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PHYSICAL DISCRETIZATION APPROACH TO EVALUATION OF ELASTIC MODULI OF HIGHLY FILLED GRANULAR COMPOSITES

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Abstract—A structural model for highly filled granular composites is suggested. Composites investigated represent rather dense random assemblies of hard spherical particles embedded in softer elastic matrices. The concept forming the foundation of the model approach is the so-called physical discretization principle which comes from the fact that the really complicated field interactions in the system considered here can be substituted by simpler linear elastic ones. The indispensable stages of model development are the generation of random composite architecture and the solution of some special boundary problems. The model reveals clearly the influence of structural parameters on macroscale behavior. The predicted properties proved to be in reasonable agreement with experimental observations. Of particular concern is the insight into the internal pattern of structural forces that makes it possible to obtain new information about the peculiarities of composite internal elastic resistance.

1. INTRODUCTION

At present, the structural modelling of mechanically non-homogeneous composite materials seems to be one of the most promising research trends. Structural models help researchers to get clear views of the relationships between micro- and macromechanical behaviour of composite materials. This is especially important when improvements in properties are sought.

There exists a great variety of materials whose main structural feature can be represented as assemblies or clusters of rather closely spaced round particles such as polymeric matrix composites, filled rubbers, some natural materials like sand and so on. Two different approaches to structural properties investigations of these materials are developed. The first one which might be referred to as "geomechanical" focuses attention mainly on such characterizations of objects as particle packings, contact forces, load transfer through the assemblies etc. (Bazant *et al.*, 1990; Cundall and Strack, 1979; Kawai, 1980; Zubelewicz and Mroz, 1983). The "geomechanical" approach assumes particles interact by means of cohesionless contact forces.

The second approach deals with granular composites. Here, the interaction between particles is not direct. It takes place through the matrix layers that divide particles. Such systems are usually regarded as continuous at microlevel and homogeneous at macrolevel. That is why, possibly, principles of continuum mechanics are more often applied to solving structural properties problems for these materials (Christensen, 1979; Shermergor, 1977).

Both trends, reflecting different industrial interests, seem to progress rather independently in spite of the existence of rather deep structural similarity of the objects investigated. We think that their joining might be fruitful in the field of the structural properties investigations. This paper is an attempt to apply the merits of cohesionless discrete geomechanics to the continuous media composite problems. We shall describe a new method of relating micro- and macroelastic parameters for highly filled granular composites characterized by the high mechanical non-homogeneity of constituent elements (particles are much stiffer than matrices). This approach will be referred to as the physical discretization method (PDM).

2. BASIC IDEAS OF PHYSICAL DISCRETIZATION

The idea of the method can be clearly seen when one considers the interaction of two closely spaced solid spheres embedded in the infinite soft elastic matrix. The spheres are perfectly bonded to the matrix. Equal opposite forces are applied to the centres of the spheres along the centre lines (Fig. 1).

Let us analyse the peculiarities of the stress state in the matrix surrounding spheres. An available solution of this problem (Chen and Acrivos, 1978; Svistkov, 1982) shows that the elastic energy stored in the matrix layer between the spheres is many times greater than that in the rest of the surrounding matrix. This matrix layer can be regarded as the elastic bridge or spring transferring the interaction force between the spheres. On these grounds we may attempt to substitute the field interaction between two spheres by the equivalent linear interaction brought about through a phantom elastic rod element. The realistic composite structure may, then, be represented as a network of such connected elastic rod elements. They may differ both in length and elastic stiffness to reflect the random nature of composite structure. Such a "framework" description seems to be much simpler for mathematical treatment than the original field representation.

To find an equivalent pass from the field to the rod element construction, the solution of the boundary problem shown in Fig. 1 is obviously needed. This solution has been obtained earlier on the basis of an analytical approach and has been reported in (Garishin, 1988).

