{B} - 2\frac{\partial W}{\partial I_2}\mathbf{B}^2 \tag{3}$$

This general theory being established, the major difficulty remains the choice of the function W. Many developments were proposed in the bibliography (see for example the review of Boyce [12]). A "good" hyperelastic model can be defined by four major qualities:

- first, it should be able to accurately reproduce the whole "S" shaped response of rubbers,
- second, the change of deformation modes should not be problematic, i.e. if the model behaves satisfactorily in uniaxial tension, it should also be quite accurate in simple shear or in equi-biaxial extension,
- third, the number of relevant material parameters must be as small as possible, in order to reduce the number of experimental tests needed for their identification,
- finally, the mathematical formulation has to be quite simple to render possible the numerical implementation of the model.

Taking into account these four pre-requisites, three constitutive equations can be selected: the recent models of Arruda and Boyce [13], and Gent [14], and the "older" formulation of Hart-Smith [15]. Through the rest of the paper, they will be referred to as AB, G and HS theories, respectively.

The aim of the present work is to demonstrate the equivalence of these three constitutive equations. More precisely, their small and large strain characteristics will be thoroughly examined. As Boyce [16] recently compared the AB and G approaches, the emphasize is laid on the equivalence of the HS model with the two other ones. In the next Section, the three strain energy functions are briefly recalled. Then, the theoretical relationships between material parameters of the models are established in Section 2. The relevance of these results is demonstrated in Section 3. Finally, some concluding remarks are proposed.

1 Strain energy functions

In this section, the strain energy functions that define the HS, AB and G models are briefly recalled.

1.1 The HS model

In the 60's, Hart-Smith [15] proposed an empirical form of W, that satisfactorily reproduces the response of rubberlike materials for the entire range of deformation endured by rubbers:

$$W_{HS} = C_1 \int \exp[C_3(I_1 - 3)^2] dI_1 + C_2 \ln\left(\frac{I_2}{3}\right) \tag{4}$$

where C_1, C_2 and C_3 are the three material parameters. In this equation, the first right-hand side part of W describes the global response of the material. It only depends on the first strain invariant I_1. The second term that involves I_2 is based on the work of Gent and Thomas [17] and improves the accuracy of the model for moderate strain (less than 150%). It can be related to the Mooney-Rivlin theory that involves both I_1 and I_2 and is widely recognized for its ability to satisfactorily reproduce rubber like behavior for small and moderate strain [11].

1.2 The AB model

The model of Arruda and Boyce [13] is a molecular based constitutive equation, also known as the 8-chains model. Following Kuhn and Grün [18], the behavior of an individual chain is considered non-gaussian. In order to describe the response of the polymer network, this individual response should be integrated in an unit sphere defined by its chain density. Nevertheless, the corresponding constitutive equation necessitates numerical integration [19]. So, in order to overcome this difficulty, privileged directions for chains orientation are defined by the half diagonals of a unit cube contained in the unit sphere. The corresponding strain energy function is given by:

$$W_{AB} = C_R N \left\{ \sqrt{\frac{I_1}{3N}}\beta + \ln\left(\frac{\beta}{\sinh\beta}\right) \right\} \tag{5}$$

where $\beta = \mathcal{L}^{-1}\left(\sqrt{I_1/3N}\right)$ with $\mathcal{L}$ being the Langevin function defined as $\mathcal{L}(x) = \coth(x) - 1/x$. In the previous equation, C_R and N are the two material parameters. Due to the non-gaussian nature of the chain modelling, $3N$ stands for the maximum value that I_1 can reach and represents the extensibility limit of chains.

This model only depends on the first strain invariant I_1 , and it is well-known that its performances at moderate strain are not very good [19]. Very recently, Meissner and Matejka [20] proposed the improvement of chains models by

adding a function of I_2 to the strain energy. Moreover, Fried [21] explained the dependance of W on I_2 invoking physical considerations.

1.3 The G model

Recently, Gent [14] proposed an empirical relation for the strain energy function:

$$W_G = -\frac{E}{6}\ln\left(1 - \frac{I_1 - 3}{J_m}\right) \tag{6}$$

where E and J_m are the two material parameters. J_m represents the maximum value of $I_1 - 3$ that can be undergone by the material. It is the empirical counterpart of the extensibility limit of chains previously defined for the AB model.

