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Research Paper

A Theory of Undamageable Graphene

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It is the aim of this work to develop and extend the theory of undamageable materials to graphene. An undamageable material is a material where the value of the damage variable remains zero throughout the deformation process. It is anticipated that the constitutive equations for undamageable graphene can be modeled with differential equations for the case of graphene. The equations are solved for three cases: $n = 1$, $n = 2$, and the general case of n. It is hoped that undamageable graphene can be achieved in the laboratory in the near future when the manufacturing technology advances so as to produce such materials.

Key words: undamageable material; graphene; strain energy equivalence; continuum damage mechanics; elastic stiffness degradation.

1. Introduction

The theory of undamageable materials has been developed recently by Voyiadjis and Kattan [40, 41, 43, 45]. This theory has been developed within the framework of continuum damage mechanics. Continuum damage mechanics has advanced rapidly since the early work of Kachanov [13]. In particular, advances in damage has been introduced by many authors with important insights and developments in continuum damage (Kachanov [13], Kattan and Voyiadjis [15–17], Krajcinovic [18], Lee *et al.* [22], Sidoroff [31], Voyiadjis and Kattan [33, 35–37]). Further research along this topic was undertaken later by Kattan and Voyiadjis [14], Ladeveze and Lemaitre [19], Voyiadjis *et al.* [47], Voyiadjis and Kattan [38, 39, 42, 44].

Further developments in damage mechanics appeared rapidly in the past few years (Celentano *et al.* [5], Doghri [9], Hansen and Schreyer [10], Kattan and Voyiadjis [14, 15], Ladeveze *et al.* [20], Lee *et al.* [22], Lubineau [27],

Lubineau and Ladeveze [28], Luccioni and Oller [29], Rice [30], Voyiadjis [32], Voyiadjis and Kattan [34, 35]). Basaran and Yan [1] and Basaran and Nie [2] used an alternate approach to damage characterization based on an entropy generation rate as a damage metric rather than the damage potential surface. Practical examples of elastic stiffness degradation in highly jointed rock masses are given by Cai and Horii [3].

Research continued in damage mechanics by several researchers who applied the concept to other topics: damage and elasto-plastic models (Chaboche [6], Kattan and Voyiadjis [14, 15], Ladeveze and Lemaitre [19], Ladeveze *et al.* [20], Lee *et al.* [22], Voyiadjis [32], Voyiadjis and Kattan [34, 35], Voyiadjis *et al.* [48]), damage and elasto-viscoplastic models (Chaboche [7], Lemaitre and Chaboche [25]), damage and continuum damage models (Voyiadjis and Kattan [33]), and coupled elasto-plastic damage models (Chow and Jie [8], Lemaitre [23, 24]). The damage variable or damage tensor represents the various types of damages at the micro-scale level such as micro-cracks, micro-voids, and micro-cavities (Lubarda and Krajcinovic [26], Voyiadjis and Kattan [33]). Some researchers postulate the use of two independent damage variables to accurately describe the case of isotropic damage (Cauvin and Testa [4], Ju and Chen [11, 12]).

Within the framework of continuum damage mechanics, the concept of the Representative Volume Element (RVE) is used where discontinuities such as micro-cracks and micro-voids are not considered at all. Thus macroscopic internal variables are used to lump the effect of all damages. As a result of this concept, a thermodynamically consistent formulation is obtained (Hansen and Schreyer [10], Kattan and Voyiadjis [16, 17], Voyiadjis *et al.* [46]).

In an undamageable material the value of the damage variable remains zero throughout the deformation and damage process. In this way the material cannot be damaged under any loading condition whatsoever. Thus it is given the name undamageable material. It has been proved by Voyiadjis and Kattan [40, 41, 43, 45] that the value of the damage variable remains zero during the loading stage.

The constitutive equations for undamageable materials have been derived mathematically based on differential equations. There is a parameter n that increases from 1 for linear elastic materials to infinity for undamageable materials. The value of the damage variable is zero when n equals infinity (Voyiadjis and Kattan [40, 41, 43, 45]).

It is the aim of this work to extend the theory of elastic undamageable materials to graphene. It is noted that graphene is a very important material used in the industry today. Developing the theory of undamageable graphene is possible using differential operators. The differential equations have been solved for three cases: $n = 1$, $n = 2$, and the general case of n.

Undamageable graphene is a hypothetical material that does not exist today but may lead the scientists to certain material parameters that are needed to capture this behavior. It is hoped that the manufacturing technology will advance in the near future so as to produce such materials. Once undamageable materials are produced, it will be easy to follow this up with the production of undamageable graphene.