The stiffness of the equivalent rod was calculated as follows. At first the displacement dL of the sphere centres under the forces F_l and the energy W stored in the realistic continuous system was determined. Then this energy was equated with that of the elastic rod element having the length L equal to the distance between the centres of the spheres and stretched to the displacement dL. The magnitude of the stiffness G_l could then be represented as

$$G_l = 2WL/dL.$$

Knowing the stiffness of the rod element we are able to calculate the magnitude of the forces F_l under any imposed displacement dL of the rod ends

$$F_l = G_l\,dL/L.$$

Approximate estimates have shown that only the longitudinal stiffness may be taken into account while cross-bending and twisting stiffnesses can be neglected when the spheres are spaced close to each other.

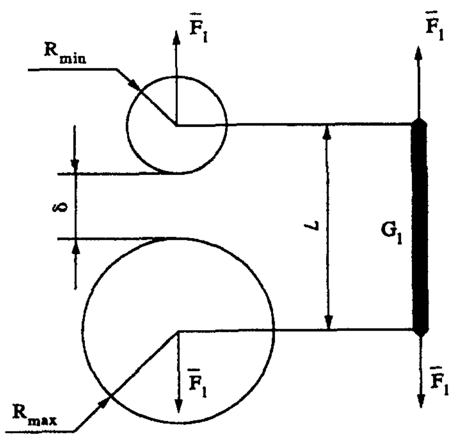

Fig. 1. Calculation scheme for the definition of rod element elastic characteristics.

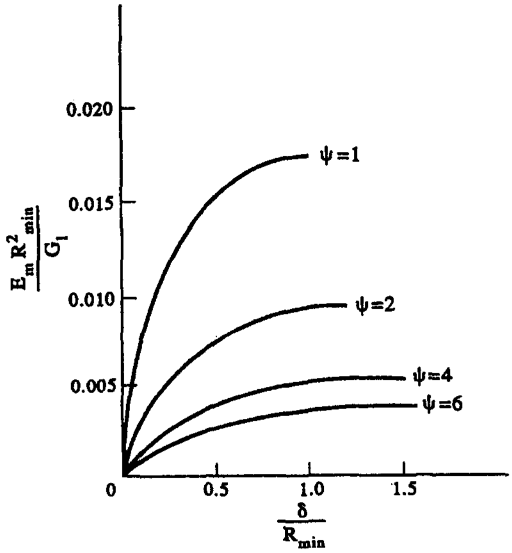

Fig. 2. The dependence of non-scale rod compliance ($E_m R_{min}^2/G_l$) on the relative spacing between the spheres δ/R_{min} and the ratio of their sizes $\psi = R_{max}/R_{min}$.

Having fulfilled a series of calculations for various imposed conditions (sizes of the spheres, spacings between them, matrix moduli) the dependence of non-scale rod compliance ($E_m R_{min}^2/G_l$) on these parameters has been obtained. The results are depicted in Fig. 2, where G_l is the stiffness of the equivalent rod; E_m is the Young's modulus of matrix; R_{min} is the radius of the minor sphere; ψ is the ratio of the radius of the larger sphere R_{max} to R_{min}; δ is the distance between the spheres. It is seen clearly that rod compliance (and stiffness G_l accordingly) are highly non-linear functions. The uniformly sized spheres turn out to be sensitive to each other when spacing between them becomes less than their radii. A very steep rise in stiffness is observed at gaps less than 0.3 of the radius. An empirical approximation for the stiffnesss of the equivalent rod covering these results has been suggested:

$$G_l = E_m R_{min}^2 \psi^{0.95} (1 - \exp(-4.4\delta/R_{min}))^{-1}. \qquad (1)$$

It is obvious that besides the rod stiffnesses, the inner geometry of the composite material is to be described to enable its macroscopic mechanical evaluation. This is the next point to be regarded.

