Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 366 (2007) 129–136
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Estimation of elastic properties of nuclear fuel material using longitudinal ultrasonic velocity – A new approach

K.K. Phani [a,*], Dipayan Sanyal [a], A.K. Sengupta [b]

[a] Central Glass and Ceramic Research Institute, 196 Raja S.C. Mullick Road, Kolkata 700 032, India
[b] Radio Metallurgy Division, Bhaba Atomic Research Centre, Mumbai 400 085, India

Received 28 September 2006; accepted 12 December 2006

Abstract

A novel methodology has been suggested for estimation of elastic properties of nuclear fuel materials based on porosity and longitudinal ultrasonic velocity. New correlations have been proposed between the elastic moduli, namely, the Young's modulus, shear modulus, bulk modulus and Poisson's ratio, and the longitudinal ultrasonic velocity. The theoretical predictions agreed extremely well with data reported for uranium dioxide and uranium nitride, respectively. The proposed method promises to be extremely useful for non-destructive evaluation of elastic properties of irradiated fuel materials which are not amenable to shear velocity measurements due to extreme fragility.

PACS: 28.52.Fa; 62.20.Dc

1. Introduction

The fuel materials used in the form of pellets in pressurized water reactors undergo deterioration in their mechanical properties during irradiation. Thus to assess the integrity and the life of the fuel rod it is necessary to characterize the mechanical behaviour of the irradiated fuel pellets. Generally micro-indentation and micro-acoustic techniques are used for this characterization [1–8]. While micro-indentation is used to determine plastic properties and indirectly through modeling elastic properties, micro-acoustic technique characterizes elastic properties using the relations provided by the physical acoustics theory. In this paper we shall be dealing with micro-acoustic characterization only. In this technique the effects of the fuel pellet porosity on their acoustical properties are first quantified and then their intrinsic elastic properties are deduced from these measurements.

For complete characterization of elastic properties of a material usually two acoustical properties are required – longitudinal and shear (transverse) wave velocities of the ultrasonic wave traveling through the material. These velocities are generally measured by generating ultrasonic waves in the material by coupling a piezoelectric crystal with the sample and measuring the time of travel of the

* Corresponding author. Tel.: +91 33 24838081; fax: +91 33 24730957.
E-mail address: kkphani@cgcri.res.in (K.K. Phani).

0022-3115/$ - see front matter
doi:10.1016/j.jnucmat.2006.12.045

wave through the material. However, direct determination of shear wave velocities in the fuel pellet material like uranium dioxide is impractical, if not impossible, due to coupling difficulties. Thus in case of fuel materials sophisticated technique like high frequency acoustic microscopy [9,10] is used to determine the longitudinal and the Rayleigh wave velocities through the fuel pellet material and then the shear wave velocity is calculated by solving an eighth degree polynomial in shear velocity correlating these three velocities [1,6]. In case of UO_2, the efficiency of the longitudinal mode is too low and the measurement of the longitudinal wave velocity by acoustic microscopy fails [10]. Therefore, a separate technique, micro-echography is used to measure the longitudinal velocity. Once the longitudinal and the shear wave velocities are known the elastic properties of pellet materials can be calculated using the relations given by the physical acoustics theory correlating ultrasonic velocities with elastic properties. Considering the difficulties in determining the shear wave velocity, the main aim of this paper is to examine whether only longitudinal wave velocity measured by micro-echography or otherwise, can be used as a predictor not only of elastic properties but also density of the fuel pellets.

2. Previous works

Attempts have been made by various investigators [5,11–13] to use longitudinal ultrasonic velocity as the predictor of elastic properties of porous materials. These studies are based on the premise that the ultrasonic velocity is more sensitive to changes in micro-structural features like pore shape, inter particle bonding, etc., than the density or porosity in the material [13]. Panakkal based on his earlier studies on sintered iron powder compacts [11] gave an equation of the form [5]:

$$M = M_o - C_M(V_{L0} - V_L), \qquad (1)$$

where M is the modulus, Young's or Shear; C_M is a constant dependent on the material and V_L is the longitudinal ultrasonic velocity. The subscript 'o' refers to the value of pore free material.

