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EFFECT OF TRANSITION ZONE ON THE ELASTIC BEHAVIOR OF CEMENT-BASED COMPOSITES

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ABSTRACT

The elastic behavior of mortar and concrete as a composite material and the influence of the transition zone on the elastic modulus is studied. The theoretical bounds of Hashin-Shtrikman for the modulus of elasticity of two-phase composite material are used as a criteria in the analysis. Available experimental results with variable W/C ratio, aggregate volume and variable E-modulus of the matrix and the aggregate are considered. A notable influence of the transition zone on the elastic behavior of the composite is detected. It is demonstrated as a divergence of the experimental data from the general trend of the theoretical bounds with the increase of the aggregate volume. The influence of the transition zone on the overall elastic properties of the composite is strongly related to the water content and with the reduction of W/C ratio it changes from negative into positive.

Introduction

Macroscopically the deformation behavior of concrete is strongly related to the properties and the volume of the matrix (cement stone or mortar) and the aggregate. Many two-phase composite models have been developed to predict elastic deformation of concrete. However they cannot adequately predict the experimental results. One of the possible reasons for this could be the properties of the interface or the transition zone (TZ), the influence of which on the elastic modulus has not received adequate attention.

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The simplest two-phase composite models are the parallel - Voigt [1] and the series - Reuss [2], which assume respectively equal strain and equal stress in the two phases. It was demonstrated by Hill [3], that these models are the upper and the lower bounds for the effective modulus of the two phase composite. Different combinations of the upper and the lower bonds have been proposed for predicting the effective modulus of concrete as a composite material [4,5,6]. For statistically isotropic and homogeneous material, Hashin and Shtrikman [7] derived bounds (HS), which are the best (the closest) bounds for estimating the effective modulus of elasticity of a two-phase composite [8]. The distance between them is strongly dependant on the moduli ratio of the two phases. When the ratio of the two moduli is close to 1, HS bounds can successfully be used for predicting the effective modulus, while at higher ratios they do not have prognostic value. Nevertheless they can be used as a good test for a two-phase composite elastic behavior.

A number of experimental studies confirm the validity of HS bounds [8] for different composite materials. Mandel and Dantu [9] experimentally verified the validity of the HS bounds for cement concretes and mortars. They showed that their results lie below the lower HS bound. Watt and O'Connell [10] pointed out that low results in such investigations can be related to the non compensation of porosity in the specimens. They stated that it is also necessary to consider the possible anisotropy of the specimens and the experimental error.

It was pointed out by Monteiro [11] that the models of Yoan and Guo [12] and Mindess [13] which apply the Hirsch-model for description of the interface layer may not be theoretically correct. For the assessment of the influence of the interface layer, Monterio proposed that HS bounds should be used. He suggested that an experimental check should be made to see if the values of E-modulus of concrete as a function of the aggregate volume are inside HS bounds. Monterio stated that "if the results are outside these bounds that will mean the influence of the interface layer is significant and will show the need of three-phase models".

Baalbaki, Aitchin and Ballivy [14] studied the influence of different aggregates on the E-modulus of high-strength concrete and comparing their experimental results with different two-phase model values, concluded that high-strength concrete can not be modeled as a two-phase material. Nilsen and Monteiro [15] analyzed Hirsch's data by calculating HS bounds and concluded that concrete cannot be considered a two phase material and the transition zone (interface layer) should be included as a third phase.

In this paper the available experimental data is analyzed with the help of HS bounds and the results indicate that there is a notable effect of TZ on the elastic properties of concrete.

Description of Experimental data and Calculation of HS Bounds

Numerous papers can be found in the literature on experimental investigations with cement concretes and mortars which contain data for the modulus of elasticity of the aggregate (Ea) and the matrix (Em) (paste or mortar) and the modulus of elasticity of the composite (concrete or mortar) (Ec) for different volume fractions of the aggregate (Va).

Table 1 summarizes the available data which were considered to have adequate information for the purpose of the current analysis. The corresponding values for the lower and upper HS bounds computed for each set of data are also shown in the table.