2 Comparison of materials parameters

The present section is devoted to the comparison of the three models. As these constitutive equations are supposed to be qualitatively efficient for the entire range of strains, both small and large strain responses will be compared. As mentioned above, the I_2 terms in constitutive equations can be seen as corrections of the phantom network theory. So, we propose to only compare the part of functions W that involves I_1 , i.e. the phantom part of constitutive equations. Consequently, the following HS strain energy will be considered through the rest of the paper:

$$W_{HS} = C_1 \int \exp[C_3(I_1 - 3)^2]dI_1 \tag{7}$$

in which only two material parameters are considered, i.e. C_1 and C_3.

Moreover, recalling that a previous work demonstrated the equivalence of the AB and the G approaches [16], the present work emphasizes the comparison of the HS model with them.

2.1 Polynomial expansion

Before explicitly comparing the models, their polynomial expansion in terms of I_1 are established. This will highly simplified the comparison of their small strain responses. The HS strain energy function (7) can be developed into an

infinite series of powers of I_1:

$$W_{HS} = C_1 \sum_{i=0}^{\infty} \frac{C_3^i}{(2i+1)i!}(I_1 - 3)^{2i+1} \tag{8}$$

It is to note that this expression is a particular case of the general expansion proposed by Rivlin [22]:

$$W = \sum_{(r,s)=(0,0)}^{\infty} C_{rs}(I_1 - 3)^r (I_2 - 3)^s \tag{9}$$

where the dependance on I_2 is not considered, said $j = 0$, and only odd powers of I_1 are relevant. The five first terms of the development of W_{AB} , given by Eq. (5), into a series of powers of I_1 are:

$$W_{AB} = C_R \left[\frac{1}{2}(I_1 - 3) + \frac{1}{20N}(I_1^2 - 9) + \frac{1}{1050N^2}(I_1^3 - 27) + \frac{19}{7000N^3}(I_1^4 - 81) + \frac{519}{673750N^4}(I_1^5 - 243) + ...\right] \tag{10}$$

This expansion is also a particular case of the Rivlin's series (9), but, in comparison with Eq. (8), it is expressed with both odd and even powers of I_1. Similarly to previous expansions,W_G (see Eq. (6)) can be expanded into a series of powers of I_1:

$$W = \frac{E}{6} \sum_{n=1}^{\infty} \frac{1}{J_m^{n+1}}(I_1 - 3)^n \tag{11}$$

Here, both odd and even powers of I_1 are involved.

2.2 Small strain behavior

The small strain stiffness of the models are respectively defined by first terms of Eqs (8), (10) and (11). Then, the three strain energy functions reduce to the neo-Hookean expression [11]:

$$W_{NH} = \frac{1}{2}C(I_1 - 3) \tag{12}$$

where C is a material parameter. Moreover, the first parameters of the HS, AB and G models are proportional. They are simply related to C by the following equation:

$$C = 2C_1 = C_R = \frac{E}{3} \tag{13}$$

As mentioned above, the three models are characterized by their ability to describe the strain-hardening of the material that takes place under large strain. This strain-hardening phenomenon is mainly due to the extensibility limit of polymer chains. According to Eq. (3) and taking into account that the considered strain energies do not depend on I_2, i.e. $\partial W/\partial I_2 = 0$, the stress-strain relationship reduces to:

$$\sigma = -p\mathbf{I} + 2\frac{\partial W}{\partial I_1}\mathbf{B} \tag{14}$$

in which only the first derivative of W with respect to I_1 is involved. In the three cases, this derivative is given by:

$$\frac{\partial W_{HS}}{\partial I_1} = C_1 \exp\left[C_3(I_1-3)^2\right] \tag{15}$$

$$\frac{\partial W_{AB}}{\partial I_1} = \frac{C_R N}{2\sqrt{3I_1}}\mathcal{L}^{-1}\left(\sqrt{\frac{I_1}{3N}}\right) \tag{16}$$

$$\frac{\partial W_G}{\partial I_1} = \frac{E}{6}\,\frac{1}{1-\frac{(I_1-3)}{J_m}} \tag{17}$$

