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Critical reevaluation of the prediction of effective Poisson's ratio for porous materials

K. K. PHANI*, DIPAYAN SANYAL
Central Glass and Ceramic Research Institute, 196 Raja S. C. Mullick Road, Kolkata 700032, India
E-mail: kkphani@cgcri.res.in

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An equation has been derived for the prediction of the Poisson's ratio of porous materials having ribbon like pores. The equation fits the data of silica gel with ribbon like pores quite well for the whole range of porosity. For spherical pores, a relation derived by previous researchers showed an anomalous variation in Poisson's ratio with porosity. It has been shown that it was due to the mathematical form of the function chosen to describe the relation between the bulk and Young's moduli with porosity. In this analysis, a modified form of the function to describe Young's and bulk modulus with porosity has been suggested for spherical pores. The derived relation for variation of Poisson's ratio with porosity shows good agreement with the prediction by self consistent theory.

1. Introduction

Many engineering materials, such as, ceramics, composites and metals sintered from powder compacts are inherently porous, with a wide variety and distribution of pore sizes and shapes depending on the synthesis route adopted in material preparation. Estimation of the effective elastic moduli of such materials characterized by microscopic heterogeneity in pore shapes and sizes has been a matter of considerable interest for several decades. Experimental studies, based on ultrasonic longitudinal and shear wave measurements have been conducted for porous materials for estimation of the Young's modulus and shear modulus as functions of the average porosity (P) determined from the ratio of the bulk density to the theoretical density of the material [1–7]. The resulting data have yielded semi-analytical expressions for prediction of the effective moduli [7–10]. More fundamental predictions of these moduli have also been attempted from micromechanical theories by several researchers [11–18]. Distinctions have been made between spherical pores [11–18] and non-spherical pores [19–22] to derive various predictive models. Among the elastic moduli, the importance of the Poisson's ratio for structural calculations cannot be overemphasized. For example, in the analysis of bonded dissimilar materials, a mismatch in Poisson's ratio can give rise to singular stress fields [15]. In the earth sciences too, the sensitivity of the Poisson's ratio to pore structure has been recognized as an effective tool for studying the latter [23, 24]. However, in comparison with the effective Young's modulus (E_e) and the effective bulk modulus (K_e), less attention has been paid to the prediction of the effective Poisson's ratio (ν_e) for porous materials [15, 17, 18, 25]. Researchers are divided on the issue whether Poisson's ratio is a function of porosity of materials. While some declare ν_e as constant [26], others claim it to be a function of porosity [15, 17, 18, 25]. Such debates possibly arise out of the fact that the Poisson's ratio is usually derived as a function of E_e and K_e whose relative dependence on porosity may accentuate or dampen the dependence of ν_e on P. Previous investigations on this issue, especially that of Arnold *et al.* [25] have been far from conclusive. It is worthwhile to present the salient features of the Arnold's model which merit closer scrutiny. Arnold *et al.* [25] have observed that the Poisson's ratio (ν_e) versus porosity (P) relation is concave downward upto a P value of 0.4 and is convex upward above this value. This is clearly depicted in Fig. 1. It shows a distinct point of inflexion at $P = 0.4$ with a sharp kink. From Fig. 1, we can further infer that the experimental data of Ashkin *et al.* [4], with which Arnold *et al.* [25] compared their theoretical prediction, do not indicate such a trend, except at porosity higher than 0.5. Arnold *et al.* [25] have also not offered any physical explanation to the peculiar nature of their theoretical curve in Fig. 1 and chose to offer the validity of their analysis as "a challenge". In fact, a close look at the analysis of Arnold *et al.* [25] reveals the following anomalies:

1. The data of Ashkin *et al.* [4] is for colloidal gel derived silica having ribbon like pore structure, as reported by the authors, whereas the analysis of

