Macromechanical evaluation of random strength of heterogeneous materials

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The random strength of heterogeneous materials is considered as a function of volume fraction of the material components. A general macromechanical probabilistic model is created that permits one to consider all experimental results jointly, and therefore, minimize the experimental programme. A probabilistic description and corresponding method of statistical treatment, taking into account variability of the volume fraction on both the average strength and strength scatter, is offered. Experimental confirmation of the approach is presented using three examples of polymer blends of polyethylene and polypropylene. The influence of volume fraction scatter is analysed in detail, and it is noted that the scatter may lead to a significant increase in variability of the strength. A generalization of the approach is considered for other relevant problems, such as random thermal expansion as a function of temperature and random strength of multi-component (hybrid) materials.

1. Introduction

Reliable design of materials and structures may be carried out only using well-defined information regarding properties of materials and service conditions. Generally, the properties, such as strength, possess significant variability which may be described in a probabilistic framework. Moreover, the failure of materials has an obvious stochastic nature. Thus, the random response of materials should be taken into account along with information about the stochastic nature of loading and environments. It gives an opportunity to develop probabilistic design methods, numerically predict reliability as a probability of meeting the certain product requirements [1], and solve important practical problems, such as size effect evaluation, probabilistic optimization, statistical treatment of experimental results, etc.

Evaluation of the random strength of composite materials is especially difficult because of their heterogeneity and a practically infinite number of possible microstructures. In other words, a particular strength distribution is required for each possible microstructure. Existing methods of micromechanics allow one to predict the deterministic response of many composite (heterogeneous) materials from the properties of components and the geometry of their distribution. There are, however, many instances where the microstructure of the material is too complex for welldefined micromechanical modelling and analysis. Moreover, properties of the components cannot always be assumed to be the same as the pure constituents because of physical-chemical changes that occur during materials manufacturing and storage. One can also note complex changes in the material microstructure due to, for example, interface debonding or accumulation of microdamages. Consequently, a stochastic "macromechanical" analysis is often the only way to obtain a reliable statistical information on the heterogeneous materials strength. This usually requires the use of labour-intensive and time-consuming experimental programmes, since it is necessary to obtain statistical information separately for each considered microstructure and/or environmental condition.

An approach which allows one to minimize the amount of data required is to create a general macromechanical probabilistic model which analyses all experimental results together. Thus, an objective of the paper is to develop a simple and less time-consuming method of macromechanical evaluation of the random strength of heterogeneous materials and show its application for typical polymer compositions.

2. Probabilistic model of strength

Generally, the reliability of materials or structures, H, may be evaluated as [1]

$$H = P\left\{\tilde{R} \ge \tilde{Q}\right\} \tag{1}$$

where $P\{\tilde{R} \ge \tilde{Q}\}$ is the probability of the random event $\tilde{R} \ge \tilde{Q}$; \tilde{Q} is the random parameter(s) of loading; and \tilde{R} is the random characteristic(s) of the strength response. (Here and further, sign "" above variables shows their random nature; sign"" represents the average value.) Numerical evaluation of the reliability, using the Equation 1, depends on the statistical distributions of \tilde{R} and \tilde{Q} . Solutions to particular problems may be found, for example, in [2]. Although any experimentally determined statistical distributions of strength may be used in principle, such distributions as normal, Weibull, and log-normal are the most popular ones in numerous engineering and scientific applications. These distributions may be determined by two statistical parameters only. Reliable evaluation of the main statistical characteristics may therefore be expressed in terms of the average value and the standard deviation.

In describing deterministically the strength of a heterogeneous material as a function of volume fraction, each potential mechanism of failure should be considered separately. Therefore, the particular mode of failure at a given volume fraction is determined by the mechanism providing the minimum value of strength and, generally, a deterministic evaluation of the strength may be represented as a piecewise-smooth function of the volume fraction. In considering the problem in a stochastic framework, all possible mechanisms are analysed together using certain probabilistic models, i.e. all mechanisms are taken into account with different probabilities depending on the volume fraction. Separate potential mechanisms of failure are not considered in detail using a "macromechanical" approach. However, one can make a conclusion that the dependence of the random strength on the volume fraction may be written as "one" stochastic smooth function in a macromechanical statement of the problem.