3. GENERATION OF RANDOM GRANULAR STRUCTURES

Many approaches have been developed for generating random structures of spherical particles. Most of them cannot be used for mathematical synthesis of the high density random assemblies (Volkov and Stavrov, 1978). The others cannot escape anisotropy in a generated structure (Jodrey and Tory, 1979).

We have used an algorithm that is free of these deficiencies. The basic idea of the method was proposed by Mason (1967) who suggested synthesizing such systems by simulation of radial gravitation field influence. Initially a necessary number of points are generated in a "box" space. Then these points are turned into small spheres of an imposed radius. If two spheres overlap, they are moved apart along the line of centres until they are just touching. The spheres are then increased in size by a chosen increment and the process is repeated. Such structures can reach a limiting random packing density of 0.64 which is close to the volume densities of realistic packings.

We have expanded Mason's approach into the generation of multimodal packings that can be both dense and slightly rarefied. The procedure consists first of creating dense packing of unimodal spheres. Then an imposed number of arbitrary chosen spheres continue increasing incrementally in size according to Mason's scheme (with some necessary improvements) until the prescribed size ratio is reached. The mathematical details of the generation of such systems have been described in Garishin and Chait (1983) and Garishin (1984).

Radial distribution functions have been calculated for mono- and bifractional packings and compared with experimental data reported in Oda (1977) and Scott (1962). A close conformity of these results has been observed.

Mathematically, the structures generated were characterized by the coordinates of the sphere centres and the magnitudes of their radii. Other information concerning the inner geometry of the structures could be easily calculated.

4. FRAMES ELASTICALLY EQUIVALENT TO CONTINUOUS MEDIA

To transform the continuous representation into a rod-like one it suffices to substitute the distances between the centres of the spheres for rod elements of the same length and ascribe to them the stiffnesses determined by the expression (1). It is evident that the construction obtained may be regarded as a variety of random three-dimensional frameworks consisting of elastic ball-jointed rods. The example of such a transformation for a plane granular assembly is shown in Fig. 3.

Now, we can proceed to the calculation of the macroelastic parameters of the framework. A spherical cluster containing about 600 spheres is generated. Then, a cube including 300–400 spheres and 1000–2000 rod elements is cut out of this cluster. This cube will be later referred to as a sample. The stiffness and length of rod elements are calculated.

The statement of the problem is the following (Garishin, 1989):

The prescribed uniform longitudinal displacement at one end of the sample and restraint against displacement at the opposite end are imposed. The calculation procedure is that of conventional finite element analysis. Several cross-cuts perpendicular to the elongation of the sample are made. For each cross-section the projections of rod element forces in the direction of elongation are summed to yield the total force per given cross-section. The averaged force per cross-section is then obtained. This one divided per cross-section area is identified with the stress acting in the sample. The sample strain is calculated by dividing the imposed displacement of the end of the sample by the sample length. Taking the ratio of the stress to the strain we obtain the Young's modulus E_c.

The magnitude of a representative volume has been estimated. It was shown that the number of particles needed to get a reasonable statistical significance must be taken to be not less than 300.

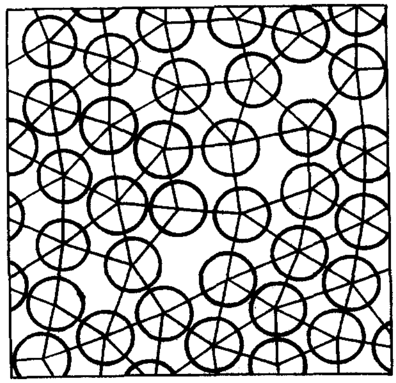

Fig. 3. An example of physical discretization of granular composite structure (plane case).

5. ANALYSIS OF THE VALIDITY OF THE PHYSICAL DISCRETIZATION METHOD

Direct comparison of this approach with any well documented theoretical solutions which, undoubtedly, were the best way of testing, is not possible because of the lack of such solutions, as far as we know.