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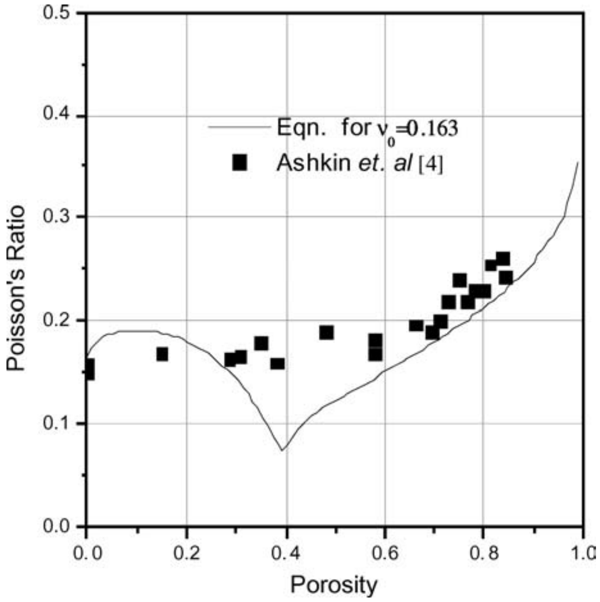

Figure 1 Variation of the Poisson's ratio of porous gel-derived silica with porosity.

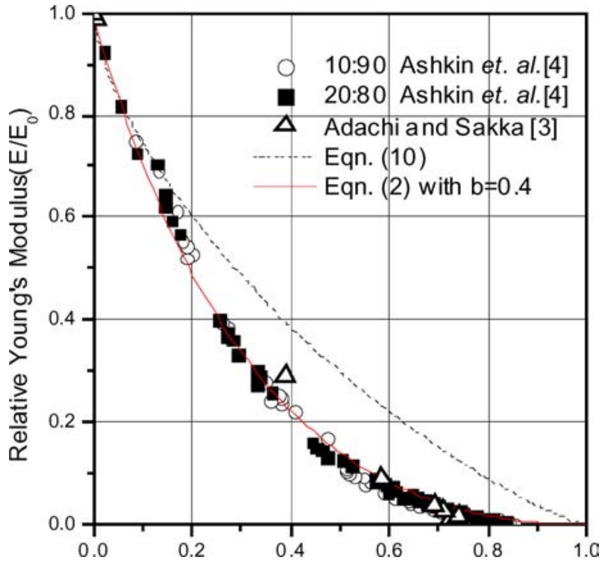

Figure 3 Variation of the relative Young's modulus with porosity for gel derived silica.

Arnold *et al*. [25] is for spherical pores. Such an effort implicitly assumes that porosity is the only variable required for describing the relation of the variation of the elastic properties, such as, Poisson's ratio. It is well established now that besides porosity, the pore structure is an important parameter for predicting the variation of the elastic properties [8, 9, 27].

2. While analyzing the bulk modulus data of glass with spherical pores reported by Walsh *et al*. [30], Arnold *et al*. [25] attempted a blending of two Equations 5 and 6, presented in the next section, with a sigmoidal function, s. The resulting Equation 8 shows an abrupt change in the slope of the curve at a porosity level of 0.4 which cannot be described by any physical phenomena. Fig. 2 brings about this issue quite clearly. Since this sigmoidal function appears at the denominator of the relation for the variation of Poisson's ratio with porosity derived by Arnold *et al*. [25], it can be argued that this function may have contributed to the kink shown in Fig. 1 at $P = 0.4$.

