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INHOMOGENEOUS INTERFACIAL TRANSITION ZONE MODEL FOR THE BULK MODULUS OF MORTAR

Melanie P. Lutz[1], Paulo J.M. Monteiro[1], and Robert W. Zimmerman[2]

[1]Department of Civil and Environmental Engineering
University of California, Berkeley, CA 94720, USA
[2]Department of Earth Resources Engineering
Imperial College of Science, Technology and Medicine, London SW7 2BP, UK

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ABSTRACT

The macroscopic bulk modulus of mortar and concrete is modeled by assuming that each inclusion (fine or coarse aggregate) is spherical, and is surrounded by an interfacial transition zone (ITZ) in which the elastic moduli vary smoothly as a power-law function of radial distance from the center of the inclusion. The exponent in the power law can be chosen based on the estimated thickness of the ITZ, or by fitting the power law to measured porosity profiles. For this model, an analytical expression has been found by Lutz and Zimmerman (J. Appl. Mech., 1996) for the macroscopic bulk modulus. The macroscopic bulk modulus depends on known properties such as the elastic moduli of the bulk cement paste and the inclusions, the volume fraction of the inclusions, the elastic moduli at the interface between the cement paste and inclusion, and the thickness of the ITZ. In this paper the inhomogeneous ITZ model is used to analyze the data of Wang et al. (Cem. Conc. Res., 1988) on the bulk modulus of mortar containing sand inclusions. By fitting the measured moduli to the model predictions, we can estimate, in a non-destructive manner, the elastic moduli within the ITZ. For Wang's specimens, it is inferred that the elastic moduli at the interface with the inclusions is 30-50% less than in the bulk cement paste. © *1997 Elsevier Science Ltd*

Introduction

The earliest and simplest models of the elastic moduli of cementitious materials were based on the assumption that concrete (or mortar) consists of two phases: aggregate (or sand) particles and cement paste. Under this assumption, simple mixing rules, such as a volume-average of the stiffnesses (Voigt model) or a volume-average of the compliances (Reuss model), can be used to estimate the effective elastic moduli. More sophisticated models can be used that account for the fact that the cement paste is the connected phase, whereas the inclusions form a disconnected, dispersed phase. Various mathematical theories have been proposed to predict the effective elastic moduli of this type of particulate composite. Zimmerman et al. (1) used the Kuster-Toksöz theory (2) to study the effect of sand inclusion concentration on the effective moduli of mortar. Yang and Huang (3) used the Mori-Tanaka theory (4) to account for the presence of both sand and aggregate inclusions, each having their own elastic moduli. A review of two-component models for the macroscopic elastic moduli of concrete and mortar has been given by Mehta and Monteiro (5).

Recently, however, it has become recognized that the cement paste should not be considered to be a homogeneous phase. It is now known that the structure of the cement paste in the vicinity of the inclusions differs from that of bulk cement paste (5,6). The region in which the presence of the inclusions affects the properties of the cement paste is known as the Interfacial Transition Zone (ITZ). In this zone the porosity is greatest near the inclusions, and decreases with increasing distance from the inclusion (7). Neubauer et al. (8) and Ramesh et al. (9) have developed models in which the ITZ is represented by a thin, shell-like region that surrounds each inclusion. These three-shell (inclusion/ITZ/cement paste) models should be more realistic than two-component models, but still represent an oversimplification, in that they assume that the elastic moduli are uniform within the ITZ. Lutz et al. (10) proposed a model in which the elastic moduli in the cement paste decay according to a power law function of the distance from the center of the inclusion. An analytical solution for the effective bulk modulus of this inhomogeneous ITZ model has been derived by Lutz and Zimmerman (11). In the present paper we use this inhomogeneous ITZ model to analyze measurements of elastic moduli made by Wang et al. (12) on a suite of water-saturated mortar specimens that contained varying volume fractions of sand inclusions.