Examining these equations, it is obvious that the three functions exhibit an upturn for large values of I_1. Moreover, for both AB and G models, $\partial W/\partial I_1$ tends asymptotically to infinity as the first invariant approaches a limiting value. Vertical asymptotes correspond with values of I_1 equal to $3N$ and J_m+3 for AB and G constitutive equations, respectively. Thus, the second material parameters of AB and G potentials are obviously related by:

$$J_m = 3(N-1) \tag{18}$$

Consider now the HS model. It does not exhibit an asymptotic behaviour, because it involves an exponential-like function. Nevertheless, in order to ensure that models reproduce the same behavior at large strain, it is important that they admit similar slopes in this range of deformation. Here, the HS model is only compared with the G model, the equivalence of AB and G models being already established in Eq. (18). Slopes of stress-strain curves can be expressed in terms of the second derivatives of W with respect to I_1 that are:

$$\frac{\partial^2 W_{HS}}{\partial I_1^2} = 2C_1C_3(I_1-3)\exp\left[C_3(I_1-3)^2\right] \tag{19}$$

$$\frac{\partial^2 W_G}{\partial I_1^2} = \frac{E}{6J_m}\,\frac{1}{\left[1-\frac{(I_1-3)}{J_m}\right]^2} \tag{20}$$

Thus, to ensure that the responses of HS and G models are similar, these two derivatives should be equal. Eliminating their first material parameters, i.e. C_1 and E respectively, by the use of Eq. (13), it yields to:

$$2C_3(I_1-3)\exp\left[C_3(I_1-3)^2\right] = \frac{1}{J_m}\,\frac{1}{\left[1-\frac{(I_1-3)}{J_m}\right]^2} \tag{21}$$

which lies the second material parameters C_3 and J_m, and depends on I_1. This equation means that the equality of slopes depends on the considered deformation level. In order to solve this equation, it can be casted under the following form:

$$C_3(I_1-3)^2\exp\left[C_3(I_1-3)^2\right] = \frac{(I_1-3)}{2J_m}\,\frac{1}{\left[1-\frac{(I_1-3)}{J_m}\right]^2} \tag{22}$$

The solution of this equation can be expressed in terms of the *LambertW* function, denoted here W^L:

$$C_3(I_1-3)^2 = W^L\left(\frac{(I_1-3)}{2J_m}\,\frac{1}{\left[1-\frac{(I_1-3)}{J_m}\right]^2}\right) \tag{23}$$

where W^L is defined to be the function satisfying (see Corless et al. [23] and the references herein for details):

$$W^L(x)\exp\left[W^L(x)\right] = x \tag{24}$$

So, defining now the reduced first strain invariant by:

$$\alpha = \frac{I_1-3}{J_m} \tag{25}$$

where $\alpha \in [0,1]$ as J_m represents the asymptotic value of I_1-3, and considering some basic algebraic manipulations, Eq. (23) can be transformed into:

$$C_3J_m^2 = \frac{1}{\alpha^2}W^L\left(\frac{\alpha}{2(1-\alpha)^2}\right) \qquad \text{for} \qquad \alpha \in [0,1] \tag{26}$$

This equation relates the second material parameters C_3 and J_m that satisfy the equality of slopes at large strain for HS and G models. As mentioned above, it depends on α, i.e. on the strain level. Nevertheless, as the G model increases asymptotically to infinity in the neighborhood of $I_1 = J_m + 3$, i.e. $\alpha = 1$, and the HS model is an exponential-like function in this range of I_1, Eq. (26) is not defined for $\alpha = 1$. In practice, the equivalence between the two constitutive equations can only be demonstrated on a given range of α

(or of I_1). In this way, a mean constant value of the function in the right-hand side of Eq. (26) can be adopted to relate C_3 and J_m. This permits to omit the dependence on α of $C_3 J_m^2$. Let us denote $[\alpha_{\min}, 1[$ with $\alpha_{\min} > 0$, the range in which the HS and G models are close to. Thus, in this range, the two parameters can be linked by a constant coefficient:

$$C_3 J_m^2 = k \qquad \text{with} \qquad k = \frac{1}{1-\alpha_{\min}} \int_{\alpha_{\min}}^{1} \frac{1}{\alpha^2} W^L \left(\frac{\alpha}{2(1-\alpha)^2} \right) d\alpha \quad (27)$$

So, the value of k depends on the range in which the equivalence of the two models is recognized. Figure 1 presents the evolution of k as a function of $\alpha_{\min}$. As shown by the graph, k tends to infinity as $\alpha_{\min}$ tends to 0 or 1. The practical choice of the bound $\alpha_{\min}$ and the corresponding value k will be examined in the next section for two examples.