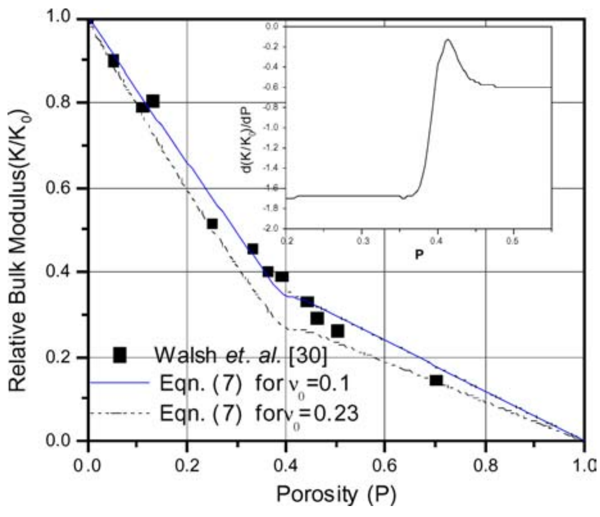

Figure 2 Variation of the bulk modulus with porosity for glass.

3. While comparing the data of Ashkin *et al*. [4] with the theoretical predictions, Arnold *et al*. [25] implicitly assumed that Equation 10 describes the variation of Young's modulus in materials with spherical pores. However, it fails to fit the data of Young's modulus versus porosity reported by Ashkin *et al*. [4], as shown in Fig. 3. This goes to confirm, yet again, the inadequacy in the analysis of Arnold *et al*. [25].

The primary objectives of this paper are twofold—firstly, to find a Poisson's ratio versus porosity relation taking into account the ribbon like pore shape for the data of Ashkin *et al*. [4] and secondly, to examine mathematically, the nature of variation of the ν_e versus P for spherical pores.

2. Prediction of Poisson's ratio for materials with ribbon like pores

The effective Poisson's ratio is related to the two elastic moduli, E_e and K_e as follows:

$$\nu_e = 0.5 - \frac{E_e}{6K_e} \tag{1}$$

It is imperative that suitable expressions for E_e and K_e be first constructed for ribbon like pore structure, before deriving the relation for ν_e versus P. For this purpose, we used the data reported by Ashkin *et al*. [4] for Young's modulus variation for porosity. Ashkin *et al*. [4] studied the change in elastic moduli with porosity for gel-derived porous silica sintered from a relative density of 0.15 to full density. The gels were made from colloidal silica, potassium soluble silicate and formamide and the gel structure was varied by changing the ratio of weight percentages of colloidal silica to potassium soluble silicate from 5:95 to 25:75. Further details of their experimental study can be found in [4]. They fitted their data to the relation of Young's modulus derived by Nielsen [13]:

$$\text{Relative modulus } \{E/E_0, K/K_0 \text{ or } G/G_0\} = (1-P)^2 \Big/ \left[1 + \left(\frac{1}{b} - 1\right) P\right] \quad (2)$$

where b is a term related to the pore morphology. Ashkin *et al*. [4] obtained the best fit of the measured Young's modulus data with the theoretical prediction from Equation 2 for $b = 0.4$. Fig. 3 depicts the quality of fit of these data against the solid line drawn using Equation 2 with $b = 0.4$ which pertains to ribbon like pore structure.

To derive the Poisson's ratio versus porosity relation, data for bulk modulus of silica samples with similar pore structures are required. Unfortunately, bulk modulus data were not reported by Ashkin *et al*. [4]. For this we used the data reported by Adachi and Sakka [3] for silica gel prepared by the sol-gel method from a tetramethoxysilane solution, where porosity varied in the range from 0 to 0.726. The details of their experimental work is reported in [3]. In their work, Adachi and Sakka [3] tabulated the Young's modulus, shear modulus and bulk modulus data estimated from the longitudinal and transverse velocities of sound wave measured from the resonance of vibrating cubic samples as per Goto and Soga's technique [37]. Before using these data, it is necessary to check that this material has similar pore structure with that of Ashkin *et al*. [4]. To verify the same, we plotted their Young's modulus data in Fig. 3. The data closely followed the curve predicted by Ashkin *et al*. [4] based on Equation 2 with $b = 0.4$ over the whole range of porosity. Thus it will be reasonable to assume that the material used by Adachi and Sakka [3] had similar pore structure as that of Ashkin *et al*. [4] and their bulk modulus data can be used for the analysis of Equation 1. The bulk modulus data of Adachi and Sakka [3] are shown in Fig. 4. The data were again fitted to Equation 2 which yielded a value of $b = 0.476$.