However, some qualitative theoretical considerations concerning the application of the physical discretization method can be made. The distributions of spacings between the neighbouring spheres at various concentrations have been studied (Fig. 4). (Two particles are considered to be neighbours if the line between their centres does not cross any other sphere.) It is seen that there exist rather high concentrations above which the maximum spacings exceeding one radius may be regarded as negligible. It means that this is the range of concentrations where the interaction, in the sense of Section 2, becomes substantial, thus enabling us to use the offered approach (PDM). At lower concentrations of unimodal sphere systems the mean distances between nearest neighbours become more than one radius and the main postulate of the proposed approach about the predominant role of central line interactions ceases to be valid. The analysis of monofractional sphere random distributions for various filler loadings has revealed that at volume concentrations less than 0.3 the quantity of interparticle spacings exceeding one radius becomes considerable. In this region of concentration neglecting the stored shear and twist energy must lead to modulus underestimations. The other difficulty with low concentrations is that the notion of neighbouring sphere becomes indefinite.

As to the upper concentration limit of the PDM application, it can be defined from the consideration that at very high concentrations of spheres close to the packing density, the true stiffness of the model basic element must be substantially higher than the one calculated as indicated in Section 2 because the effective modulus of the medium surrounding the basic element in the composite material is much higher than that of the pure matrix taken into account in the above approach.

Envisaging the modulus-concentration relationship over the whole range of concentrations we conclude that the E_c predicted by the PDM results must lie somewhat below the true ones. The underestimation seems to be more pronounced at lower and higher concentrations.

Published theoretical data of this sort being absent, we see the only possibility for quantitative PDM evaluation is that of the direct comparison of our theoretical calculations with some experimental data. The following initial data were introduced for theoretical calculations: the ratio of the matrix modulus E_m to the sphere modulus E_p was taken to be equal to 10^4; both matrix and spheres were assumed to be incompressible; the radii of all

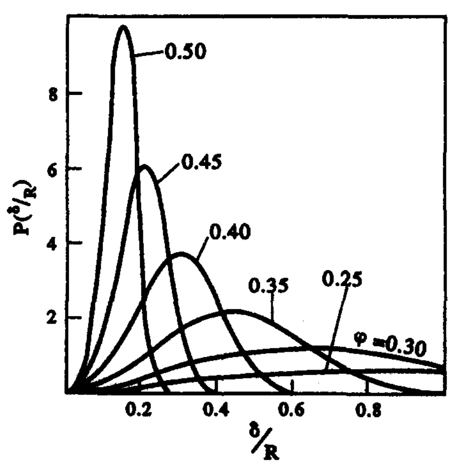

Fig. 4. The distributions of spacings in random structures of equal spheres for various concentrations of filler φ.

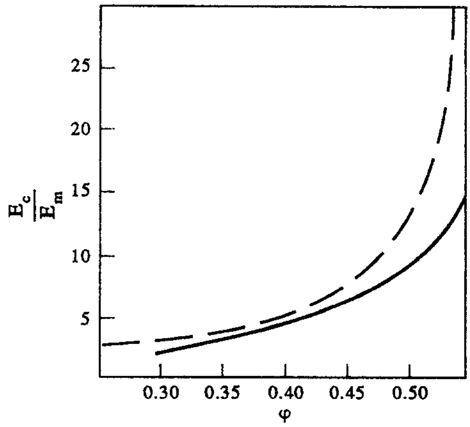

Fig. 5. The modulus–concentration relation for a monofractional granular composite. Dashed line corresponds to experimental data (Farris, 1968), solid line corresponds to the model result.

spheres were equal to $R = 1$. In Fig. 5, the well established experimental modulus–concentration curve (Farris, 1968) is compared with our prediction. A concentration range between 0.3 and 0.5 (Fig. 6) can be judged as one where the approximate theoretical results seem to be the best. Here, the underestimation error lies within 30%.