Model of the Transition Zone

Lutz et al. (10) proposed the following conceptual model of concrete or mortar (Fig. 1). The aggregate particles are assumed to be spherical, with radius a. The elastic moduli $\{K_{in}$, G_{in} within the inclusions are assumed to be uniform. Outside of each inclusion, the elastic moduli are assumed to vary smoothly with radius, according to the following power law equations:

$$K(r) = K_{cp} + (K_{if} - K_{cp})(r/a)^{-\beta}, \tag{1}$$

$$G(r) = G_{cp} + (G_{if} = G_{cp})(r/a)^{-\beta} \tag{2}$$

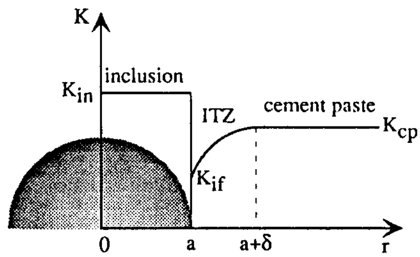

FIG. 1.
Schematic modulus profile around an aggregate particle in concrete (from Eqs. 1, 2).

where the subscript *cp* refers to the bulk cement paste, and the subscript *if* refers to the interface with the inclusion. The moduli $\{K_{cp}, G_{cp}\}$ are those of the cement paste far from the inclusion, and reflect the effect of any pores that are present; they are not the elastic moduli of a hypothetical non-porous cement paste. Furthermore, if the pores of the cement paste are saturated with water, $\{K_{cp}, G_{cp}\}$ also reflect that fact; the effect of pores, and pore saturants, on the elastic moduli of the cement paste has been studied by Zimmerman et al. (1) using the Kuster-Toksöz model. For our present purposes, we treat the water-saturated, porous cement paste as a single, macroscopically homogeneous phase.

Lutz and Zimmerman (11) found an exact solution to the problem of hydrostatic compression of a cement paste containing a single inclusion surrounded by an inhomogeneous ITZ, and then used this solution to estimate the effective bulk modulus of a material that contains a random dispersion of such inclusions. The results of their analysis coincide with the predictions of the Mori-Tanaka (4) and Kuster-Toksöz (2) theories in the limiting case when the ITZ is absent. In order for their analytical solution to be applicable, the power-law exponent β must be an integer; this poses no limitation on the practical applicability of the model, as will be seen below.

If the elastic moduli profile within the ITZ were known, the parameter β could be found by performing a least-squares fit of the profiles to the functions (1, 2). In the absence of such detailed elastic moduli profiles, Lutz et al. (10) showed that if the "thickness" of the ITZ is estimated based on visual observation of SEM micrographs, β could be found from the equation

$$\beta \approx 2.303a/\delta. \tag{3}$$

This relation can be obtained from Eqs. 1 and 2 by assuming that the visually-estimated outer edge of the ITZ corresponds to the point at which the power-law term has decayed to 10% of the value that it has at the interface with the inclusion. As β must be an integer, the predictions of eq. (3) should be rounded off to the nearest integer.

Although no measurements of moduli profiles within the ITZ are yet available, porosity profiles have been measured (7,13). The porosity profiles should be closely related to the moduli profiles, because the increase in porosity near the inclusions is one of the main factors that leads to a local degradation of the elastic moduli. To develop a relationship between the increase in porosity and the decrease in moduli, we consider the effect of adding an additional incremental amount of porosity to the "bulk" cement paste. If we let ϕ_{cp} denote the porosity of the bulk cement paste, and let $\Delta\phi(r)$ denote the increment in porosity at some radial distance r, the effective elastic moduli at r are given by (14)

$$K(r) \approx K_{cp}\left[1-\alpha_K \Delta\phi(r)\right] = K_{cp}\left[1-\alpha_K\left\{\phi(r)-\phi_{cp}\right\}\right] \tag{4}$$

$$G(r) \approx G_{cp}\left[1-\alpha_G \Delta\phi(r)\right] = G_{cp}\left[1-\alpha_G\left\{\phi(r)-\phi_{cp}\right\}\right] \tag{5}$$