Combining Equations 1 and 2, we obtain the following relation describing variation of effective Poisson's ration with porosity for ribbon shaped pores:

$$v_e = 0.5 - \frac{3(1-2v_0)[1+1.5P]}{1+1.1P} \quad (3)$$

A plot of Equation 3 for $v_0 = 0.163$ for silica [4] is shown in Fig. 5 along with the data of Ashkin *et al*. [4]. Also shown in the plot are the Poisson's ratio values calculated from the elastic and shear modulus data reported in Adachi *et al*. [3]. As can be seen from Fig. 5, the predicted values show reasonable agreement with the data, having the maximum deviation of +20% from the predicted value. Considering the fact that the Poisson's ratio is more sensitive to the error of measurements, this deviation is within reasonable limits. Also the trend predicted by the above equation agrees with the observation by Ashkin *et al*. [4] that, for their material, Poisson's ratio increases with increase in porosity. The nature of the variation predicted by Equation 3 is also in qualitative agreement with the

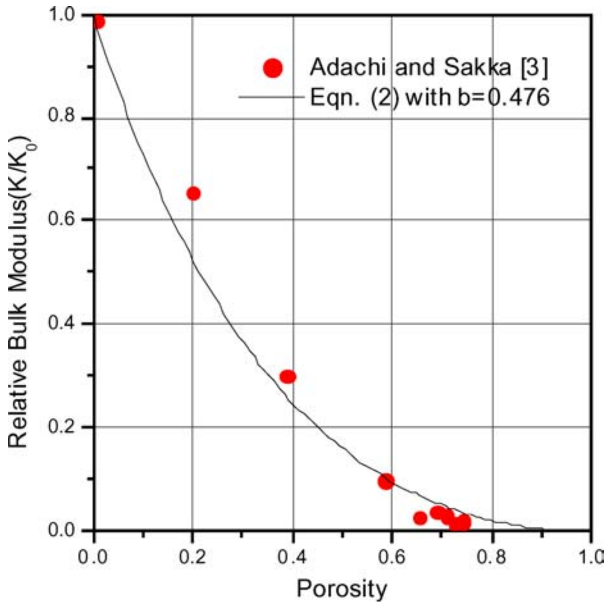

Figure 4 Variation of the relative bulk modulus with porosity for gel derived silica.

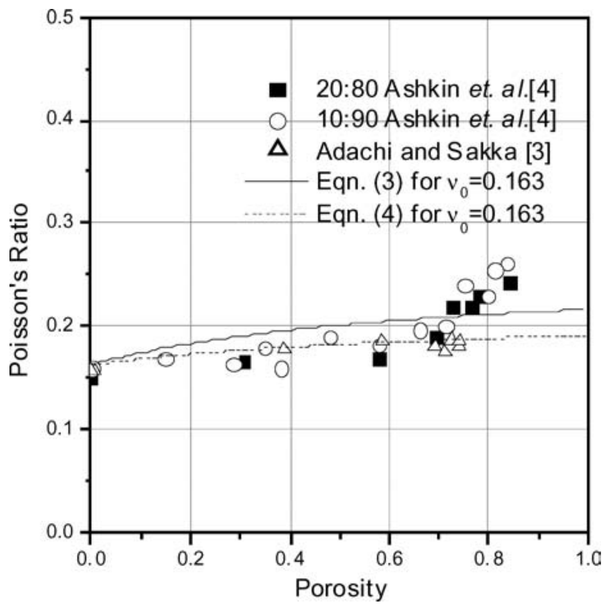

Figure 5 Variation of the Poisson's ration with porosity for nonspherical pores.