Hirsch [4] investigated the influence of Ea and Va on Ec. Cement paste with W/C=0.4 was used as a matrix. Six different kinds of aggregates: steel, sand, crushed glass, gravel, crushed limestone and lead were used. Va was varied between 0.2 and 0.57. Secant and dynamic moduli were measured after 3, 7, 14, 28 and 60 days. Only the data for secant modulus after 28 days are used here. Mandel and Dantu [9] studied the influence of Ea, Va and Em on Ec. Different cement mortars were used as a matrix and the aggregates were diorite and glass. Counto [5] studied the influence of Ea and Va on Ec and the creep of concrete. Mortar was used as matrix and the aggregates were: steel, gravel and glass. Anson and Newman [16] investigated the influence of Em and Va on the Poison's coefficient of mortars and concretes. W/C was varied from 0.3 to 0.6. The volume of the aggregates, quartz sand and gravel, was changed in a wide range - from 0.322 to 0.725. The elastic moduli were evaluated at 0.22-0.27 of the compressive strength after several loadings and unloadings of the specimens.

Stock, Hannant and Williams [17] investigated the influence of Va (0.2-0.8) on the mechanical properties of concrete. In order to obtain constant W/C of the matrix, the absorption of the aggregates was tested and was considered in the preparation of the specimens. A special technique was used for obtaining a homogeneous distribution of the aggregates in the volume of the specimens. Ec was tested in compression (at 0.33fc) and in tension (at 0.5ft). The authors reported standard deviation and coefficient of variation for each set of experiments.

Calculation of HS Bounds

HS bounds are calculated for every experimental result according the following equations:

$$K_m=E_m/(3(1-n_m)) \quad (1)$$
$$G_m=E_m/(2(1+n_m)) \quad (2)$$
$$K_a=E_a/(3(1-n_a)) \quad (3)$$
$$G_a=E_a/(2(1+n_a)) \quad (4)$$
$$Kc_{(-)}=K_m+V_a/(1/(K_a-K_m)+3V_m/(3K_m+4G_m)) \quad (5)$$
$$Kc_{(+)}=K_a+V_m/(1/(K_m-K_a)+3V_a/(3K_a+4G_a)) \quad (6)$$
$$Gc_{(-)}=G_m+V_a/(1/(G_a-G_m)+6V_m(K_m+2G_m)/(5G_m(3K_m+4G_m))) \quad (7)$$
$$Gc_{(+)}=G_a+V_m/(1/(G_m-G_a)+6V_a(K_a+2G_a)/(5G_a(3K_a+4G_a))) \quad (8)$$
$$Ec_{(-)}=9Kc_{(-)}Gc_{(-)}/(3Kc_{(-)}+Gc_{(-)}) \quad (9)$$
$$Ec_{(+)}=9Kc_{(+)}Gc_{(+)}/(3Kc_{(+)}+Gc_{(+)}) \quad (10)$$

where:

E - Young's modulus;	m - matrix;
K - bulk modulus;	a - aggregate;
G - shear modulus;	c - composite;
n - Poison's ratio;	(-) - value for lower HS bound;
V - relative volume of the phase;	(+) - value for upper HS bound.

For calculation of HS bounds, the bulk and the shear moduli of the matrix and the aggregates must be known but usually they are not provided by the researchers as part of the experimental data. However they can be evaluated by equations (1-4) using the data for the Young's moduli and knowing the Poison's ratios of the phases.

In most studies considered in **Table 1**, there was no experimental data provided on Poison's ratio of the constituent materials - cement paste, mortar and aggregates. According to Anson and Newman [16], the Poison's ratio for cement paste is close to 0.25 and is not influenced by W/C, while for the mortars it is between 0.17 and 0.23 and depends strongly on the sand content and W/C. Mandel and Dantu [9] used the value of 0.25 for cement paste and mortar in their study. Nilsen and Monteiro [15] reported a value of 0.26 after testing the original matrix used by Hirsch. In this study values of 0.25 for cement pastes and 0.20 for cement mortars are assumed.

The Poison's ratio for the coarse aggregates can vary between 0.10 and 0.20. In this investigation, an average value of 0.15 is assumed for the coarse rock aggregates. The values of 0.25 for glass, 0.30 for steel and 0.45 for lead are assumed as used by Mandel and Dantu [9].

The calculations show that a change in the Poison's ratio for one of the phases in the range between 0.1-0.2 influences the results for HS bounds less than 1.0% (usually less than 0.5%, especially for LHS). This means that any inaccuracies in the assumed values for the Poison's ratios will have insignificant effect on the results of the analysis conducted in this paper.