The dependence of the random strength, \tilde{R} , of a two-component heterogeneous material on volume fraction, ψ , may be represented in a general form as:

$$\tilde{R}(\psi) = \bar{R}(\psi) + \tilde{r}\bar{R}^*(\psi) \tag{2}$$

Here, \tilde{r} is the random variable, which determines the strength scatter; $\bar{R}(\psi)$ is the deterministic function of ψ showing the strength average value; $\bar{R}^*(\psi)$ is the deterministic function reflecting the dependence of the strength scatter on volume fraction (here, ψ reflects the volume content of one of the two components). We also suppose that distribution of \tilde{r} is determined by the following main statistical characteristics: σ_r is the standard deviation and $\bar{r} = 0$ is the average value. For practical applications, it is convenient to use power series for approximations of the functions $\bar{R}(\psi)$ and $\bar{R}^*(\psi)$:

$$\bar{R}(\psi) = \sum_{j=0}^{m} \bar{R}_{j} \psi^{j}$$
 $\bar{R}^{*}(\psi) = \sum_{j=0}^{m} \bar{R}_{j}^{*} \psi^{j}$ (3)

It is easily shown that in the case of the non-random nature of the volume fraction, the average value, \overline{R} , and the standard deviation, σ_R , of strength may be evaluated as:

$$\bar{R} = \bar{R}(\psi) \qquad \sigma_R = \sigma_r \bar{R}^*(\psi)$$
 (4)

In a case of a normally distributed variable \tilde{r} , the distribution of \tilde{R} will also be normal, with a probability density function, p_R , as:

$$p_R(R, \psi) = \frac{1}{(2\pi)^{1/2} \sigma_R(\psi)} \exp\left\{-\frac{[R - \bar{R}(\psi)]^2}{2\sigma_R^2(\psi)}\right\}$$
(5)

Let us note that any other distributions may be utilized in principle. For example, for the two-parameter Weibull distribution in the form

$$p_R(R, \psi) = \frac{b}{A^{\beta}} R^{\beta - 1} \exp\left[-\left(\frac{R}{A}\right)^{\beta}\right]$$
(6)

The respective statistical characteristics A and β may be calculated on the basis of the following relationships [3]:

$$\frac{\Gamma(1+2/\beta)}{\Gamma^2(1+1/\beta)} - 1 = v_R^2(\psi) \qquad A = \bar{R}(\psi)/\Gamma(1+1/\beta) \quad (7)$$

where $v_R(\psi) = \sigma_R(\psi)/\overline{R}(\psi)$ is the coefficient of variation of strength; Γ is a Gamma-function.

3. Evaluation of statistical parameters

Strength parameters of Equations 2 and 3, \bar{R}_j , \bar{R}_j^* ; j = 1, ..., m; and σ_r , can be evaluated using a statistical treatment of relevant experimental data. However, application of the classical least squares method may lead to incorrect results, because, in a general case, the strength scatter also depends on volume fraction. Fig. 1 illustrates the difference between the classical and proposed methods: in the case of the least squares method application, the strength scatter is constant (Fig. 1a), while the introduced probabilistic model takes into account the actual variation (Fig. 1b). Obviously, in a particular case of the model when $\bar{R}^*(\psi) = 1 = \text{constant}$, statistical evaluation of the parameters is reduced to application of the least squares method.

We suppose that there is a sample of experimental values R_i , ψ_i , i = 1, ..., n where *n* is a number of the measurements. In contrast with the classical least squares method, the difference between the measured $(\tilde{R}(\psi))$ and expected mean $(\bar{R}(\psi))$ values will be a function of volume fraction:

$$\tilde{R}(\psi) - \bar{R}(\psi) = \tilde{r}\bar{R}^*(\psi) \neq \text{constant}$$
 (8)

However, the random expression

$$\frac{\tilde{R}(\psi) - \bar{R}(\psi)}{\bar{R}^*(\psi)} \tag{9}$$

is not a function of ψ and should be distributed as \tilde{r} . Therefore, the appropriate minimization procedure to obtain \bar{R}_j may be written by analogy with the least



Figure 1 Schematic distribution of the strength scatter using (a) the classical least squares method and (b) the approach presented.

