REVIEW OF COMMON PHENOMENA AND PROBLEMS OF PARTICULATE REINFORCED HYPER VISCOELASTIC RUBBER-LIKE COMPOSITE MATERIALS

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ABSTRACT

The main physico-mechanical properties and phenomena of particulate reinforced hyper viscoelastic composites are listed. The common difficulties and problems of mathematical modeling of such materials, which researchers face, are described. Classical approaches to theoretical research and numerical simulation are considered. Physical and geometrical nonlinearity, dewetting, effects of strain rate sensitivity, Mullins effect, temperature effects, dilatation, size dependence, superimposed hydrostatic pressure sensitivity and coupled bulk and deviatoric behavior are discussed in the framework of macroscopic point of view. Special attention is paid to the particulate reinforced composites damage initiation and propagation problems. It is considered two approaches based on introduction of dewetting surface similar to a plastic surface. The first approach uses plastic flow equations similar to the classical theory of plasticity and the second one deals with the hyper viscoelastic constitutive equations supposed to be dynamically changed i.e. shear and volumetric stiffness should be reduced during the dewetting process.

1. INTRODUCTION

Particulate reinforced hyper viscoelastic composites are widely used in space and defense industry. For example, all modern solid propellants use elastomeric rubber-like binder, which is filled by high percentage of solid particles. The behavior of particulate reinforced materials with rubber-like binder is complicated by the fact that in addition to the common problems of rubber-like materials such as physical and geometrical (finite strains) nonlinearity, effects of strain rate sensitivity (viscoelasticity), Mullins effect and temperature effects new phenomena are supplemented. The application of the load causes complex stress – strain state in the material binder, filler and interfaces between the particles and binding matrix (1). Interfacial debonding between the particles and the filler and the subsequent void formation, also called dewetting, is one of the main reason of the occurrence of a set of phenomena such as strong physical and geometrical nonlinearity, dilatation, superimposed hydrostatic pressure sensitivity and coupled bulk and deviatoric behavior.

The variety and complexity of behavior of investigated material make researchers combine the classical approaches with specially developed technics. Thus, in order to take into account strain rate effects in the hyperelastic materials, it was made an assumption that the instantaneous response of the material follows from the hyperelastic constitutive equations and the strain rate effect could be accounted by the introduction of the linear viscoelastic equations with only difference, that in the convolution equations hyperelastic relations take place instead of the linear elastic constitutive equations (2).

To develop the damage model the changes that occur during the dewetting process must be mathematically described. Dewetting has many similarities with yielding in the classical plasticity theory. Thus, the majority of the researchers define the dewetting surface (criterion) similar to the plastic surface in the 6 dimensional stress space. When for some point in the stress space the dewetting surface equation is satisfied, it becomes clear that in the corresponding point of the space there is a damage (dewetting) onset.

For the most viscoelastic materials, not only the available force for the damage initiation is rate-dependent, but also the resistance against the damage growth is rate-dependent (3). This means that the dewetting surface must be strain rate sensitive.

It is observed that the strength of particulate reinforced materials increases with superimposing of the pure hydrostatic pressure during the test. For example, the tensile strength in the uniaxial tensile test can be increased around two times with the supplementing of the hydrostatic pressure around 20 atmospheres into the test conditions. From the mathematical modeling point of view this means that the properties of the material are stress state sensitive and this fact has to be taken into account during the simulation i.e. the dewetting criteria must be stress state sensitive. From the physical point of view, this phenomenon can be explained by the fact that the main reason of dewetting is the formation of voids between the binder and the particles and the superimposed hydrostatic pressure helps “close” these voids, and dewetting (damage) starts much later upon this hydrostatic pressure and therefore, the maximum tensile strain and the nominal maximum stress increase.

In the rubber-like materials and particulate reinforced rubber-like materials so-called Mullins effect plays a very important role. This effect is the phenomenon of stress softening, commonly observed in filled rubber elastomers because of the damage associated with straining. When an elastomeric test specimen is subjected to simple tension from its virgin state, unloaded, and then reloaded, the stress required on reloading is less than that on the initial loading for stretches up to the maximum stretch achieved during the initial loading (4).