Analysis of Experimental Data

Interpretation of experimental results with HS bounds

Fig.1 shows representative sets of data from different authors together with the calculated HS bounds. The graphs give the link between the volume fraction of the aggregate and the elastic modulus of the composite in relation to the elastic moduli of the matrix and the aggregate. The data sets chosen, show ordinary cement composites - concretes and mortars with various W/C ratio, various type of matrix and aggregates at different testing conditions. Va is varied in a broad range - from 0.2 to 0.8. It can be seen from the figure that the experimental data follow the main trend of the theoretical HS bounds for a two-phase composite. At the same time most of the points lie very close and some of them slightly below the lower HS bound.

For a precise assessment of the situation of every experimental point or a given set of data to HS bounds, the experimental error for the Em, Ea and Ec should also be considered. However, experimental errors for Ec and Em could be found only in the paper of Stock, Hannant and Williams [17] with an average value of the coefficient of variation of 4.6% for compression and 6.8% for tension tests. There were no data at all for the experimental error of Ea, where the values were often taken from other studies or from handbooks. For some aggregates this error can be significant [18]. Considering the possible experimental error, HS bounds should be regarded not as lines but as ranges corresponding to the variations in the values of Em and Ea with an accepted probability of occurrence. For illustration, dashed lines are shown on **Fig.1** to give the ranges in which HS bounds can vary when Em and Ea change within ±5%. In analyzing the location of Ec data to HS bounds their experimental error should also be

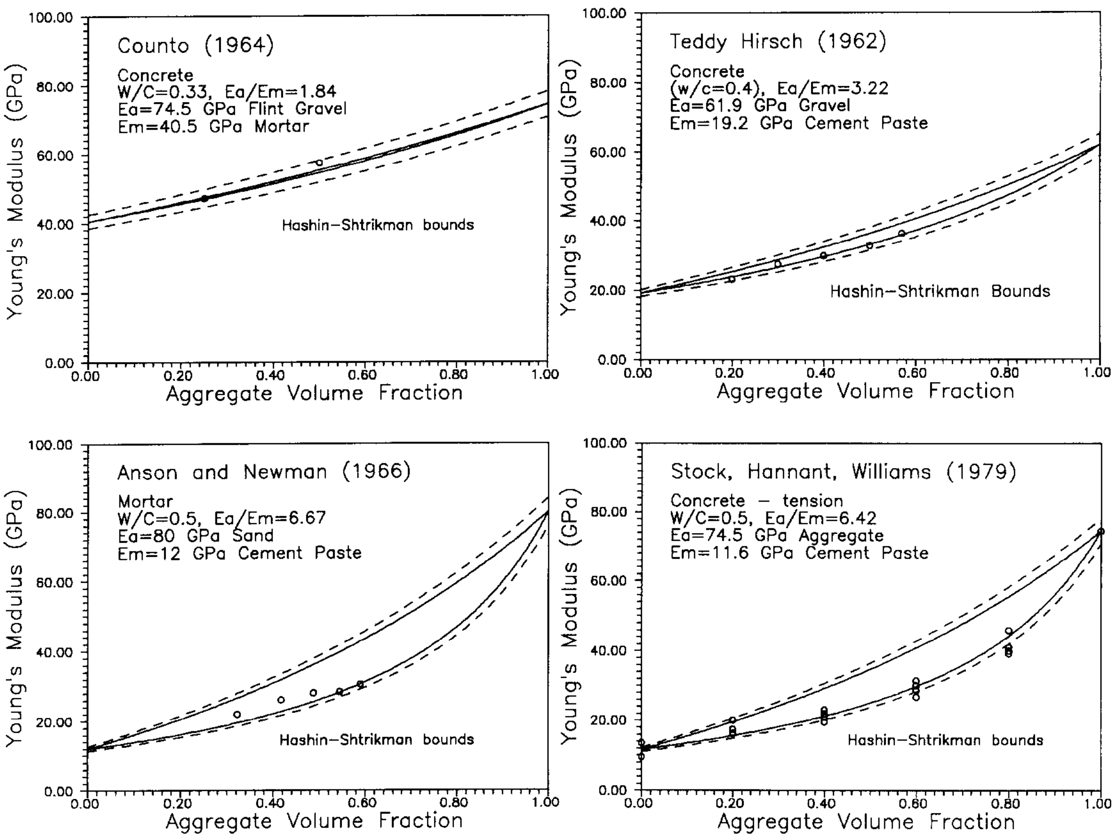

FIG.1.
Experimental data sets with corresponding HS bounds.

considered, however, due to lack of reported information on the experimental errors, a correct individual statistical analysis for every point or set of data cannot be made.