squares method as:

$$\Phi(\bar{R}_j) = \frac{1}{n} \sum_{i=1}^n \left\{ \frac{R_i - \bar{R}(\psi_i)^2}{\bar{R}^*(\psi_i)} \right\} \to \min$$
(10)

where R_i is the *i*th experimental value of the strength corresponding the volume fraction ψ_i : $\overline{R}(\psi_i)$ and $\overline{R}^*(\psi_i)$ are the theoretical (expected) values given by Equation 3 at $\psi = \psi_i$ as:

$$\bar{R}(\psi_i) = \sum_{j=0}^{m} \bar{R}_j \psi_i^j \qquad \bar{R}^*(\psi_i) = \sum_{j=0}^{m} \bar{R}_j^* \psi_i^j \quad (11)$$

The minimization procedure is carried out by letting:

$$\partial \Phi(\bar{R}_j) / \partial \bar{R}_j = 0 \qquad j = 0, \dots, m$$
 (12)

The coefficients \overline{R}_j may thus be obtained as:

$$[\bar{R}] = [B]^{-1}[C]$$
(13)

where $[\bar{R}]_t = [\bar{R}_0 \bar{R} \dots \bar{R}_m]$ and components of the matrices [B] and [C] are calculated as:

$$B_{jk} = \sum_{i=1}^{n} \frac{\psi_i^{j+k-2}}{\left(\sum_{l=0}^{m} \bar{R}_l^* \psi_i^l\right)^2} \quad C_j = \sum_{i=1}^{n} \frac{\psi_i^{j-1} R_i}{\left(\sum_{l=0}^{m} \bar{R}_l^* \psi_i^l\right)^2} \quad (14)$$

Let us note that in the classical least squares method, the function $\overline{R}^*(\psi_i) = \sum_{l=0}^m \overline{R}_i^* \psi_i^l$ is assumed to be equal to 1. One can see from Equations 13 and 14 that determination of the coefficients \overline{R}_j (j = 0, ..., m) are based on information regarding \overline{R}_j^* (j = 0, ..., m), which is also unknown. Thus, the following iterative procedure is proposed (Fig. 2). At the first iteration, we



Figure 2 Flow-chart of numerical evaluation of the statistical parameters.

suppose that the function $\overline{R}^*(\psi)$ is a constant, i.e. $\overline{R}_0^* = 1$; $\overline{R}_j^* = 0$; j = 1, ..., m. Then, after the calculation of \overline{R}_j using the Algorithm 13–14, the "actual absolute errors" $\Delta R(\psi_i)$ are calculated for each measurement (i = 1, ..., n) as:

$$\Delta R(\psi_i) = |R_i - \bar{R}(\psi_i)| \tag{15}$$

Calculation of \overline{R}_{j}^{*} (j = 0, ..., m) is carried out on the basis of the classical least squares method for sample of $\Delta R(\psi_i)$; i = 1, ..., n. The standard deviation of \tilde{r} may be obtained as:

$$\sigma_r = \left\{ \frac{1}{n} \sum_{i=1}^n \left[\frac{R_i - \bar{R}(\psi_i)}{\bar{R}^*(\psi_i)} \right]^2 \right\}^{1/2}$$
(16)

This cycle of statistical evaluation is developed again using obtained values of \overline{R}_j^* (j = 0, ..., m). A condition of convergence is based on the required accuracy. That is the difference in results between the nearest iterations should be less than desired accuracy.

4. Experimental programme

Let us consider, as an example of the proposed approach, statistical evaluation of random tensile strength of three different polymer blends. The components of the blends are: low density polyethylene, polypropylene, maleated polyethylene (Polybond 3001) and maleated polypropylene (Polybond 3009). The maleated polyolefins were commercial products supplied by Uniroyal Chemical. Pellets of these materials were blended in a twin screw Brabender mixer at 190 °C for 12 min at 50 r.p.m. Three different types of blends were thus obtained: mixtures of neat polyethylene and polypropylene (material "a"); mixtures of polyethylene and polypropylene with 25% by weight of maleated polyethylene and maleated polypropylene (material "b"); and mixtures of maleated polyethylene and maleated polypropylene (material "c"). Dogbone-shaped specimens were compression moulded in a Wabash heat press at 200 °C for 5 min and then water quenched under pressure. Tensile strength was measured with an Instron 1011 tensile machine, equipped with a 100 lb (45.4 kg) load cell; the crosshead speed was 10 mm min⁻¹.