Eq. (1) can be derived for porous materials containing spherical pores using the elasticity theory from the treatment given in Ref. [14]. From the theories of physical acoustics one would expect the modulus to be at least a second degree polynomial function of ultrasonic velocity if Poisson's ratio remains invariant with porosity. Mathematically, Eq. (1) represents a piecewise approximation of the polynomial with a straight line over a narrow domain – extending the domain will bring in a large error in the estimated values. Thus Eq. (1) can be valid only over a very narrow and low range of pore volume fraction ($p \leqslant 0.05$).

The material constant, C_M is a function of Poisson's ratio, ν_o, of the pore free material and its value can be calculated from the expressions given in Ref. [11]. Taking $\nu_o = 0.316$ for polycrystalline UO_2 calculated from single crystal data of UO_2 [4], C_M values for UO_2 work out to be 28.8 and 26.7 for Young's and shear moduli, respectively. This indicates similar nature of variation of Young's and shear moduli with longitudinal velocity. This is in agreement with the analysis made by Yeheskel and Tevet [12] for porous ceramics based on self-consistent theory of Zhao et al. [15]. Panakkal [5] obtained these values as 69.3 and 25.0 for Young's and shear moduli respectively, by fitting Eq. (1) to his data by least square analysis indicating that the variation of Young's modulus is much more pronounced with longitudinal velocity than that of shear modulus with the same. This anomaly with the theory possibly arises because of the range of porosity over which the data has been fitted. Whereas Eq. (1) is considered to be valid over $p \leqslant 0.05$, it has been fitted over the range $0 \leqslant p \leqslant 0.3$.

Yeheskel [13] based on his studies on sintered iron compacts gave a semi-analytical relation between the normalized elastic moduli, M^* (shear or bulk moduli of porous compacts normalized with respect to the moduli of pore free material) and the normalized longitudinal velocity V_L^*

$$M^* \approx (\alpha V_L^{*3} + \beta V_L^{*2})C(\nu), \qquad (2)$$

where α, β are obtained from the relation

$$\rho^* \approx \alpha V_L^* + \beta, \qquad (3)$$

where ρ^* is the normalized density (normalized with respect to theoretical density), α is the reciprocal of the slope of ρ^* versus V_L^* linear correlation and the term β is a function of the tap density and the constant α multiplied by the increase in normalized longitudinal velocity due to sintering (for details reference may be made to Appendix B of Ref. [13]). $C(\nu)$ is a function of Poisson's ratio and is given by

$$C(v) = \frac{(1-2v)}{(1-v)} \frac{(1-v_o)}{(1-2v_o)}, \quad (4)$$

for shear modulus and

$$C(v) = \frac{(1+v)}{(1-v)} \frac{(1-v_o)}{(1+v_o)}, \quad (5)$$

for bulk modulus, v_o is Poisson's ratio of the pore free material. The term $C(v)$ is dimensionless and its value approaches one as Poisson's ratio of the porous material approaches that of the fully dense material. To determine M^* from Eq. (2) solely on longitudinal velocity either $C(v)$ must be expressed in terms of V_L^* or it may be neglected. Neglect of $C(v)$ leads to the uncertainties in the determination in M^* and the error is large with low values of Poisson's ratio [13]. Yeheskel [13] proposed the estimation of $C(v)$ values from Eqs. (4) or (5) using the relation

$$v = v_o (V_L^*)^m, \quad (6)$$

where m is an exponent, the value of which depends on the processing routes of the material. This means that in order to use longitudinal velocity as the sole predictor of elastic properties, v must be determined either from shear wave velocity measurements or by some other means. It may also be noted that α, β and m are the parameters which are dependent on the micro-structural features of the material like pore shape, inter particle bonding, etc., and therefore, their values will be dependent on the processing parameters of the material. So, no material specific unique values for these parameters can be specified based on a single experiment.