The current work lays the foundation for the theory of elastic undamageable graphene. Since the constitutive equations for undamageable materials resemble those of rubber and biological tissue, it is anticipated that undamageable materials, and in particular undamageable graphene, will have rubber-like or biological-like components.

2. What is meant by an undamageable material?

Some elaboration is given on the new concept of undamageable materials. These hypothetical materials were proposed recently by Voyiadjis and Kattan [40, 41, 43, 45]. These types of material compare with rubber materials and biological tissue. The authors also made comparisons between undamageable materials and various nonlinear elastic materials.

Undamageable materials are proposed and designed in such a way so as to maintain a zero value for the damage variable throughout the deformation process. This formulation was presented by Voyiadjis and Kattan [40, 41, 43, 45] in great detail within the concept of continuum damage mechanics. Thus, it can be seen that undamageable materials are desirable since they cannot be damaged at all. It is hoped that the manufacturing technology will reach a stage in the future where the realization of this type of material can be achieved.

To show that such materials maintain a zero value of the damage variable (when $n \to \infty$) throughout the deformation process, we need to modify the classical definition of the effective stress to become as follows:

$$\overline{\sigma} = \frac{\sigma}{\sqrt[n]{1-\varphi}}, \tag{2.1}$$

where φ is the damage variable taking values between 0 and 1, and n is the same exponent used in the expressions of the elastic strain energy below. We can now perform the following trick when n approaches infinity:

$$\overline{\sigma} = \frac{\sigma}{\sqrt[n]{1-\varphi}} = \frac{\sigma}{(1-\varphi)^{1/n}} = \frac{\sigma}{(1-\varphi)^{1/\infty}} = \frac{\sigma}{(1-\varphi)^{0}} = \frac{\sigma}{1} = \sigma. \tag{2.2}$$

Thus, we obtain the undamageable material in this case. In their previous publications detail, Voyiadjis and Kattan [40, 41, 43, 45] presented the concept of undamageable materials using the definition of the damage variable in

terms of elastic stiffness degradation. We have now supported that formulation by presenting the concept of undamageable materials using a slightly modified form of the effective stress basesd on the cross-sectional area reduction as shown in Eqs. (2.1) and (2.2). It should be noted that Eq. (2.1) is the road that leads to Eq. (2.2) and undamageable materials.

The authors need to clarify the distinction between Eqs. (2.1) and (2.2). It is Eq. (2.2) that describes undamageable materials. It is the case when n approaches infinity when one obtains the undamageable material. Again Eq. (2.1) is the road that leads to Eq. (2.2) and undamageable materials. Furthermore the modification that was made to the equation of effective stress to obtain Eq. (2.1) is not arbitrary. There is a formal and consistent derivation of Eq. (2.1) that appears in the authors' previous work [42]. According to the proof one starts with the expression of the higher order elastic strain energy from and proceeds to derive Eq. (2.1). Equation (2.1) is not a choice but is the result of a formal derivation. The value of the effective stress remains unchanged irrespective of the value of the damage variable – this results from the infinity in Eq. (2.2) and applies only to Eq. (2.2). It does not apply to Eq. (2.1). It is hoped that this explanation will remove any misunderstandings between Eqs. (2.1) and (2.2).

3. Theoretical formulation

The elastic constitutive relation for graphene can be written in the following form:

$$\sigma = E\varepsilon + D\varepsilon^2, \tag{3.1}$$

where σ is the stress, ε is the strain, E is the elastic modulus (Young's modulus), and D is the third order elastic stiffness. Equation (3.1) has been verified experimentally by Lee *et al.* [21]. It should be noted that for graphene, while E has a positive value, D has a negative value.

The elastic strain energy U is obtained from the stress using the following relation:

$$U = \int \sigma \, \mathrm{d}\varepsilon. \tag{3.2}$$

Substituting for σ from Eq. (3.1) into Eq. (3.2), one obtains the following expression for the elastic strain energy:

$$U = \frac{1}{2}E\varepsilon^2 + \frac{1}{3}D\varepsilon^3. \tag{3.3}$$

Equation (3.3) for the elastic strain energy of graphene can be re-written as follows:

$$U = \frac{1}{2}\sigma^* \varepsilon, \tag{3.4}$$

where σ^* is given by:

$$\sigma^* = E\varepsilon + \frac{2}{3}D\varepsilon^2. \tag{3.5}$$

It should be noted that σ^* appears in instead of σ in the expression of U of Eq. (3.4). Consequently, the usual strain energy function $U = \frac{1}{2}\sigma\,\varepsilon$ still applies for graphene but is slightly changed, as shown in Eq. (3.2).