variation predicted by the self-consistent theoretical analysis of Dunn and Ledbetter [15] for needle shape pores given by the following equation, as shown in Fig. 5:

$$v_e = \frac{-15v_0 + P(8v_0 - 5)(v_0 + 1)}{-15 + 4P(4v_0 - 5)(v_0 + 1)} \quad (4)$$

3. Prediction of Poisson's ratio for materials with spherical pores

For analysis of data for spherical pores, we again need the experimental data of Young's modulus and bulk modulus of materials having spherical pores. Arnold *et al*. [22], in their analysis have used the data reported by Walsh *et al*. [30] for glass (54.4 wt% SiO_2, 14.4 wt%

B_2O_3, 14.1 wt% CaO, 10 wt% Al_2O_3, 6.5 wt% Na_2O and 0.7 wt% K_2O) with spherical pores having $\nu_0 = 0.23$. Arnold *et al*. [25] analysed these data in terms of two equations. For low concentration of spherical pores, they adopted the equation derived by Ondracek [28, 29]:

$$K_{P,1} = K_0 \frac{2(1-2\nu_0)(3-5P)(1-P)}{2(3-5P)(1-2\nu_0)+3P(1+\nu_0)} \quad (5)$$

For high porosity range, they used the the expression derived by Walsh *et al*. [30]:

$$K_{P,2} = K_0 \frac{2(1-2\nu_0)(1-P)}{3(1-\nu_0)} \quad (6)$$

They then combined these two equations by:

$$K_P = (1-s)K_{P,1} + sK_{P,2} \quad (7)$$

where s is the sigmoidal function given by

$$s = \frac{1}{1+\exp\{-100(P-0.4)\}} \quad (8)$$

The variation of bulk modulus with porosity as per Equation 7 is shown in Fig. 2 for two values of Poisson's ratio, $\nu_0 = 0.1$ and $\nu_0 = 0.23$. The curve drawn by Arnold *et al*. (Fig. 3 in [25]) was possibly drawn for $\nu_0 = 0.1$ which was not explicitly mentioned by them. The curve was also possibly smoothened as shown by dotted line in Fig. 2. For the correct magnitude of $\nu_0 = 0.23$, the plot is drawn in dashed line in Fig. 2 and fits the data of Walsh *et al*. [30] rather poorly. The plot also reveals two distinct regimes with negative slopes—before and after the porosity value of 0.4, pertaining to Equations 5 and 6, respectively. Near the porosity value of 0.4, the blending of these equations is not sufficiently smooth and shows an abrupt change in continuity, which is clearly brought about by plotting the first derivative of the relative bulk modulus with respect to porosity. This plot is shown in the inset of Fig. 2 for porosity values ranging from 0.2 to 0.5. From the physical point of view, it is unlikely that the modulus versus porosity curve will show such inflexions in the slope over the entire porosity range. This possibly arises due to form of the function, s, given by Equation 8. Incidentally, the kink in the Poisson's ratio-porosity curve in Fig. 1 corresponds to this porosity range only.

To verify the same, the Equations 5 and 6 were blended with a polynomial function of the first order, of the form:

$$K_P = (aP+b)K_{P,1} + (cP+d)K_{P,2} \quad (9)$$

The parameters a, b, c and d of the above equation were obtained by least square regression analysis by fitting the equation to the bulk modulus data with additional conditions that at $p = 0$, $K/K_0 = 1$ and at $p = 1$, $K/K_0 = 0$. The fitted equation is shown in Fig. 6 with