Seven different compositions were prepared for each type of blend at $\psi = 0$; 0.1; 0.3; 0.5; 0.7; 0.9; 1. Here, ψ is the volume fraction of polypropylene in the "a" blends; the total volume fraction of neat and maleated polypropylene in the "b" blends; and the volume fraction of maleated polypropylene in the "c" blends.

Values for the \bar{R}_j , \bar{R}_j^* , σ_r of Equations 2 and 3 used to characterize the random strength (at m = 4) are presented in Table I. (The requirement of accuracy has been chosen as 0.001 for σ_r .) There is, in general, a difference between statistical parameters evaluated at the first iteration (the classical least squares method) and at the last iteration (the approach presented). Although the main differences are shown for parameters \bar{R}_j^* , significant distinctions may be noted for σ_r and \bar{R}_j as well. Experimental distributions of

TABLE I	Statistical	parameters	of	tensile	strength
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Parameter	Material "a" Iteration	Material "a" Iteration		Material "b" Iteration		Material "c" Iteration	
	the 1st	the last	the 1st	the last	the 1st	the last	
n	50	50	41	41	37	37	
σ_r	0.963	1.098	0.646	1.399	0.659	1.150	
\overline{R}_0	36.97	36.84	38.50	38.44	37.51	37.48	
\overline{R}_1	- 63.78	- 51.73	-29.50	-32.24	- 6.89	-6.24	
\overline{R}_2	109.42	50.52	- 17.89	1.06	- 51.36	- 53.66	
\overline{R}_3	-131.72	-44.18	30.95	- 3.38	89.21	90.62	
\overline{R}_4	59.57	18.98	-8.68	9.65	- 44.45	-44.06	
\overline{R}_0^*	0.698	0.709	0.382	0.402	0.492	0.486	
\bar{R}_1^*	3.906	-0.133	1.645	2.550	2.242	2.180	
\overline{R}_{2}^{*}	-8.077	18.235	-4.565	-9.041	-14.124	-13.930	
\overline{R}_{3}^{*}	1.259	-44.857	5.039	10.817	27.728	27.803	
$ar{R}_4^*$	2.380	26.275	- 2.249	- 4.546	- 16.167	- 16.397	

 \bar{R}_j, \bar{R}_j^* in MPa.



Figure 3 Experimental distributions of \tilde{r} (here and further, indices a, b, c correspond to the material "a", "b", and "c" respectively).



Figure 4 Dependence of the average strength on volume fraction: -a; -b; ----c.

 \tilde{r} calculated using Expression 9 are shown in Fig. 3. The distributions reflect the random nature of strength and permit one to predict the random strength distribution at any value of volume fraction. The non-linear character of the average strength $\overline{R}(\psi)$ is shown in Fig. 4. Although the non-linearity of the average strength may be estimated using conventional statistical methods, the non-linearity of the dependence of the strength scatter on volume fraction, $\overline{R}^*(\psi)$, can be taken into account only by using the approach presented. The experimental results presented in Fig. 5 show the complex nature of functions $R^*(\psi) = |R_i - \overline{R}(\psi_i)|$, and especially, for materials "a" and "b". Therefore, the non-linear character of the strength should be taken into consideration not only for analysis of the average characteristics, but also for the scatter.

5. Effect of volume fraction scatter

In a more general case, volume fraction should be considered as a random parameter as well. Variations in the volume fraction inevitably occur as the result of non-uniform blending procedures, especially resulting in non-uniform distribution of components at the microlevel. Let us assume that the random volume fraction $\tilde{\Psi}$ is determined by the average value $\bar{\Psi}$ and the standard deviation σ_{ψ} . Therefore, non-linear function $\tilde{R}(\tilde{\Psi})$ in accordance with Equation 2 may be



Figure 5 Experimental dependence of the function \overline{R}^* on volume fraction.

written in the following form:

$$\widetilde{R}(\widetilde{\Psi}) = \sum_{j=0}^{m} \overline{R}_{j} \widetilde{\Psi}^{j} + \widetilde{r} \sum_{j=0}^{m} \overline{R}_{j}^{*} \widetilde{\Psi}^{j}$$
(17)