In this study, an attempt is made to asses the situation of the data as a whole. For this purpose a generalized presentation of all experimental data is made. Since most of the points are situated close to the lower HS bound (LHS) a normalization to this bound is chosen as most appropriate. The normalized data are presented graphically in **Fig.2.** Analysis show that the data exhibits a nearly normal distribution with mean value +2.67%, which has a 95% confidence limits of ±2.7% and standard deviation of 12.25%. These results show that LHS bound can be successfully used for modeling the elastic behavior of concrete as a composite. Other two-phase composite models can also be applied, but for good predictability the values computed should lie close to the LHS bound.

A conclusion can be made that statistically the elastic behavior of concrete closely resemble that of a two-phase composite. However there is still a significant number of experimental data situated outside the bounds. The analysis show a notable shift of all data towards the lower HS bound. As mentioned above this can be due to ignoring the effect of air content in the composite, or due to anisotropy in some of the specimens because of segregation. It can also be attributed to the influence of the

TABLE 1

Experimental Data for Modulus of Elasticity of Cement-Based Composites and the Calculated Hashin-Shtrikman Bounds

Author	W/C	Matrix		Aggregate			Composite	HS bounds		Ec-LHS
[year]		Type	Em	Type	Ea	Va	Ec	LHS	UHS	LHS
			GPa		GPa		GPa	GPa	GPa	%
1	2	3	4	5	6	7	8	9	10	11
Teddy	0.40	Cement	19.20	Steel	207.00	0.30	37.30	31.94	54.78	16.78
Hirsch		Paste				0.40	44.50	38.29	69.27	16.22
[1962]						0.50	53.40	46.45	85.47	14.96
				Sand	75.90	0.20	25.70	24.40	26.88	5.33
						0.40	33.90	31.23	35.90	8.55
				Glass	72.40	0.20	26.10	24.30	26.46	7.41
						0.30	30.80	27.30	30.55	12.82
						0.40	35.30	30.80	34.98	14.61
						0.50	37.40	35.00	39.81	6.86
						0.57	42.80	38.20	43.46	12.04
				Gravel	61.90	0.20	23.00	23.72	25.21	-3.04
						0.30	27.40	26.40	28.56	3.79
						0.40	29.70	29.44	32.18	0.88
						0.50	32.60	32.92	36.10	-0.97
						0.57	36.10	35.67	39.05	1.21
				Lime-	31.90	0.20	20.20	21.25	21.38	-4.94
				stone		0.30	20.60	22.35	22.52	-7.83
						0.40	22.30	23.51	23.71	-5.15
						0.50	23.90	24.72	24.95	-3.32
						0.57	23.40	25.62	25.84	-8.67
				Lead	15.00	0.20	17.20	18.57	18.52	-7.38
				Drops		0.40	16.70	17.80	17.73	-6.18
						0.50	16.20	17.37	17.3	-6.74
Mandel	0.429	Mortar	31.30	Glass	74.20	0.35	39.90	41.70	43.60	-4.32
and						0.45	43.10	45.20	47.20	-4.65
Dantu						0.55	46.70	49.40	51.20	-5.47
[1963]				Glass	74.20	0.35	39.30	41.70	43.60	-5.76
						0.45	42.60	45.20	47.20	-5.75
		Mortar	36.40	Diorite	104.70	0.40	49.50	51.90	60.40	-4.62
						0.50	55.00	57.60	62.30	-4.51
						0.55	56.80	60.90	65.60	-6.73
		Mortar	33.70	Diorite	104.70	0.33	42.50	47.40	51.10	-10.34
						0.42	47.10	52.10	56.80	-9.60
						0.52	52.70	58.10	64.30	-9.29
		Mortar	32.60	Diorite	104.70	0.33	41.90	46.10	50.30	-9.11
						0.42	47.50	50.90	56.90	-6.68
						0.52	51.80	57.10	62.60	-9.28

TABLE 1 (Contd.)