The effect of the temperature upon the mechanical behavior of the particulate reinforced composite materials produces a series of competing effects. As the temperature decreases the strength of the filler-to-polymer bond increases, the polymer strength increases. At the same time, the maximum nominal strain decreases with the increasing of the temperature.

Because of the nature of the localized dewetting in the particulate reinforced materials, it is found that most of the results of mechanical testing must be referred to the dimensions of the test specimen: length, width and thickness. An especially critical feature of this behavior is noted when considering alternate layers of locally yielded and non-yielded materials (5, 6).

The distortional response of particulate reinforced composite is well studied and it is not difficult to handle it using the classical methods of the hyperelasticity theory. However, the bulk response is further complicated by the fact that the total dilatation observed can be a relatively large positive quantity.

Summing up above-written it is possible to conclude that to simulate the particulate reinforced materials behavior it is necessary to create an approach, which will allow making predictions taking in account all phenomena in the framework of one universal material model i.e. the constitutive equations. To develop the correct constitutive equations it is necessary to make a lot of different type tests to obtain the information about physico-mechanical behavior of the material.

2. DEWETTING EFFECT

Any research on particulate reinforced materials does not do without studying the dewetting effect. The dewetting effect plays one of the main roles in the understanding of particulate reinforced materials damage nature. The effect of particle rotation on strain is to produce a repacking of the particles, which leads to the formation of voids within the medium, resulting from the failure, or dewetting of the bond between the binder and the filler. After an initial linear region, nonlinear stress-strain behavior under loading resulted from the progressive failure of the bonds at the interfaces between the fuel matrix and the oxidizer particles (7). Figures **1** and **2** show the typical nonlinear stress-strain behavior of the dewetted specimen during the uniaxial tensile test, which displays itself in softening of the material. Figure **1** (8) represents test results for deferent superimposed pressures (15, 50 and 165 psi) and Fig. **2** (9, 10) shows test results for different strain rates (0.0067%, 0.067%, 1.67% min^{-1}).

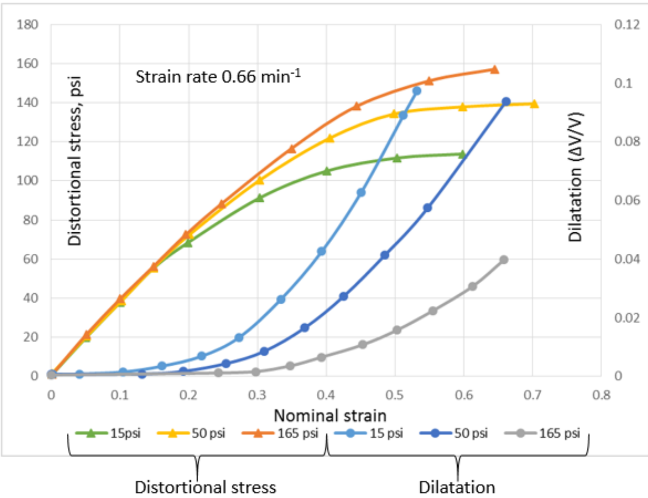

Fig. **1**. Nonlinear stress-strain behavior of particulate reinforced rubber-like material for different superimposed hydrostatic pressures (15, 50, 165 psi) during uniaxial tensile test.

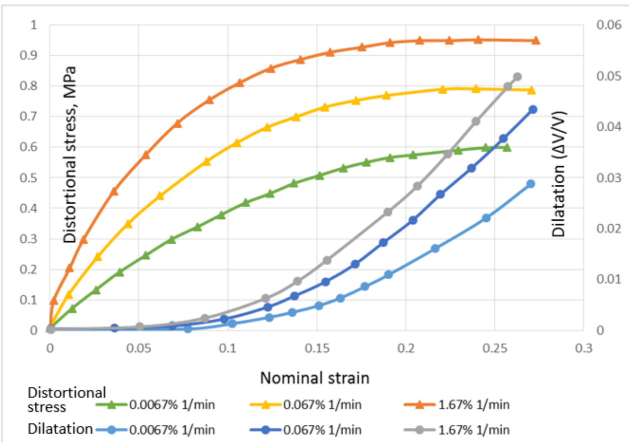

Fig. **2**. Nonlinear stress-strain behavior of particulate reinforced rubber-like material for different strain rates (0.0067%, 0.067%, 1.67% min^{-1}) during uniaxial tensile test.