Then, the main statistical characteristics of $\tilde{R}(\tilde{\Psi})$ can be obtained using a linear approximation [4] in the following form

$$\tilde{R}(\tilde{\Psi}) \approx \frac{\partial R(\Psi = \Psi, \tilde{r} = \bar{r})}{\partial \Psi} (\tilde{\Psi} - \bar{\Psi}) + \frac{\partial R(\tilde{\Psi} = \bar{\Psi}, \tilde{r} = \bar{r})}{\partial r} (\tilde{r} - \bar{r}) + \bar{R}(\bar{\Psi}) \quad (18)$$

Taking into account Approximations 2–3, one can easily obtain:

$$\tilde{R}(\tilde{\Psi}) \approx \bar{a}\tilde{\Psi} + \bar{b}\tilde{r} + \bar{c} \tag{19}$$

where

$$\bar{a} = \sum_{j=1}^{m} j\bar{R}_{j}\bar{\psi}^{j-1} \quad \bar{b} = \sum_{j=0}^{m} \bar{R}_{j}^{*}\bar{\psi}_{j}$$
$$\bar{c} = -\sum_{j=0}^{m} (j-1)\bar{R}_{j}\bar{\psi}^{j} \qquad (20)$$

Thus, the average strength $\overline{R}(\widetilde{\Psi})$ and the standard deviation of $\sigma_R(\widetilde{\Psi})$ are calculated as:

$$\bar{R}(\tilde{\Psi}) \approx \bar{a}\bar{\Psi} + c = \sum_{j=0}^{m} \bar{R}_{j}\bar{\Psi}^{j}$$
(21)

$$\sigma_R(\tilde{\psi}) \approx (\bar{a}^2 \sigma_{\psi}^2 + \bar{b}^2 \sigma_r^2)^{1/2}$$
(22)

Expression 22 reflects the dependence of the strength variability on the volume fraction scatter. Fig. 6 shows, as an example, dependence of $\sigma_R(\tilde{\psi})$ on σ_{ψ} for the above-mentioned materials "a", "b", and "c". Significant influence of the volume fraction scatter on random strength distribution can be seen. Therefore, reducing the variability of volume fractions leads to a reduction of strength scatter, and hence, to an increase in reliability.

6. Generalization of the model

Although the approach presented is considered chiefly for random strength evaluation, its application may be



Figure 6 Dependence of the strength standard deviation on the standard deviation of volume fraction at $\bar{\psi} = 0.5$: ---a; ---b; ----c.

useful in certain adjacent problems of stochastic mechanics of materials. In more general form, a random function $\tilde{Y}(x)$ may be written as

$$\tilde{Y}(x) = \bar{Y}(x) + \tilde{r}\tilde{Y}^*(x) \tag{23}$$

where \tilde{Y} is some response; x is a relevant parameter of structure or environments; approximation of $\bar{Y}(x)$ and $\bar{Y}^*(x)$ are chosen, for example, in the form of power series. Then, the method of statistical evaluation of functions $\bar{Y}(x)$, $\bar{Y}^*(x)$ and parameter σ_r may be carried out on the basis of the above-mentioned algorithm as well.

Let us further consider an example regarding random thermal expansion. Generally, thermal expansion of typical composite and polymer materials is a nonlinear function of temperature *T*. Therefore, a thermal expansion function, $\alpha(T)$, should be used instead of a traditional coefficient of linear expansion, α , [5]. Random function of thermal expansion may be represented as

$$\tilde{\alpha}(T) = \bar{\alpha}(T) + \tilde{r}\bar{\alpha}^*(T) \tag{24}$$

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where

$$\bar{\alpha}(T) = \sum_{j=0}^{m} \bar{\alpha}_j T^j \qquad \bar{\alpha}^*(T) = \sum_{j=0}^{m} \bar{\alpha}_j^* T^j \qquad (25)$$