1	2	3	4	5	6	7	8	9	10	11
				Iron	104.80	0.25	54.30	50.63	52.67	7.25
Upendra		Mortar	40.50			0.50	71.70	63.60	66.95	12.74
Counto	0.33			Flint	74.50	0.25	47.20	46.98	47.49	0.47
[1964]				Gravel		0.50	57.40	54.58	55.36	5.17
				Glass	72.40	0.25	46.90	46.73	47.19	0.36
						0.50	54.30	53.94	54.62	0.67
				Poly-	0.29	0.25	22.90	24.63	2.16	-7.02
				thene		0.50	14.50	13.96	0.93	3.87
Anson	0.30	Cement	23.00	Sand	80.00	0.386	33.20	35.58	39.61	-6.69
and		Paste				0.486	39.20	40.02	44.78	-2.05
Newman	0.40		17.00			0.352	27.90	27.13	32.78	2.84
[1966]						0.449	31.80	31.06	38.02	2.38
Mortar						0.520	31.20	34.41	42.17	-9.33
						0.576	34.20	37.40	45.65	-8.56
	0.50		12.00			0.322	21.90	19.53	26.73	12.14
						0.417	26.00	22.73	31.98	14.39
						0.487	28.00	25.54	36.19	9.63
						0.544	28.30	28.19	39.85	0.39
						0.589	30.40	30.56	42.90	-0.52
	0.60	Cement	8.00	Sand	80.00	0.298	22.10	13.18	21.75	67.68
		Paste				0.389	21.40	15.49	26.82	38.15
						0.459	23.20	17.65	31.06	31.44
						0.515	24.60	19.68	34.71	25.00
						0.561	25.60	21.61	37.88	18.46
Anson	0.50	Cement	12.00	Sand	69.00	0.63	34.90	30.90	40.34	12.94
and		Paste		and		0.66	34.20	32.69	42.32	4.62
Newman				Gravel		0.67	35.40	33.65	43.34	5.20
[1966]						0.68	36.20	34.20	43.92	5.85
Concrete						0.70	38.60	35.62	45.36	8.37
						0.73	39.60	37.06	46.77	6.85
	0.50	Mortar	28.30	Gravel	69.00	0.18	34.90	32.89	33.75	6.11
		(1:2.5)				0.25	34.20	34.89	36.04	-1.98
						0.28	35.40	35.94	37.23	-1.50
						0.30	36.20	36.56	37.92	-0.98
						0.35	38.60	38.09	39.61	1.34
						0.40	39.60	39.62	41.28	-0.05
Stock	0.5	Cement	11.60	Graded	74.50	0.20	17.80	15.59	19.64	14.18
Hannant		Paste		Aggreg.		0.40	21.40	21.22	29.25	0.85
Williams		Tension				0.60	29.00	29.76	40.97	-2.55
[1979]						0.80	41.30	44.28	55.63	-6.73
		Cement	13.40	Graded	74.50	0.20	15.80	17.75	21.36	-10.99
		Paste		Aggreg.		0.40	23.20	23.77	30.81	-2.40
		Compre-				0.60	30.70	32.63	42.26	-5.91
		ssion				0.80	39.10	47.03	56.45	-16.86

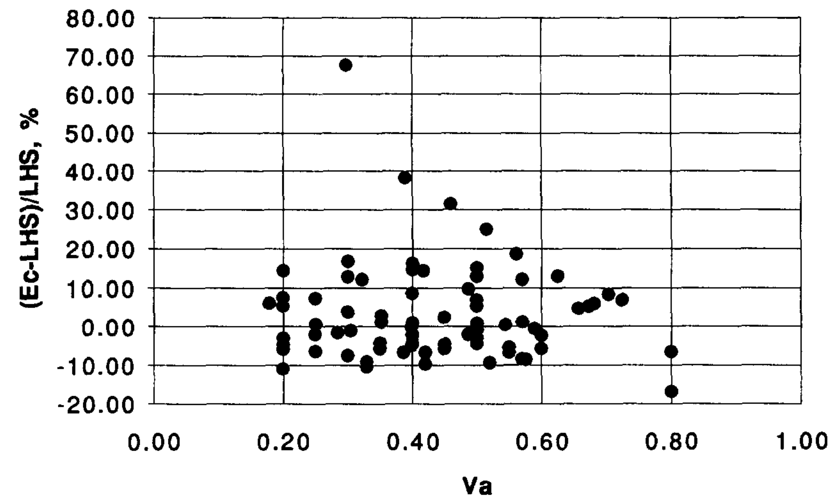

FIG.2.
Experimental data, normalized to the lower HS bound.

transition zone which has a different structure and probably different elastic properties than those of the matrix.