3. DEWETTING SURFACE

As written before, the dewetting surface can be defined similar to the plastic surface in the general form $F(p,q)=0$, where $p=-\frac{1}{3}(\sigma_{11}+\sigma_{22}+\sigma_{33})$, $\mathbf{S}=\boldsymbol{\sigma}+p\mathbf{I}$, $q=\sqrt{\frac{3}{2}\mathbf{S}\cdot\cdot\mathbf{S}}$, σ_{ij} - stress tensor components. When for some point in the stress space the equation $F(p,q)=0$ is satisfied it becomes clear that in the corresponding point of space there is a damage (dewetting) onset. Further, there are two main approaches to simulate the dewetting process under loading.

The first approach is the usage of strain decomposition (11) $d\boldsymbol{\varepsilon}=d\boldsymbol{\varepsilon}^{e}+d\boldsymbol{\varepsilon}^{dew}$, where the total strain $d\boldsymbol{\varepsilon}$ is the sum of the elastic strain $d\boldsymbol{\varepsilon}^{e}$ and the dewetting strain $d\boldsymbol{\varepsilon}^{dew}$. In this case during the positive loading it is assumed that the dewetting strains are normal to the dewetting surface i.e. $d\boldsymbol{\varepsilon}^{dew}=d\lambda\frac{\partial F}{\partial\boldsymbol{\sigma}}$, where λ is the scalar function of the current dewetting strains and the current dewetting stress.

The second approach is that the hyper visco-elastic constitutive equations supposed to be dynamically changed i.e. shear and volumetric stiffness should be reduced during the dewetting process. Such an approach can be met in works (12-15). In terms of this approach, it can be noticed the similarity between the modelling of the particulate reinforced rubber-like materials and the modeling of the fiber reinforced composite materials. For example, the idea of the dynamic changing of the stiffness is well described in the article (16) for the multilayered composites.

It is very important to understand that nonlinear elastic or plastic behavior are indistinguishable from dewetting during the loading; however, these three are significantly different upon examination of the unloading behavior (11). The main difference between plasticity and dewetting is that the dewetting effect does not suppose to have large irreversible strains like in plasticity. Figure **3** (11) schematically shows the difference between plasticity and dewetting.

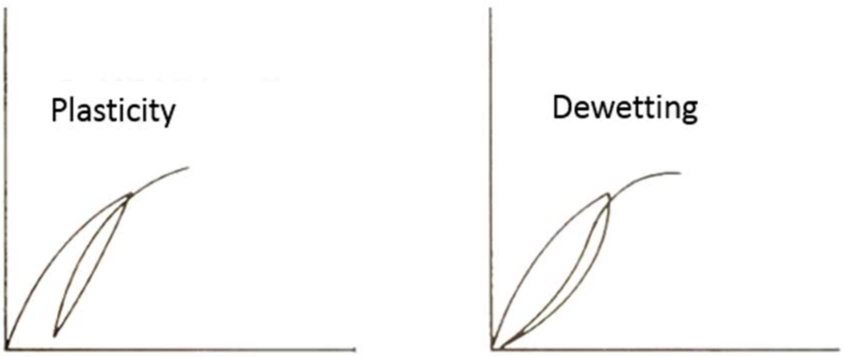

Fig. **3**. Difference between plasticity and dewetting.

Also there are several approaches for the dewetting surfaces. Here there are some of them:

$F(p,q) = q^2 - \mu p^2 - d^2$, where μ, d can be functions of strains, strain rates and temperature (9-14),

$F(p,q) = \begin{Bmatrix} q^2 - B, p > 0 \\ q^2 + Ap^2 - B, p < 0 \end{Bmatrix}, A, B$ could be constants or functions of strains, strains rates and temperature (7). Here we see a good idea to use stress state sensitive criteria. There are several works where we can see the usage of the dewetting surface similar to Drucker-Prager criteria of plasticity $F(p,q) = q - \mu p - d$, for example in the article (11).