The coefficients α_K and α_G depend primarily on pore shape, but also, to a lesser extent, on the Poisson ratio of the cement paste, ν_{cp}. For the two "extreme" cases of spherical and crack-like voids, these coefficients are (5 and15) spherical pores: $$\alpha_K = \frac{3(1-\nu_{cp})}{2(1-2\nu_{cp})}, \qquad \alpha_G = \frac{15(1-\nu_{cp})}{(7-5\nu_{cp})}; \tag{6}$$

cracks: $$\alpha_K = \frac{4(1-\nu_{cp}^2)}{3\pi(1-2\nu_{cp})\varepsilon}, \qquad \alpha_G = \frac{8(1-\nu_{cp})(5-\nu_{cp})}{15\pi(2-\nu_{cp})}; \tag{7}$$

where $\varepsilon \ll 1$ is the aspect ratio of a typical crack in the ITZ. For the range of Poisson ratios commonly observed in cement paste, $0.15 < \nu_{cp} < 0.30$, these coefficients are nearly independent of ν_{cp}. For a typical value such as $\nu_{cp} = 0.2$, the coefficients are

spherical pores: $$\alpha_K = 2.0, \qquad \alpha_G = 2.0; \tag{8}$$

cracks: $$\alpha_K = 0.679/\varepsilon, \qquad \alpha_G = 0.362/\varepsilon. \tag{9}$$

Spherical pores therefore cause proportionately the same decrement in the bulk modulus as in the shear modulus, whereas crack-like voids decrease K to a much greater extent than G.

According to Eq. 4 and 5, the incremental moduli profiles (i.e., $K(r) - K_{cp}$) should be proportional to the incremental porosity profile. Hence, the accuracy with which the moduli profiles can be fit with power law functions can be estimated from the extent to which porosity profiles can be fit with power law functions of the form

$$\phi(r) = \phi_{cp} + (\phi_{if} - \phi_{cp})(r/a)^{-\beta} \tag{10}$$

Fig. 2 shows the porosity profile around an aggregate particle in a concrete having a water/cement ratio of 0.4, after one year of hydration, as measured by Crumbie (13) using backscattered electron imaging. Although the porosity profile within the ITZ depends on factors such as the water/cement ratio and the chemical composition of the cement paste, it

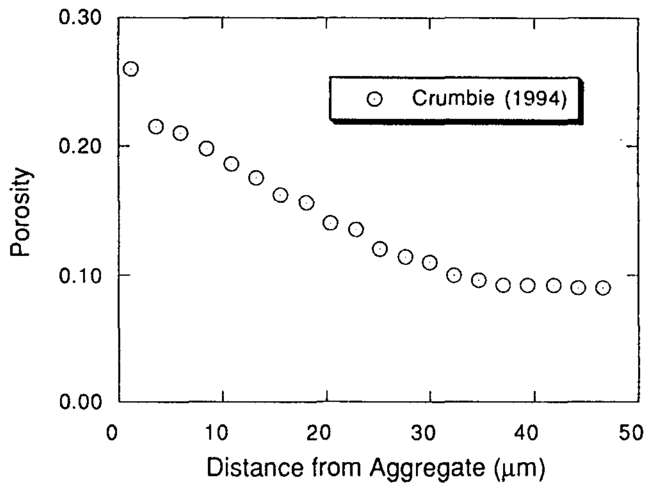

FIG. 2.
Porosity profile around an aggregate particle in concrete (after Ref. 13).

seems to be independent of the size of the inclusions (16,17). Hence, it seems plausible to assume that the profile measured by Crumbie is at least qualitatively representative of typical porosity profiles within the ITZ.

As the variable in the power law function is the distance from the center of the inclusion, rather than the distance from the outer edge of the inclusion, we cannot perform the curve-fit without assuming a value for the inclusion size. In order to span a large range of possible sizes, Lutz et al. (10) considered inclusions having radii of 50 μm and 500 μm, each surrounded by an ITZ having the same porosity profile as measured by Crumbie (13). Although the optimal value of β depended strongly on the inclusion radius, in both cases a power law provided a very good fit to the porosity data. They also found that the optimal values of β found from the curve-fitting procedure were in very close agreement with the values obtained from Eq. 3, if the "thickness" of the ITZ is taken to be about 40 μm, which seems reasonable (see Fig. 2). Our conclusion is that it seems reasonable to represent the porosity profiles, and hence the elastic moduli profiles, by power-law functions of the radius.