The Influence of Transition Zone on the Elastic Behavior of Concrete

The influence of transition zone (TZ) on the elastic properties of concrete depends mainly on volume and modulus of elasticity of TZ. The volume of the TZ is closely related to the total aggregate surface or the interface surface. If Va is increased at a constant specific surface of the aggregate, the total interface area will be increased which increases the volume of the TZ. This will result in a reduction of the volume of the matrix. If the elastic properties of TZ differ significantly from those of the matrix this must result in a divergence of the experimental data from the general trend of HS bounds for a two-phase composite. At higher Ea/Em, HS bounds are further apart, but as it was already shown, the experimental data lie mainly close to the LHS bound, so, it is reasonable that the divergence from the LHS bound should be considered and further analyzed.

Fig.3 shows the data sets of **Fig.1**, normalized to LHS bound. The linear regression lines show how the data diverge from LHS bound while Va increases. The divergence for every set of data can be characterized by one value i.e. the slope of the linear regression line. This value gives the rate of relative change of the elastic modulus of the composite per unit volume change of the aggregate, or this is a characteristic for the influence of TZ. It would be more correct to relate this value to the change of the volume of TZ or to the change of the aggregate surface (the total interface surface), however this is not possible due to lack of more detailed experimental information.

It can be seen from **Fig.3**, that the values which represent the influence of the TZ are not the same for the different sets of data. An explanation for this can be found considering some of the main factors influencing the properties of the TZ.

W/C ratio is the principle factor governing the structure and the properties of the cement paste in the composite (mortar, concrete) and it is also the main factor