Since the behavior of the particulate reinforced composites is stress state sensitive, it is important to introduce the parameter which can be used to classify the stress states. Stress triaxiality parameter $k = -\frac{p}{q}$ best of all is suitable for this role. It possesses the following values:

$$k = -\frac{p}{q} = \begin{Bmatrix} -\infty, \sigma_{11} = \sigma_{22} = \sigma_{33} < 0 \\ -\frac{2}{3}, \sigma_{11} = \sigma_{22} < 0, \sigma_{33} = 0 \\ -\frac{1}{3}, \sigma_{11} < 0, \sigma_{22} = \sigma_{33} = 0 \end{Bmatrix}, \quad k = -\frac{p}{q} = \begin{Bmatrix} +\infty, \sigma_{11} = \sigma_{22} = \sigma_{33} > 0 \\ +\frac{2}{3}, \sigma_{11} = \sigma_{22} > 0, \sigma_{33} = 0 \\ +\frac{1}{3}, \sigma_{11} > 0, \sigma_{22} = \sigma_{33} = 0 \end{Bmatrix}.$$

In the work (17) the stress triaxiality parameter is successfully used to develop constitutive equations for hyper-elastic materials.

The idea of stress state dependent plastic surface is described in the article (18). In this article, the plastic surface is taken in the form of modified Drucker-Prager criteria $F(p,q) = q - \mu p - d$ where μ, d are the functions of plastic strains and superimposed hydrostatic pressure. Results, represented in the work (18) in combination with the research (17) can be used to develop the dewetting surface i.e. the dewetting criterion for the particulate reinforced rubber-like materials in the following form: $F(p,q) = q - \mu p - d(\varepsilon^{dew}, k)$.

3. SUPERIMPOSED HYDROSTATIC PRESSURE

The superimposed hydrostatic pressure sensitivity is illustrated on the Fig. **1**. On this figure it is possible to observe how the ultimate strength of material is increased by superimposing of the pressure. As it has already been noted, hydrostatic pressure helps avoid voids formation, therefore it is visible that the dilatation decreases with the increase of the pressure. However, superimposed hydrostatic pressure has no noticeable effect on the investigated material behavior until dewetting is observed.

4. STRAIN RATE EFFECT

Figure **2** represents strain rate sensitivity. With increasing the strain rate, the maximum nominal stresses increase. The dilatation also increases with the increase of the strain rate. It is possible to see (Fig. **2**) that the strain rate influences the undewetted and dewetted regions of stress-strain curves.

General equation for hyper-visco-elastic material can be written in the following form (2):

$$\boldsymbol{\tau}^D(t) = \boldsymbol{\tau}_0^D(t) + dev(\int_0^{\tau} \frac{\dot{G}(\tau')}{G_0} \overline{\mathbf{F}}_t^{-1}(t-t') \cdot \boldsymbol{\tau}_0^D(t-t') \cdot \overline{\mathbf{F}}_t^{-T}(t-t') d\tau'),$$

$$\boldsymbol{\tau}^H(t) = \boldsymbol{\tau}_0^H(t) + \int_0^{\tau} \frac{\dot{K}(\tau')}{K_0} \boldsymbol{\tau}_0^H(t-t') d\tau',$$

where dev – the deviator operator, $\boldsymbol{\tau}_0^D(t)$ and $\boldsymbol{\tau}_0^H(t)$ - the deviatoric and the hydrostatic parts of the instantaneous Kirchhoff stress tesor, $\boldsymbol{\tau}^D(t)$ and $\boldsymbol{\tau}^H(t)$ the deviatoric and the hydrostatic parts of the entire Kirchhoff stress tensor, G_0 and K_0 - are the instantaneous shear and bulk moduli, $G(t)$ and $K(t)$ - time-dependent shear and bulk relaxation moduli, $\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}$, $\mathbf{x}$ - the current position of a material point, $\mathbf{X}$ - the reference position of the same point, $J = \det(\mathbf{F})$, $\overline{\mathbf{F}} = J^{-\frac{1}{3}}\mathbf{F}$, $\overline{\mathbf{F}}_t(t-t')$ - is the distortional deformation gradient of the state at $t-t'$ relative to the state at t, $\frac{d\tau}{dt} = \frac{1}{A_\theta(\theta(t))}$, θ is a temperature and $A_\theta(\theta(t))$ is the shift function and can be used to take into account the temperature.