Fig. 1. Curve of Eq.(27): k vs $\alpha_{\min}$

3 Examples

Finally, two examples are studied to demonstrate the equivalence of the three constitutive equations, and the relevance of Eqs (13), (18) and (27) that lie the material parameters.

3.1 Fit of the G theory

The first example consists in using the G model as a reference. In order to build fictitious materials and their corresponding data sets to be fitted, the

two parameters E and J_m are considered to vary in $]0, 10]$ and $[7, 100]$ respectively. Values of J_m stand for limits of extensibility between 200% and 900% in uniaxial tension.

First, J_m is set to 60 and E adopts the three values 0.1, 1 and 10. In these three cases, the function $\partial W_G/\partial I_1$ given by Eq. (17) is discretized using a large number of values, and the corresponding stress functions of HS and AB models, respectively given by Eqs (15) and (16), are fitted with these data. Fitting results are obtained using two different algorithms. First, an optimization program based on a genetic algorithm estimates approximately the material parameters. Second, these parameters are precisely determined with the help of a classical steepest descent algorithm using the genetic algorithm results as initial guess solutions. The linear relation eq. (13) between first parameters of the three models is exactly recovered for the three tests.

Second, the initial stiffness E is set to 1, and J_m varies between 7 and 100. First parameters of HS and AB models are calculated according to Eq. (13), said $C_1 = 6$ and $C_R = 3$. For the considered values of J_m, the method employed above for first parameters is used to determine both C_3 and N. Let us first examine the results for the AB model. Figure 2 shows the comparison between fitted results and the theoretical relation Eq. (18). The fitted results agree well with the theory and the linear relation between the asymptotic limit of the two models is satisfactorily recovered.

Fig. 2. Comparison of second parameters for the G and AB models: (∘) fitted results, (—) theoretical results of Eq. (18)

As mentioned above, the case of the HS model is more difficult. Our goal is to verify Eq. (27). Thus, the interval of α should be defined to calculate the integral in this equation. In order to choose the lower bound $\alpha_{\min}$, we consider that slopes of HS and G models should not be compared for small strain, because

only the large strain responses of these models are examined. As proposed by Kucherskii [24], the neo-Hookean formulation is valid in the first third of the deformation range. Thus, only the two thirds of the deformation range has to be retained for the large strain response, i.e. $\alpha_{\min} \geq 1/9$. Moreover, as models behaviors are highly different in the neighborhood of $\alpha = 1$, because the G model tends asymptotically to infinity and the HS model grows exponentially, we consider that the equivalence should be demonstrated for more than a third of the deformation range, said $\alpha_{\min} \leq 4/9$. Consequently, according to Eq. (27) and noting that k does not evolve monotonously in $\alpha_{\min} \in [1/9, 4/9]$ (see Figure 1), the range in which k varies can be easily established:

$$2.83 \leq k \leq 2.98 \tag{28}$$

Finally, Figure 3 presents the comparison between fitted and theoretical results. The two boundary values of k given in Eq. (28) are considered. This figure shows that fitted results are satisfactorily reproduced by Eq. (27), with a slight dependance on the value of $\alpha_{\min}$.

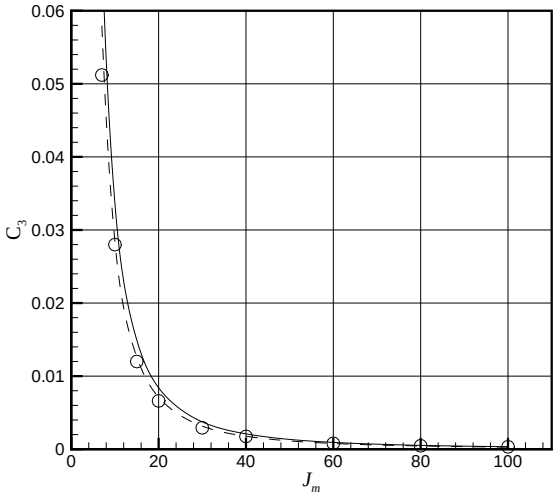

Fig. 3. Comparison of second parameters for G and HS models: (∘) fitted results, (- - -) theoretical results of Eq. (27) for $k = 2.83$, (—) theoretical results of Eq. (27) for $k = 2.98$.