In order to develop the theory of undamageable graphene, one needs to use higher order energy expressions like $\frac{1}{2}\sigma\,\varepsilon^2$, $\frac{1}{2}\sigma\,\varepsilon^3$, ..., $\frac{1}{2}\sigma\,\varepsilon^n$, where n is a positive integer that goes to infinity. One now considers first the case when $n = 1$.

Case $n = 1$. Consider the following expression for the stress σ:

$$\sigma = Ef'(\varepsilon) + \alpha D f(\varepsilon), \tag{3.6}$$

where $f(\varepsilon)$ is an unknown function of the strain, and α is a constant. In the case of graphene, $f(\varepsilon) = \frac{1}{2}\varepsilon^2$ and $\alpha = 2$.

Starting with Eq. (3.6), one proceeds with the following long derivation. Substituting Eq. (3.6) for σ into Eq. (3.2) (this is valid for the case $n = 1$), one obtains the following expression for the elastic strain energy for this case:

$$\mathrm{d}U = \big[Ef'(\varepsilon) + \alpha D f(\varepsilon)\big]\,\mathrm{d}\varepsilon. \tag{3.7}$$

However, $U = \frac{1}{2}\sigma\,\varepsilon$, substituting into this expression Eq. (3.6) for σ, and differentiating the results, one obtains the following expression for $\mathrm{d}U$:

$$\mathrm{d}U = \frac{1}{2}\big[Ef'(\varepsilon) + \alpha D f(\varepsilon)\big]\,\mathrm{d}\varepsilon + \frac{1}{2}\big[Ef''(\varepsilon) + \alpha D f'(\varepsilon)\big]\varepsilon. \tag{3.8}$$

Solving Eqs. (3.7) and (3.8) for $\mathrm{d}U$ and re-arranging the terms, one obtains:

$$\frac{Ef''(\varepsilon) + \alpha D f'(\varepsilon)}{Ef'(\varepsilon) + \alpha D f(\varepsilon)} = \frac{\mathrm{d}\varepsilon}{\varepsilon}. \tag{3.9}$$

The above is a second-order differential equation in $f(\varepsilon)$. Subject to the initial condition that when $\varepsilon = 0$, $f(\varepsilon) = 0$, one solves the above equation by integrating both sides to obtain:

$$f(\varepsilon) = \frac{C\varepsilon}{\alpha D} - \frac{CE}{(\alpha D)^2}\Big(1 - \mathrm{e}^{-\alpha D\varepsilon/E}\Big), \tag{3.10}$$

where C is the integration constant. Considering the special case when ε is small, one uses the Taylor series expansion of the exponential function as follows:

$$\mathrm{e}^x = 1 + x + \frac{x^2}{2} + ... \tag{3.11}$$

Taking only the first two terms in the expansion, one applies this to the exponential function in Eq. (3.10) to obtain:

$$f(\varepsilon) \approx \frac{1}{2}\frac{\varepsilon^2}{E}. \tag{3.12}$$

It should be noted that the approximate expression obtained above is the correct expression for $f(\varepsilon)$ and resembles that of the elastic strain energy function for linear elastic materials. Next the case $n = 2$ is solved.

Case $n = 2$. In this section, the case when $n = 2$ is solved. Following the same procedure in the previous section, and starting with Eq. (3.6), one obtains the expression for $\mathrm{d}U$ as shown in Eq. (3.7). However, the change now is in the elastic strain energy function. One uses U in the form $U = \frac{1}{2}\sigma\,\varepsilon^2$. Substituting into this expression the formula for σ from Eq. (3.6) and differentiating the result, one obtains the following expression for $\mathrm{d}U$:

$$\mathrm{d}U = \frac{1}{2}\left[Ef'(\varepsilon) + \alpha D f(\varepsilon)\right]\varepsilon\,\mathrm{d}\varepsilon + \frac{1}{2}\left[Ef''(\varepsilon) + \alpha D f'(\varepsilon)\right]\varepsilon^2. \tag{3.13}$$

Equating Eqs. (3.7) and (3.13) for $\mathrm{d}U$ and rearranging the terms, one obtains the following differential equation:

$$\frac{Ef''(\varepsilon) + \alpha D f'(\varepsilon)}{Ef'(\varepsilon) + \alpha D f(\varepsilon)} = \frac{2(1-\varepsilon)}{\varepsilon^2}\,\mathrm{d}\varepsilon. \tag{3.14}$$