It is assumed, that the instantaneous response of the material follows from the hyperelastic constitutive equations $\boldsymbol{\tau}_0(t) = \boldsymbol{\tau}_0^D(\overline{F}(t)) + \boldsymbol{\tau}_0^H(J(t))$.

It can be used any strain energy potential where in the easy cases (not always) the strain energy is the additive function of the deviatoric and volumetric part: $U = U_{dev}(\overline{I}_1, \overline{I}_2) + U_{vol}(J)$, for example in the polynomial form

$U = \sum_{i+j=1}^{N} C_{ij}(\overline{I}_1 - 3)^i (\overline{I}_2 - 3)^j + \sum_{i=1}^{N} \frac{1}{D_i}(J-1)^{2i}$, where $\overline{I}_1 = trace(\overline{\mathbf{F}} \cdot \overline{\mathbf{F}}^T)$, $\overline{I}_2 = \frac{1}{2}(\overline{I}_1^2 - trace((\overline{\mathbf{F}} \cdot \overline{\mathbf{F}}^T) \cdot (\overline{\mathbf{F}} \cdot \overline{\mathbf{F}}^T)))$, C_{ij} and D_i - material functions.

These constitutive equations have to be complemented with the dewetting surface (criterion) and the damage parameters, which will allow operating the softening of the material during the dewetting.

5. MULLINS EFFECT

As mentioned above, the softening of the material during the unloading and reloading is known as Mullins effect (19). Figure **4** (20) shows schematic loading-unloading curves in simple tension for the material, which possesses Mullins effect.

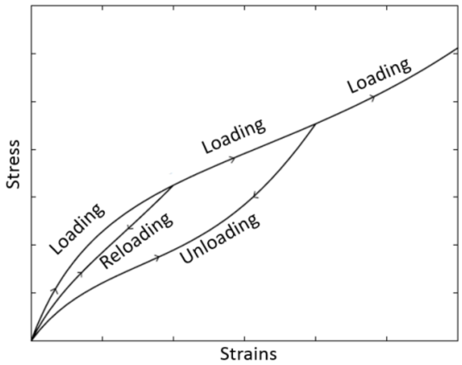

Fig. **4**. Schematic loading-unloading-reloading curves in simple tension for the material which possesses Mullins effect.

Usually Mullins effect can be accounted by definition of the damage function. For example, in (20) Mullins effect is taken into account by using of augmented energy function of the form $U(\bar{I}_1,\bar{I}_2,\eta)=\eta U_{dev}(\bar{I}_1,\bar{I}_2)+\varphi(\eta)$. The function $\varphi(\eta)$ is a continuous function of the damage variable η and is referred to as the "damage function". In the (21) the extension of the augmented energy function in the form of $U(\bar{I}_1,\bar{I}_2,\eta)=\eta U_{dev}(\bar{I}_1,\bar{I}_2)+\varphi(\eta)+U_{dev}(J)$ is proposed to account compressibility of the material. The deviatoric part of the augmented energy function is related to the deviatoric part of the energy corresponding to the primary response by the scaling factor η. The volumetric part of the augmented energy function is the same as that for the primary response. In this concept Mullins effect is associated only with the deviatoric part of the deformation. When $\eta=1$ it is required that $\varphi(\eta)=0$, such that $U(\bar{I}_1,\bar{I}_2,1)=U_{dev}(\bar{I}_1,\bar{I}_2)+U_{dev}(J)$ and the augmented energy function reduces to the strain energy function of the primary material response. This situation corresponds to η being inactive and physically represents the energy of a material point that is on the primary curve. η and consequently $\varphi(\eta)$ vary continuously as the deformation proceeds. The value of η will depend on the maximum values of the principal stretches attained on a primary loading path. On the primary curve at any point where the unloading is initiated $\eta=1$. In (20) it is described in details how to evaluate η and $\varphi(\eta)$ for the incompressible material. In (21) it is written how to evaluate these functions for the compressible material case.