3.2 Fit of Treloar's data

The second example focuses on the comparison of models for a real material. Here, experimental data of Treloar [25] are considered. The author performed a large number of experiments in different deformation modes: uniaxial tension,

pure shear, equibiaxial tension (by the use of the bubble inflation technique), and biaxial tension (by combining tension and pure shear). The corresponding stress-strain responses were thoroughly examined in the past and are not recalled here. The reader can refer to Ogden [26] for details.

Using these data, material parameters of the three constitutive equations are determined. The identification was performed using the four tests simultaneously. Fitted results for material parameters are presented in Table 1. The corresponding results for the stress-strain responses are shown Figure 4. In this figure, X-axis and Y-axis of each graph correspond to relevant stretch and nominal stress of the type of experiment. The four graphs exhibit the equivalence between the three models and demonstrate their ability to satisfactorily reproduce different deformation modes with only two material parameters.

	First parameter, MPa	Second parameter
HS model	$C_1 = 0.18$	$C_3 = 2.7\ 10^{-4}$
AB model	$C_R = 0.34$	$N = 27.9$
G model	$E = 1.0$	$J_m = 92.0$

Table 1 : Values of material parameters

Let us now examine the relevance of the previously demonstrated relationships as applied to the parameters presented in Table 1. In order to simplify the discussion, G material parameters are considered as the reference, and material parameters of AB and HS theories are compared with them.

- Concerning the first material parameter of models, we first compute $6C_1 = 1.08$ for the HS model and $3C_R = 1.02$ for the AB theory as proposed in Eq. (13). Thus, the maximum difference between these data and $E = 1$ is about 8%.
- Second, the large strain parameters J_m and N of G and AB models are analysed. According to Eq. (18), the theoretical value of N should be $(J_m + 1)/3 = 31.0$. The difference between this predicted value and the fitted result is about 10%.
- Finally, we consider the relationship between J_m and C_3. The corresponding theoretical relation that relies these data is given by Eq. (27). In order to compute k, the lower bound $\alpha_{\min}$ of the definition range should be determined. Examining experimental results for the uniaxial test shown in Figure 4(a), the small strain range, in which the neo-Hookean theory is sufficient, can be defined as $\lambda \in [1, 4]$. This interval corresponds to $I_1 \in [3, 16.5]$ and the use of Eq. (25) yields to $\alpha_{\min} \approx 0.14$. In regards with Eq. (27), k is defined by an integral from $\alpha_{\min}$ to 1. Nevertheless, considering that the last experimental point of the uniaxial tensile test corresponds with $\lambda_{\max} = 7.6$, i.e. $I_1 = 58$, the upper bound of the integration range is not equal to 1 but

Fig. 4. Fit of Treloar's data, (a) unixial tension, (b) pure shear, (c) equibiaxial tension, (d) biaxial tension: (∘) experiments, (- - -) HS model, (—) AB model, (. . .) G model.

should be replaced by $\alpha_{\max} = (I_{1\max} - 3)/J_m \approx 0.6$. In fact, it is impossible to experimentally reach the extensibility limit of chains, because samples break down for smaller stretches. Finally, the theoretical value of k to be considered is given by:

$$k = \frac{1}{10.6 - 0.14} \int_{0.14}^{0.6} \frac{1}{\alpha^2} W^L \left(\frac{\alpha}{2(1-\alpha)^2} \right) \, d\alpha \approx 2.69 \tag{29}$$

It is to note that this value is not contained in the range of Eq. (28). Using this result in Eq (3), the second parameter of the HS model C_3 should have been equal to $3.18\ 10^{-4}$. Comparing this value with the fitted result given in Table 1 leads to a discrepancy of 17.7%. In regards with differences obtained between our theoretically predicted values and fitted results, the present developments are validated. The equivalence equations (13), (18) and (27) can now be used to switch between HS, AB and G hyperelastic constitutive equations.

4 Conclusion

The present work demonstrates the equivalence between three hyperelastic constitutive equations: the HS, AB and G models. Here, only the I_1 part of the models were compared. As proposed elsewhere [20], the accuracy of the three approaches for moderate strain can be improved by adding a function of I_2 to W. This can be performed with only one additional material parameter. Note that the original Hart-Smith model included this I_2 term. The HS model presents some advantages that will highly simplify the numerical implementation: it is formulated in terms of stretch invariants and its large strain behavior is governed by an exponential-like function, so that its tangent stiffness always exists. Moreover, the equivalence demonstrated here will also render easier the identification of material parameters, because HS parameters can be now related to physical concepts through their equivalence with AB data.

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