It is evident that the framework modelling cannot accurately reflect the volume compressibility or the Poisson ratio of elastic particulate composites because no provision for volume preservation is laid down into the PDM. One should bear in mind that in our case the composite is incompressible for such are the constituent elements.

Nevertheless, there is a distinct interest in calculating the Poisson ratio for random framework systems. The calculated values of the Poisson ratio for several concentrations are as follows:

Volume concentration	0.40	0.50	0.60
Poisson ratio	0.249	0.255	0.259

One may conclude that the value of the Poisson ratio slightly increases with the concentration growth in the range studied.

6. SOME STRUCTURAL PROPERTIES RELATIONSHIPS INVESTIGATED BY MEANS OF THE MODEL

When filler loadings more than 0.6 by volume are required two or more dispersed structures are usually used. Two fraction filler loadings were investigated. The following initial data were introduced for these theoretical calculations: the ratio of matrix modulus E_m to sphere modulus E_p was taken to be equal to 10^4; both matrix and spheres were

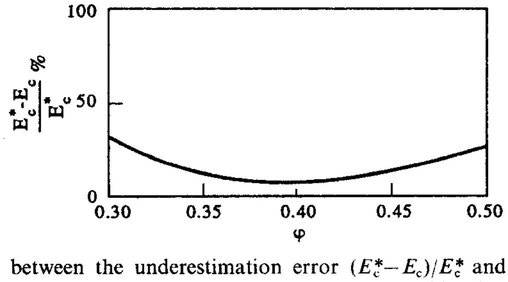

Fig. 6. The relation between the underestimation error $(E_c^* - E_c)/E_c^*$ and concentration φ for experimental (E_c^*) and model (E_c) data, depicted in Fig. 4.

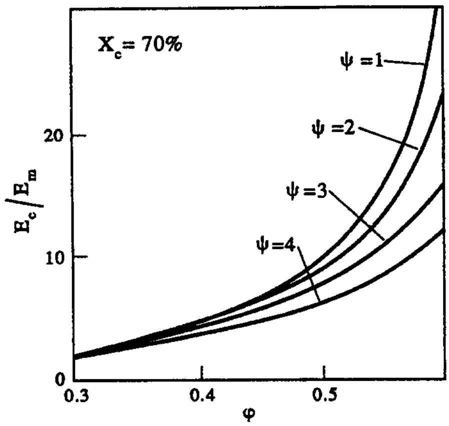

Fig. 7. The modulus–concentration relation for bifractional granular composites with the same volume of coarse particles $X_c = 70\%$ and various ratios of particle radii $\psi = 1, \ldots, 4$.

assumed to be incompressible; the spheres' radii ratio ψ (larger to minor) was varied from 1 to 4.

Figure 7 illustrates the modulus-concentration behaviour when the larger particles volume portion X_c is equal to 0.7. In Fig. 8, the curves of E_c versus X_c for various total filler loadings φ are plotted on a graph (the particle size ratio ψ is equal to 3). This numerical experience verifies the existence of the minimum E_c known from practice at certain fractions of large particles that are close to 0.7. This minimum does not vary much with filler loading. At lower concentrations it is emphasized only slightly. These calculations rather well approximate experimental results (Chong *et al.*, 1971).

The above analysis shows that theoretical prediction of the effective moduli based on two-particle approximation and knowledge of the internal constitution of composite materials appears to be realistic. This agreement substantiates the basic concepts of the physical discretization approach which provides simple and effective means of calculation of local structural forces and studying their dependence on constitutional features.