6. TEMPERATURE EFFECT

Usage of the shift function $A_\theta(\theta(t))$ defined in the previous section to process temperature effects is the most common approach. Dependence of stress-strain curves on the temperature is shown on the Fig. **5** (7).

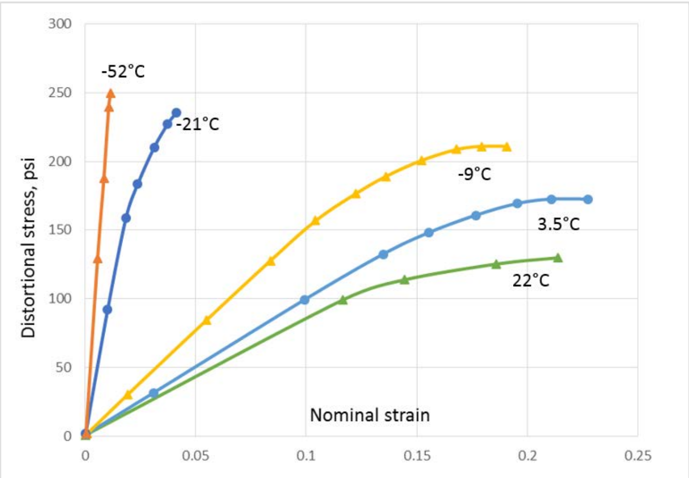

Fig. **5**. Temperature effect particulate reinforced composites.

7. SIZE DEPENDENCE

In the report (5) it is described the set of the tensile tests for the specimens with different area and with the usage of different gages length. A summary of the test parameters is represented in the Table **1**. The test data are shown on the Fig. **6**. Based on the charts from the Fig. **6,** it is possible to conclude that both area and gage length have an effect upon the measured parameters. The size effect still needs to be investigated.

Table **1**. Test parameters

Gage length (G), in	Area (A), sq in.	A/G
4.00	7.58	1.89
2.70	5.05	1.87
1.35	2.56	1.89
4.00	2.56	0.64
2.70	1.70	0.63
1.35	0.90	0.64