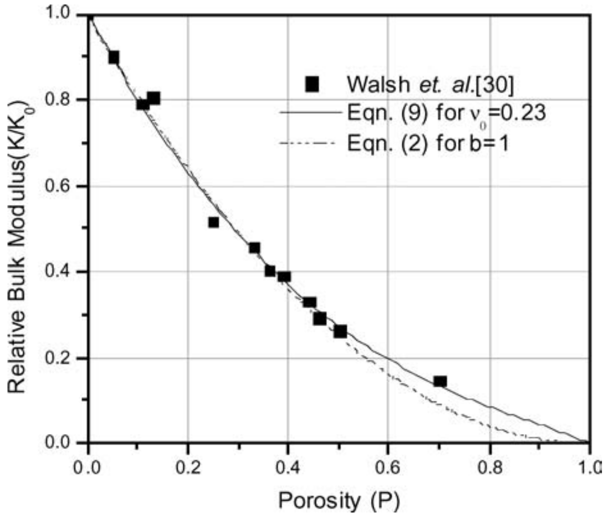

Figure 6 Variation of the relative bulk modulus with porosity for glass with spherical pores.

values of $a = -0.24$, $b = 0.019$, $c = -1.54$ and $d = 1.85$.

For Young's modulus variation with porosity, Arnold *et al*. [25] assumed the equation derived by Boccacini *et al*. [31]:

$$E = E_0(1-P^{2/3})^{1.21} \quad (10)$$

Therefore Equation 1 yields:

$$\nu_0 = 0.5 - \frac{3(1-2\nu_0)(1-P^{2/3})^{1.21}}{6[(-0.24P+0.019)K_{P,1}+(-1.54P+1.85)K_{P,2}]} \quad (11)$$

This equation is plotted in Fig. 7 as the dotted line for $\nu_0 = 0.163$. It can be seen from the figure, that the Poisson's ratio variation is now concave downwards upto a porosity of about 0.6 and then convex upwards approaching the value of 0.5 asymptotically. No kink appears in the curve as shown in Fig. 4 at $P = 0.4$, thus indicating that the nature of the curve shown in Fig. 1 possibly arises due to the form of Equation 7. The concave and convex variations in the Poisson's ratio with porosity in Fig. 1 possibly arise out of the competing slopes of the derivative of the Young's modulus and the bulk modulus with respect to porosity. It may be noted that Equation 10 shows that the Young's modulus approaches the zero porosity value asymptotically. At low values of P, $d(E/E_0)/dP$ has a very high negative slope, whereas for $d(K/K_0)/dP$ shows a smoothly varying negative slope for the whole range of porosity.

To investigate the effect of the Equation 10 on the variation of Poisson's ratio with porosity, we need to again analyze the experimental data of materials with spherical pores. Unfortunately, Walsh *et al*. [30] have not reported the Young's modulus data for their material. On the other hand, Ishai *et al*. [37] have reported the elastic modulus variation with porosity for epoxy resin with spherical pores upto a porosity of 0.72. These data are shown in Fig. 8, along with the curve for

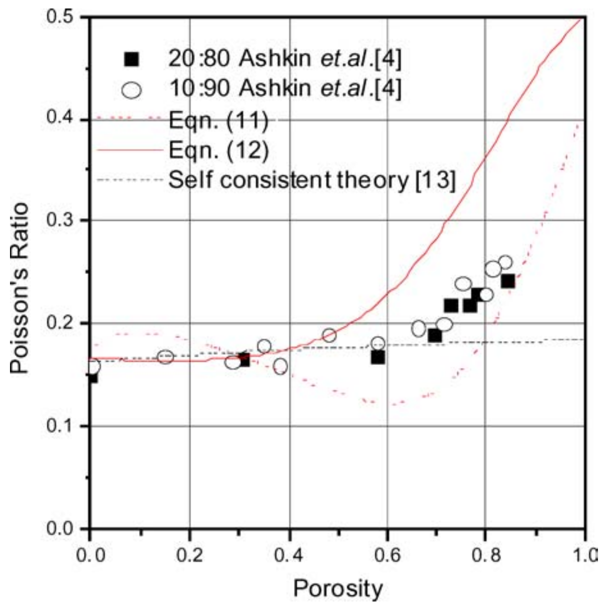

Figure 7 Variation of Poisson's ratio with porosity for spherical pores.