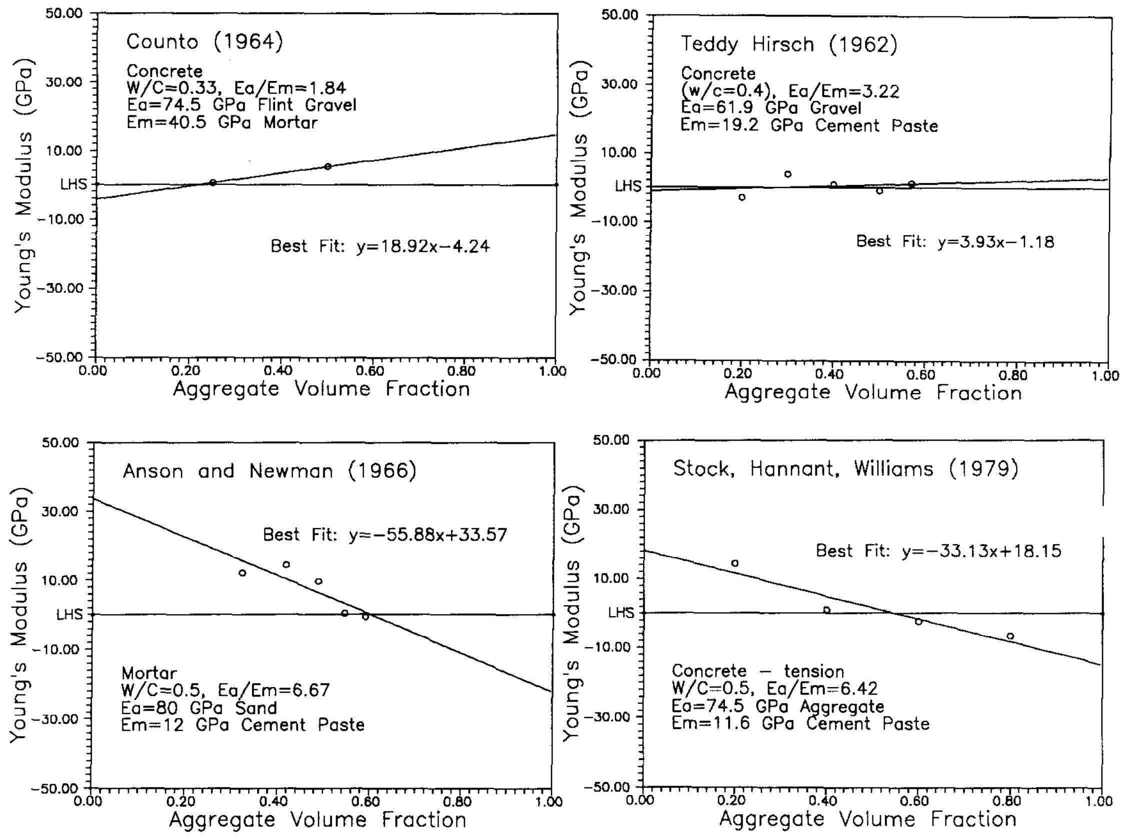

FIG.3.

Linear regression of data sets, normalized to the lower HS bound.
The slope of the regression line gives the effect of the Transition Zone.

influencing the properties and the thickness of the TZ. In order to evaluate the influence of this factor on the effect of TZ on Ec, the results for 23 data sets from five groups of authors were analyzed. **Fig.4** shows the results of the analysis. The data for concretes and for mortars are presented separately since the specific surfaces of the aggregates differ significantly. Most of the coarse aggregates used, have relatively close values of specific surface - 3-6 cm^2/cm^3 and a correct comparison between them can be made. For the sand, the specific surface values are around 100 cm^2/cm^3 and the TZ is expected to be in thinner layers.

Fig.4 shows the influence of W/C on the rate of change of the normalized values of Ec in % per unit Va, or in other words, how the effect of TZ on Ec depends on W/C. It is clearly demonstrated that there is a notable effect of TZ on the elastic modulus of concrete and that this effect is strongly related to the W/C. It can be seen that at high W/C, the effect of TZ is negative and with the reduction of W/C this effect is reduced and even turns into a positive one at low W/C. The linear regression of data for concrete (**Fig.4a**) and for mortar (**Fig.4b**) shows that the effect of TZ approaches zero at a value 0.41 for concrete and 0.37 for mortar. For convenience, a value of 0.4 can be used for mortar and concrete.

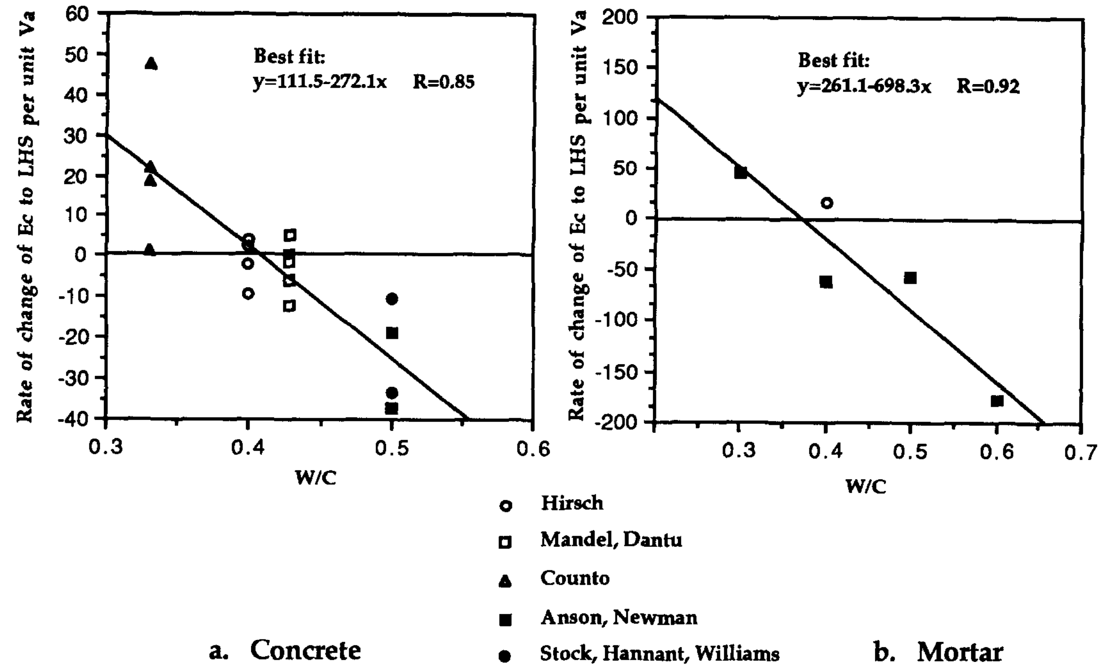

FIG.4.
The influence of W/C on the effect of Transition Zone on the modulus of elasticity of cement based composites.