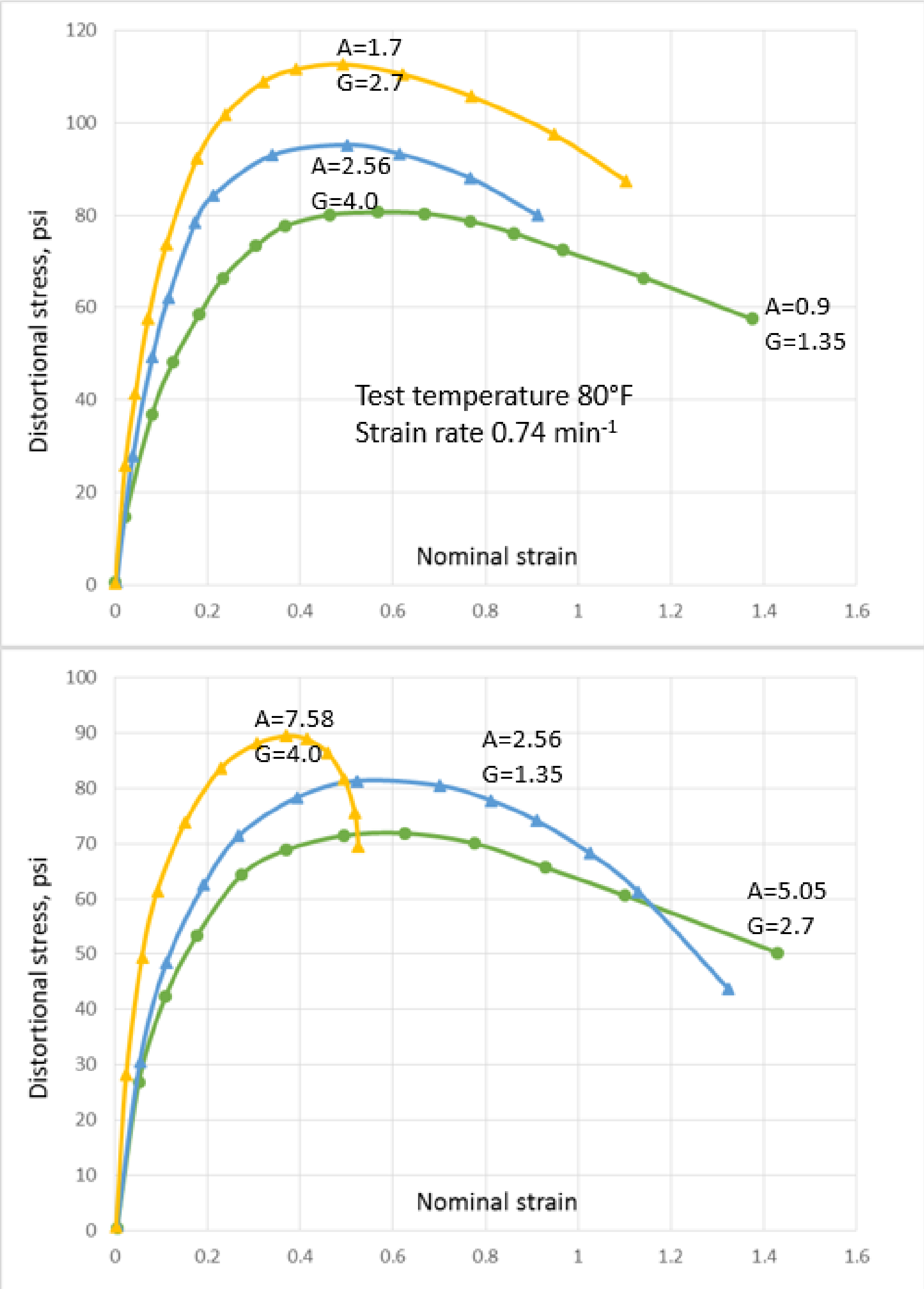

Fig. **6**. Comparison of the effect of the sample size upon the mechanical properties of the particulate reinforced rubber-like material.

8. COUPLING OF BULK AND DEVIATORIC BEHAVIOR

A 10% increase in dilatation is not uncommon in pressurized distortional experiments, even though the mean pressure is negative (8). Normally one associates increases in volume with positive normal stresses and decreases in volume with compressive normal stresses. These normal dilatational effects are observed for pure hydrostatic conditions (i.e. no distortional stresses or strains) however when distortion is present it is not uncommon to observe that the so-called bulk modulus, changes signs. Volumetric strains start being zero when superimposed pressure is zero, become slightly negative as hydrostatic pressure is applied and then increases to large positive values as the material is distorted in a constant superimposed pressure environment. In going from a negative to a positive value, the dilatation must naturally pass through zero and since the mean pressure is not generally zero at this point, the bulk moduli must start at about 1000000 psi and increase to positive infinity with distortion, change signs, becoming negative infinity, and increase to roughly 200 psi at large distortions. From the mathematical modeling point of view, this means that in the strain energy potential deviatoric and bulk terms should be coupled.

9. CONCLUSIONS

Based on the available test and analytical data it is possible to conclude that the mathematical modelling of the particulate reinforced materials is a very complex problem. The main task for researchers dealing with such materials is to develop finite strain visco-elastic constitutive equations. Also, there is a very important task to develop damage criterion (dewetting surface) and based on this criterion define the damage functions which will be able to control loading/unloading/reloading process. Based on the literature data it is possible to note that the particulate reinforced composites behavior before dewetting and after dewetting is stress state sensitive. This means that both the constitutive equations and the dewetting surface must be functions of stress state. Analysis of test results presented in the literature allows us to assume that stress triaxiality parameter can be successfully used to account the stress state sensitivity of the investigated material. The modeling problem of the hyper visco-elastic particulate reinforced rubber-like materials is not limited by mechanic or numerical mathematic. This problem should be solved in a strong cooperation with the experimenters, which should take part in the developing of the test program to obtain the proper material constants and functions.

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