Insertion of the porosity profile (10) into Eqs. 4 and 5 yields moduli profiles of the form

$$K(r) = K_{cp}\left[1 - \alpha_K\left(\phi_{if} - \phi_{cp}\right)(r/\alpha)^{-\beta}\right] \tag{11}$$

$$G(r) = G_{cp}\left[1 - \alpha_G\left(\phi_{if} - \phi_{cp}\right)(r/\alpha)^{-\beta}\right] \tag{12}$$

Comparison of Eqs. 11 and 12 with Eqs. 1 and 2 shows that the product $\alpha_K(\phi_{if} - \phi_{cp})$ is equal to the fractional difference between the bulk modulus at the interface and in the cement paste, and similarly for the shear modulus. We will refer to these fractions as the local damage parameters, $D_K = (K_{cp} - K_{if})/K_{cp}$, etc., and write Eqs. 11 and 12 as

$$K(r) = K_{cp}\left[1 - D_K(r/a)^{-\beta}\right] \tag{13}$$

$$G(r) = G_{cp}\left[1 - D_G(r/a)^{-\beta}\right] \tag{14}$$

The elastic moduli at the interface with the inclusion can therefore be expressed by, $K_{if} = K_{cp}[1 - D_k]$, and $G_{if} = G_{cp}[1 - D_G]$.

Analytical Solution for the Effective Bulk Modulus

The analytical solution developed in (10) for the effective bulk modulus of a material containing spherical inclusions surrounded by a power-law-type interfacial transition zone described by Eqs. 1 and 2 can be written as

$$\frac{K_{eff}}{K_{cp}} = \frac{1 + \left(4G_{cp}/3K_{cp}\right)fc}{1 - fc}, \tag{15}$$

where
$$f = \frac{3\left(K_{in} - K_{if}\right)\sum_{n=0}^{\infty} \Gamma_{n\beta} + \left[K_{if} + (4/3)G_{if}\right]\sum_{n=0}^{\infty} n\beta\Gamma_{n\beta}}{3\left(K_{in} - K_{if}\right)\sum_{n=0}^{\infty} \Gamma_{n\beta+3} + \left[K_{if} + (4/3)G_{if}\right]\sum_{n=0}^{\infty} (n\beta+3)\Gamma_{n\beta+3}}. \quad (16)$$

and c is the volume fraction of the inclusions. The coefficients Γ are found from the following recursion relation:

$$\Gamma_{n+\beta} = \frac{-\left\{\left(K_{if} - K_{cp}\right)\left[n^2 + (\beta-3)n - 3\beta\right] + (4/3)\left(G_{if} - G_{cp}\right)\left[n^2 + (\beta-3)n\right]\right\}}{(n+\beta)(n+\beta-3)\left[K_{cp} + (4/3)G_{cp}\right]}\Gamma_n, \quad (17)$$

with $\Gamma_0 = \Gamma_3 = 1$. From Γ_0 and Γ_3, the recursion relation (17) generates $\{\Gamma_\beta, \Gamma_{2\beta}, \ldots\}$ and, $\{\Gamma_{3+\beta}, \Gamma_{3+2\beta}, \ldots\}$, which are the only Γ coefficients that actually appear in Eq. 16. In the limiting case where the cement paste is homogeneous, all the coefficients except Γ_0 and Γ_3 vanish, and $K_{if} \to K_{cp}$, etc., in which case $f \to 3(K_{in} - K_{cp}) / (3K_{in} + 4G_{cp})$ and Eq. 15 reduces to the result found by Mori and Tanaka (4), Kuster and Toksöz (2) and others for the effective bulk modulus of a material composed of spherical inclusions in a *homogeneous* matrix. This result also coincides with the Hashin-Shtrikman lower bound (18). As shown in (11), as K_{if} decreases below K_{cp}, the effective bulk modulus falls below the Hashin-Shtrikman lower bound for a two-component material.

Application of Model to Experimental Data

Wang et al. (12) measured compressional and shear velocities on a suite of water-saturated mortar specimens that contained varying amounts of sand inclusions. The bulk moduli of their specimens can be found from the two measured wavespeeds using the relationship

$$K_{eff} = \rho_{eff}\left(V_p^2 - \frac{4}{3}V_s^2\right), \quad (18)$$

where the effective density is related to the densities of the cement paste and sand inclusions by

$$\rho_{eff} = (1-c)\rho_{cp} + c\rho_{in}. \quad (19)$$

The mortar consisted of cement paste with sand inclusions, the diameters of which ranged from 590–840 μm; hence, the mean inclusion radius was about 350 μm.