The above is a second-order differential equation in $f(\varepsilon)$. Using the MATLAB Symbolic Math Toolbox, one solves the above differential equation to obtain the following expression:

$$f(\varepsilon) = -\frac{\alpha D\varepsilon}{E}\int \frac{\mathrm{e}^{\alpha D\varepsilon/E}\,\mathrm{e}^{-2/\varepsilon}}{E\varepsilon^2}\,\mathrm{d}\varepsilon + C\mathrm{e}^{-\alpha D\varepsilon/E}, \tag{3.15}$$

where C is a constant of integration. Substituting the expression for $f(\varepsilon)$ from Eq. (3.15) into the expression for σ of Eq. (3.6), one can obtain the elastic constitutive equation for graphene when $n = 2$. Next the case for general n is solved.

General n. In this section, the case for the general value of the exponent n is solved. Starting again with Eq. (3.6) for the stress, and after differentiating it, one obtains the expression for $\mathrm{d}U$ in Eq. (3.7). However, the change now is in the elastic energy function. One uses a general expression for U as $U = \frac{1}{2}\sigma\varepsilon^n$.

Substituting into this expression the equation for the stress from Eq. (3.6), and differentiating the result, one obtains the following expression for $\mathrm{d}U$:

$$\mathrm{d}U = \frac{n}{2}\left[Ef'(\varepsilon) + \alpha D f(\varepsilon)\right]\varepsilon^{n-1}\,\mathrm{d}\varepsilon + \frac{1}{2}\left[Ef''(\varepsilon) + \alpha D f'(\varepsilon)\right]\varepsilon^{n}. \tag{3.16}$$

Equating Eqs. (3.7) and (3.16) and rearranging the terms, one obtains the following differential equation in $f(\varepsilon)$:

$$\frac{Ef''(\varepsilon) + \alpha D f'(\varepsilon)}{Ef'(\varepsilon) + \alpha D f(\varepsilon)} = \frac{2\left(1 - \frac{n}{2}\varepsilon^{n-1}\right)}{\varepsilon^{n}}\,\mathrm{d}\varepsilon. \tag{3.17}$$

The above is a second-order differential equation in $f(\varepsilon)$ for graphene for any general value of the exponent n. In order to solve the general differential equation of Eq. (3.17) one resorts to the MATLAB Symbolic Math Toolbox. The solution of the general differential equation is obtained as follows:

$$f(\varepsilon) = -\frac{\alpha D\varepsilon}{E}\int \frac{\mathrm{e}^{\frac{\alpha D\varepsilon}{E}}\,\mathrm{e}^{\frac{-2}{(n-1)\varepsilon^{n-1}}}}{E\varepsilon^{n}}\,\mathrm{d}\varepsilon + C\mathrm{e}^{\frac{-\alpha D\varepsilon}{E}}, \tag{3.18}$$

where C is a constant of integration. One then substitutes Eq. (3.18) into the expression for σ of Eq. (3.6) to obtain the general constitutive equation for graphene for any value of the exponent n. As n goes to infinity we obtain the hypothetical material called undamageable graphene.

4. Conclusion

In this work the equations of undamageable graphene are derived. The derivation follows the theory of undamageable materials presented in several papers by the authors previously. The differential equations for undamageable graphene are derived and solved for three cases. It should be noted that undamageable graphene is a hypothetical material currently and may be manufactured in the future once the technology is developed. The main property of undamageable graphene is that this material cannot be damaged under any type of loading.

The authors solve three cases. Indeed the two cases $n = 1$ and $n = 2$ will not lead to undamageable materials. But they are simple cases that can be solved. Only case 3 leads to undamageable materials when n approaches infinity. Furthermore, in the future the manufacturing technology might not be able to approach infinity or even undamageable materials. However, simple cases like $n = 2$, $n = 5$, and $n = 10$ might be realizable in the industry in the future. They will be on the road that leads to undamageable materials. The equations for $n = 5$ and $n = 10$ (or even $n = 100$) are obtained directly from Eq. (3.18) by substituting the correct value of n in the equation. There is no need to derive

these equations from scratch like what was done with the two cases $n = 1$ and $n = 2$. As n increases the reduction in the damaged stiffness decreases which emphasizes an understanding of the methodology to decrease in the percentage of stiffness reduction for the same damage input.

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