Discussion

Trying to evaluate the influence of the transition zone (TZ) on the elastic properties of the composite material - concrete, it must be recognized that the properties of TZ are not constant in direction normal to the surface of the aggregate. There is a gradient in the structure and respectively in the properties of TZ in direction normal to the surface of the aggregates. So, considering TZ as a separate phase it should be characterized with some average or equivalent properties. The pore structure, the equivalent properties, as well as the thickness of the TZ are strongly related to the water content in the cement matrix.

After the mixing of the concrete (or mortar) and during the process of initial hydration and setting of the cement paste, there is a process of migration of part of the free water to the surface of the aggregates. This rate of migration is higher for composites with higher W/C ratios. The migration of water results in reduction of the real W/C in the bulk cement paste. So it should be expected that the elasticity of the bulk cement paste (or the real matrix) will increase with the introduction of the aggregate and with the increase of its volume. As a result of the process of migration of the water to the surface of the aggregates there is a bigger W/C ratio in the TZ and reduced W/C ratio in the remaining cement paste but for the whole volume (the volume of TZ and the bulk cement paste) this W/C is equivalent to the initial W/C or the the designed value (if the absorption of the aggregates is taken into account). In other words the formation of TZ can also be regarded as a process of transfer of elasticity from the surface of the aggregates to the bulk cement paste as far as the elastic properties of the matrix and the TZ are directly related to the water content and their porous structure. It can be expected that the overall equivalent elasticity of the complex matrix (containing also the volume of the TZ) will be the same as that of a pure cement paste with the same W/C ratio. If this phenomenon exsists, the overall behavior of concrete will be the same as this of a two-phase composite. From **Fig.4** it seems that this can be true at least for a particular range of W/C ratios e.g. around 0.4.

At W/C close to 0.4 (**Fig.4**), the reducing effect of the TZ with lower equivalent elasticity is compensated by the positive effect of the increased elasticity of the bulk cement paste caused by the reduction of the real W/C. As a result there is no notable effect on Ec and with the increase of Va it tends to change as expected for a two-phase composite - following the HS bounds (LHS).

At higher W/C (W/C>0.4), the TZ is with lower equivalent elasticity than that of the matrix and in a thicker layer and in bigger relative volume to a unit interface surface. With the increase of Va, it causes reduction of Ec that is bigger than the increase of Em caused by the reduction of the real W/C in the bulk cement paste. The overall result is a significant relative reduction of Ec compared to the expected value for a two-phase composite, e.g. HS bounds (LHS).

At lower W/C (W/C<0.4), the TZ is thinner and its equivalent elastic properties are probably very close to that of the matrix or can be even higher. With an increase of Va, the influence of TZ is either negligible or positive. Em increases as a result of the reduction of the real W/C. These two coupled effects cause a notable relative increase of Ec compared to the expected value for a two-phase composite - HS bounds (LHS).

The lower the W/C, the thinner is the TZ and the greater can be the influence of the aggregate type on the equivalent properties of TZ. In high strength concretes, the thickness of the TZ is relatively small due to the use of mineral additives such as silica fume, chemical admixtures such as HRWR and extremely low W/C (W/C<0.3). For such concretes, since the elastic properties of TZ are sensitive to the aggregate type, it can have considerable effect on the elastic properties of the composite.

Conclusions

The elastic behavior of concrete is statistically close to a two-phase composite and can be successfully described by the Lower Hashin-Shtrikman bound. HS bounds prove to be an effective tool for detecting the effects of TZ on the elastic properties of concrete.

There is a notable effect of the Transition Zone on the elastic properties of concrete. It is demonstrated as a divergence of the experimental data from the general trend of Hashin-Shtrikman bounds with the increase of aggregate volume and the Transition Zone volume. This effect is strongly related to the water-cement ratio and it changes from negative at higher W/C into positive at lower W/C.

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