To apply our inhomogeneous ITZ model to the data of Wang et al. (12), we first assume that the porosity profiles in the ITZ are similar in shape and extent to those measured by Crumbie (13), although the specific values of ϕ_{if} and ϕ_{cp} may be different. As shown in Eqs. 11 and 12, if the porosity profile can be fit with a power-law function, the same should be true for the elastic moduli profiles. Fig. 3 shows the porosity profile measured by Crumbie, fit to an equation of the form of (10), using an inclusion radius of 350 μm, as is appropriate to the specimens under consideration. The optimum (integral) value of β is 22, with a correlation coefficient of $r^2 = 0.978$. Values of $\beta = 21$ or $\beta = 23$ changed the correlation coeffi-

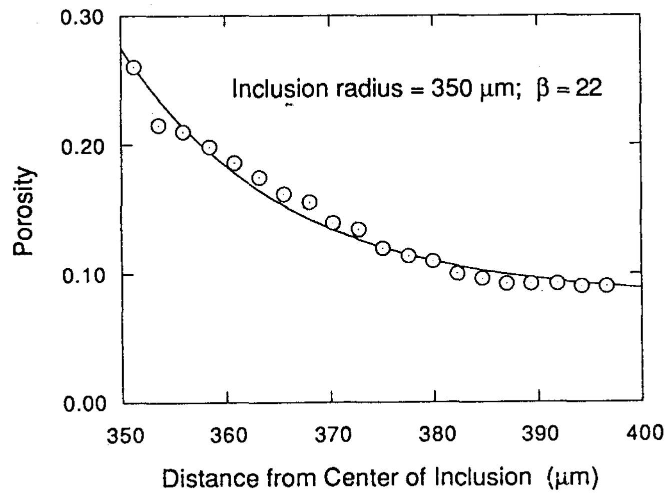

FIG. 3.
Porosity profile from Fig. 2, fit to equation (10), assuming a = 350 μm.

cient by less than 0.5%. This substantiates our earlier claim that no generality is lost by forcing β to be an integer. It is also worth noting that the curve-fir method yields a results that is consistent with that obtained from Eq. 3. If $\beta = 22$ and a = 350 μm, Eq. 3 yields and ITZ thickness of 37 μm, which seems visually consistent with Fig. 2.

Table 1 shows the parameters we used in applying our model to this data set. The properties of the bulk cement paste, $\{K_{cp}, G_{cp}, \rho_{cp}\}$, are taken from the specimen that contained no sand inclusions; the properties of the sand inclusions, $\{K_{in}, G_{in}, \rho_{in}\}$, are taken from (2). The parameters E and ν are not explicitly needed as inputs, but are listed in Table 1 for completeness; they can be calculated from the relationships

$$E = 9KG/(3K+G), \qquad \nu = (3K-2G)/(6K+2G). \tag{20}$$

The only additional parameters that are needed are the two local damage parameters. Eqs. 11-14 show that calculation of these parameters would require knowledge of the local porosity profile, and knowledge of the pore shapes within the ITZ. However, our intention is to use the model in the inverse sense, to infer the local damage parameters from the measured macroscopic bulk moduli. Lacking detailed knowledge of the pore morphology in the ITZ, we assume that the pores are nearly spherical, in which case Eq. 10 indicates that $D_G = D_K$. This is in agreement with the findings of Scrivener and Nemati (16), who observed no crack-like voids in the ITZ. Furthermore, numerical evaluation of Eqs. 15-17 indicates

TABLE 1

Physical Properties of Mortar Specimens (from Ref. 12)

Material	K (GPa)	G (GPa)	E (GPa)	ν	ρ (kg/m^3)
Sand Inclusions	44.0	37.0	86.7	0.17	2707
Cement Paste (saturated)	22.5	11.8	30.1	0.28	2182
Interface	22.5(1-D)	11.8(1-D)	30.1(1-D)	0.28	-