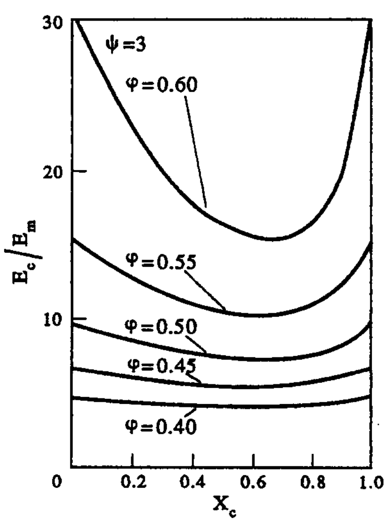

Fig. 8. The relation between bifractional and composite modulus E_c and coarse particle volume portion X_c for various concentrations φ, when size ratios of coarse and fine particles ψ equals 3.

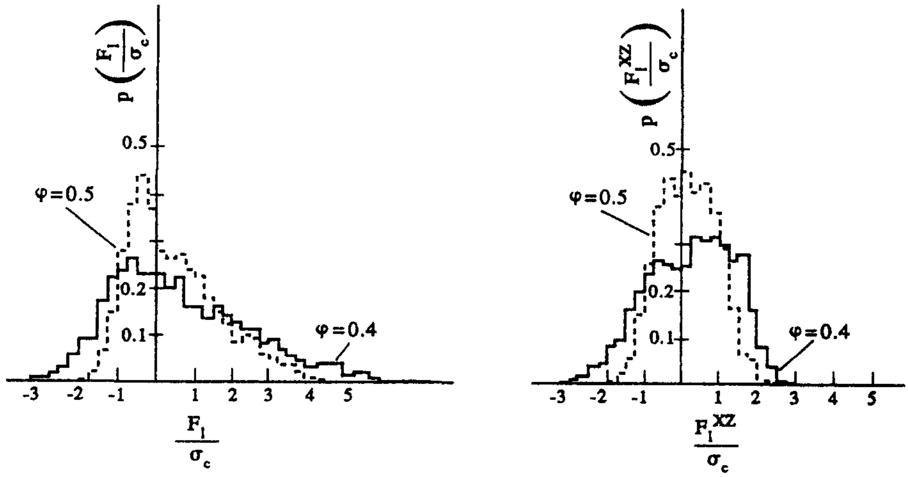

Fig. 9. The distributions of forces in rod elements E_l (a), and its projections perpendicular to the elongation cross plane F_l^{xz} (b) for various concentrations of filler. (i) $\varphi = 0.4$—solid line, (ii) $\varphi = 0.5$—dashed line.

Some new statistical structural characteristics and relationships that could not be obtained otherwise are reported below. For instance, valuable conclusions on the distribution of forces in rod elements under macrohomogeneous extension of the composite sample can be obtained. The samples containing 0.4 and 0.5 by volume spheres of one size were investigated. Figure 9 shows the distributions of forces in rod elements (F_l) and its projections perpendicular to the elongation cross plane (F_l^{xz}). The horizontal axis represents the values of the forces divided by the magnitude of macrostress σ_c and the vertical axis their probability densities. The distribution is very wide reflecting a great non-homogeneity of local stresses. Moreover, it turned out that about 40% of rod elements are not only stretched but compressed. The analysis has shown that in general those rod elements are compressed whose inclination to the cross-section planes is less than 45°. The increase in filler concentration is accompanied by the narrowing of the distribution of both the extensive and compressive maximum forces.

These results demonstrate clearly that only a minority of rod elements play an active role in the current internal elastic resistance.

The most stretched rod elements are considered to be the first candidates for failure. For that reason the structural elastic approach may be regarded as a basis for investigation of subsequent damage initiation and accumulation. This point will be the objective of our next report.

7. CONCLUSIONS

The physical discretization method (PDM) has been suggested for developing the structural model of highly filled strongly inhomogeneous particular composites. The approach is based on the two-particle approximation and applied to random composite systems.

It is oriented to the evaluation of an elastic modulus of composite materials as a function of constituent element properties, filler volume loading and particle size distributions.

The method proved to be accurate in the range of particle concentrations from 0.3 to 0.5.

It can be used for studying the structural errors in loaded materials as the point of departure in the investigation of damage origination and evolution.

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