Hence, the average, $\bar{\alpha}(T)$, and correlation, $K_{\alpha}(T, T')$, functions are determined as:

$$\bar{\alpha}(T) = M\{\tilde{\alpha}(T)\} = \sum_{i=0}^{m} \bar{\alpha}_{j} T^{j}$$
(26)

$$K_{\alpha}(T, T') = M\{ [\tilde{\alpha}(T) - \bar{\alpha}(T)] [\tilde{\alpha}(T') - \bar{\alpha}(T')] \}$$
$$= \sigma_r^2 \bar{\alpha}^*(T) \bar{\alpha}^*(T') = \sigma_r^2 \sum_{j=0}^m \bar{\alpha}_j^* T^j \sum_{j=0}^m \bar{\alpha}_j^* T'^j$$
(27)

Here, $M\{.\}$ is the operator of averaging. Random thermal strains, $\tilde{\varepsilon}(T, T_0)$, thus may be evaluated as:

$$\tilde{\varepsilon}(T, T_0) = \int_{T_0}^T \tilde{\alpha}(T) \,\mathrm{d}T \tag{28}$$

with the following average and correlation functions:

$$\bar{\varepsilon}(T, T_0) = \int_{T_0}^T \bar{\alpha}(T) \,\mathrm{d}T \tag{29}$$

$$K_{\varepsilon}(T, T', T_{0}) = \int_{T_{0}}^{T'} \int_{T_{0}}^{T} K_{\alpha}(T, T') \, \mathrm{d}T \, \mathrm{d}T'$$
$$= \sigma_{r}^{2} \int_{T_{0}}^{T} \bar{\alpha}^{*}(T) \, \mathrm{d}T \int_{T_{0}}^{T} \bar{\alpha}^{*}(T') \, \mathrm{d}T' \qquad (30)$$

where T_0 is the initial temperature. Experimental results regarding stochastic thermal expansion of composites may be found, for example, in [6, 7]. Statistical evaluation of unknown parameters may also be developed using the algorithm presented in Fig. 2.

The approach presented may also be utilized for analysis of more complex problems regarding multidimensional statements. Let us consider a hybrid composite material consisting of more than two components. In this case, function of random strength may be also written as:

$$\widetilde{R}(\psi_1, \dots, \psi_{k-1}) = \overline{R}(\psi_1, \dots, \psi_{k-1}) + \widetilde{r}\overline{R}^*(\psi_1, \dots, \psi_{k-1})$$
(31)

where k is a number of components; ψ_l is the volume fraction of *l*th component. For example of a three-component hybrid (k = 3), approximations of $\overline{R}(\psi_1, \psi_2)$ and $\overline{R}^*(\psi_1, \psi_2)$ may be written as:

$$\bar{R}(\psi_1, \psi_2) = \sum_{j'=0}^{\infty} \sum_{j''=0}^{\infty} \bar{R}_{j'j''} \psi_1^{j'} \psi_2^{j''}$$

$$\bar{R}^*(\psi_1, \psi_2) = \sum_{j'=0}^{\infty} \sum_{j''=0}^{\infty} \bar{R}_{j'j''}^* \psi_1^{j'} \psi_2^{j''} \qquad j'+j'' \leqslant m \quad (32)$$

Thus, the problem of statistical evaluation is reduced to a problem of determination of $\overline{R}_{j'j''}$, $\overline{R}_{j'j''}^*$, σ_r , and the algorithm presented in Fig. 2 allows one to receive these parameters as well. The only difference is associated with the type of approximations and the number of unknown parameters.

7. Conclusion

The approach presented possesses two main advantages. The first one is the ability to consider all experimental results together using the proposed probabilistic model. Cost and time of the experimental programme may therefore be significantly reduced. The second advantage consists of the ability to take into account the unknown nature of both $\overline{R}(\psi)$ and $\overline{R}^*(\psi)$. It allows one to consider more accurate stochastic description, and hence, to receive (or predict) more reliable results regarding actual strength response. Experimental results for three kinds of typical polymer compositions do confirm a non-linear character of $\overline{R}^*(\psi)$ and show, in a general case, the difference between the approach presented and the classical least squares method. Although the main attention has been paid to the random strength of two-component heterogeneous materials, application of the approach may be utilized for stochastic analysis of other relevant problems, such as strength of hybrid materials and macromechanical evaluation of thermal expansion. Effect of the volume fraction scatter is noted: reducing the scatter may be considered as a way of improvement of actual strength response and increasing the materials reliability.

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