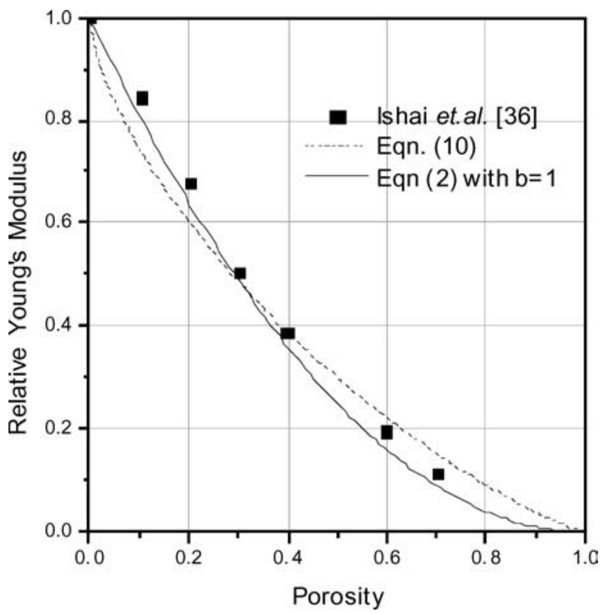

Figure 8 Variation of relative Young's modulus with porosity for epoxy resin with spherical pores.

Equation 10. It can be seen that Equation 10 fits the data well above porosity value of 0.3, whereas below 0.3 the fit is poor. Also plotted in the same figure, the curve for Equation 2 with $b = 1$ for spherical pores. The Equation 2 shows excellent agreement over the entire range of data. Thus using this equation, the Poisson's ratio versus porosity relation becomes:

$$\nu_0 = 0.5 - \frac{3(1 - 2\nu_0)(1 - P^2)}{6[(aP + b)K_{P,1} + (cP + d)K_{P,2}]} \tag{12}$$

This relation is also plotted for $\nu_0 = 0.163$ in Fig. 7, which indicates that the variation of Poisson's ratio with porosity is almost constant upto a porosity value of 0.3 and then monotonically increases to a value of 0.5. Incidentally, it may be mentioned here that the bulk modulus variation can also be described by Equation 2 with $b = 1$ (see Fig. 6). In that case, the Poisson's ratio remains constant with porosity. However, due to a lack of experimental data for Poisson's ratio of materials with spherical pores, it is difficult to verify which of these curves will describe the actual behaviour of the material. It will be of academic interest to compare these predictions with self-consistent theory which has been shown to describe the Poisson's ratio of porous material upto a porosity of 0.3 quite well [8]. The relation between the Poisson's ratio and porosity for spherical pore is given by the self-consistent theory as follows:

$$\nu_e = \frac{2\nu_0(5\nu_0 - 7) + P(5\nu_0 - 3)(\nu_0 + 1)}{2(5\nu_0 - 7) + P(15\nu_0 - 13)(\nu_0 + 1)} \tag{13}$$

This is shown, again, in Fig. 7, for $\nu_0 = 0.163$. It can be seen that upto a porosity of 0.3, Equations 12 and 13 agree quite well with each other. It can be seen from this figure, that the data of Ashkin *et al.* [4] for ribbon like pores, also fall very close to the theoretical curves predicted by Equations 11 and 12 for low porosity values. This may not be surprising, given the fact that the pores of silica gel at low concentrations, would tend to be closed and spherical in shape.

4. Conclusion

An equation has been derived for calculating Poisson's ratio of porous materials containing ribbon like pore structure. The experimental data shows fairly good agreement with that of the theoretical predictions. For spherical pores, it has been shown that the general nature of the theoretical variation of Poisson's ratio with porosity is dependent on the mathematical form of the equation chosen for describing the bulk and elastic moduli for the entire range of porosity. The exact nature of the theoretical variation could not be ascertained because of the lack of experimental data of the Poisson's ratio for materials having spherical pores. However, the theoretical predictions do not indicate any sharp transition in the Poisson's ratio versus porosity as predicted by Arnold *et al.* [25].

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