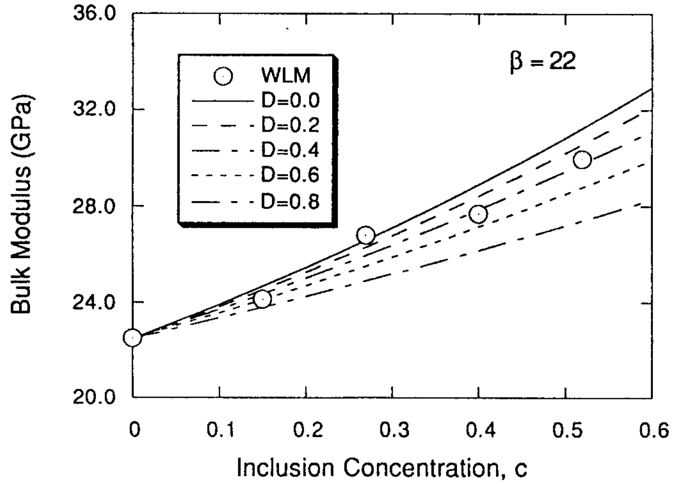

FIG. 4.
Bulk moduli measured by Wang et al. (12) using acoustic methods, compared to predictions of inhomogeneous ITZ model, for various values of the local damage parameter at the interface.

that K_{eff} is much more sensitive to D_K than to D_G, as might be expected. Hence, it seems safe to put $D_G = D_K \equiv D$, in which case K_{eff} will be a function of the local damage parameter, D, and the volume fraction of inclusions, c. We therefore calculate the $K_{eff}(c)$ curves for different values of D, and compare them to the values found from the acoustic velocity measurements; the best fit will yield an estimate of the local damage parameter at the interface.

The measured bulk moduli of the six specimens tested by Wang et al. (12) are shown in Fig. 4, along with the predictions of the model, for various values of D. Each datum point is fit by a slightly different value of D, ranging from 0.0–0.6. The single value that provides the best fit to the entire data set is about 0.4. If the excess porosity in the ITZ were indeed in the form of spherical pores, Eqs. 8 and 11 indicate that a damage parameter of 0.4 corresponds to an excess porosity at the interface of 0.20; this agrees very closely with the value 0.18 measured by Crumbie (13). This agreement lends further credibility to our assumption that the effective bulk modulus can be explained in terms of the inhomogeneous ITZ model. However, we must emphasize that the application of our model is in no way dependent upon the use of Crumbie's porosity profile. Regardless of any assumption about the nature of the porosity profile within the ITZ, our comparison of the measured data with the model predictions leads to the conclusion that the moduli at the interface are about 40% lower than in the bulk cement paste.

The curve for $D = 0.0$ coincides with the theoretical lower bound derived by Hashin and Shtrikman (18) for a material consisting of two homogeneous components. The fact that measured moduli typically fall below the lower bound demonstrates the inability of a two-component model to account for the observed macroscopic bulk moduli. The same conclusion was reached by Nilsen and Monteiro (19) after analyzing the data from (1). Our inhomogeneous ITZ model is capable of providing a rational explanation of the fact that the numerical values of the measured bulk moduli do not satisfy the two-component Hashin-Shtrikman bounds.

Summary and Discussion

A conceptual model of concrete has been proposed in which the aggregate particles are treated as spheres surrounded by a radially-inhomogeneous cement paste. The two elastic moduli in the cement paste are each represented by a constant term plus a term that varies with radius according to a power law. The exponent in the power law can be determined either using a visually-estimated "thickness" of the interfacial transition zone, or by fitting a power law to measured porosity profiles. The effective bulk modulus is then found using the exact solution recently derived by Lutz and Zimmerman (11). The only open parameter in the model is the local damage parameter, which represents the extent to which the elastic moduli at the interface are less than in the bulk cement paste. By matching the model predictions to experimental data on mortar specimens containing varying amounts of sand inclusions, we were able to estimate the bulk moduli at the interface. The estimated damage parameters for a set of saturated mortar samples (12), which were about 0.4, were consistent with the assumption that the damage was due to excess quasi-spherical porosity in the ITZ. This model offers an explanation of why some previously-measured moduli fall outside of the Hashin-Shtrikman bounds for two-component materials, and also offers a simple, non-destructive method of estimating the local elastic properties within the interfacial transition zone.

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