3. Analytical derivations

In a medium, the velocity of wave transport from some point to a neighbouring location is dependent on the interaction, point mass and medium structure. For an infinitely large solid medium physical acoustic theory indicates that sound wave velocities are functions of Young's modulus (E) and shear modulus (G) and given by the well known relations:

$$V_L = \sqrt{\frac{E(1-v)}{\rho(1+v)(1-2v)}} \quad (7)$$

and transverse or shear velocity V_S

$$V_S = \sqrt{\frac{G}{\rho}} = \sqrt{\frac{E}{2\rho(1+v)}}. \quad (8)$$

Using Eqs. (7) and (8) normalised modulus values in terms V_L^* can be written as

$$E^* = \frac{E}{E_o} = \frac{\rho}{\rho_o}\left[\frac{(1+v)(1-2v)}{(1-v)}\right]\left[\frac{(1-v_o)}{(1+v_o)(1-2v_o)}\right]V_L^{*2}, \quad (9)$$

$$G^* = \frac{G}{G_o} = \frac{\rho}{\rho_o}\left[\frac{1-2v}{1-v}\right]\left[\frac{1-v_o}{1-2v_o}\right]V_L^{*2}, \quad (10)$$

$$K^* = \frac{K}{K_o} = \frac{\rho}{\rho_o}\left[\frac{1+v}{1-v}\right]\left[\frac{1-v_o}{1+v_o}\right]V_L^{*2}. \quad (11)$$

Eq. (11) for bulk modulus, K, is derived from the relation $K = \frac{EG}{3(3G-E)}$. Subscript 'o' refers to the values of pore free or theoretically dense material.

Considering G^* first, Eq. (10) shows that the third term in square bracket on right hand side is constant for a material. Thus to express G^* in terms of V_L^* only, the variation of the second term in square bracket on the right side involving v as well as the variation of relative density with V_L^* must be known. Representing the second term by $\gamma(v)$, Eq. (10) becomes

$$G^* = \frac{\rho}{\rho_o}\frac{\gamma(v)}{\gamma(v_o)}V_L^{*2}. \quad (12)$$

Fig. 1 shows the variation $\gamma(v)$ with V_L^* for uranium dioxide and uranium nitride. These data were taken from Gatt et al. [6] and Panakkal [5] for uranium

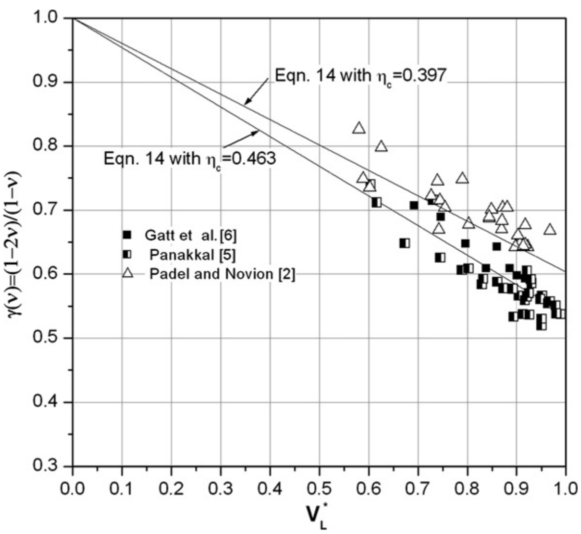

Fig. 1. Variation of $g(n)$ with V_L^* for uranium dioxide and uranium nitride.

dioxide and Padel et al. [2] for uranium nitride. Longitudinal velocity values, where not reported, were calculated from reported Young's modulus and corresponding Poisson's ratio values using Eq. (9) and values for pore free material are given in Table 1. Fig. 1 indicates that $\gamma(v)$ decreases monotonically with increasing V_L^* following a linear trend, which can be described by

$$\gamma(v) = \eta_o - \eta_c V_L^*, \tag{13}$$

where η_o and η_c are constants. The values of these constants are evaluated form the boundary conditions – $v = 0$ when $V_L^* = 0$ and $v = v_o$ when $V_L^* = 1$. Substituting these values in Eq. (13) yields $\eta_o = 1$ and $\eta_c = \frac{v_o}{1-v_o}$ and it reduces to

$$\gamma(v) = 1 - \eta_c V_L^*. \tag{14}$$

It may be noted η_c is a material specific constant depending only on v_o and its value works out to be 0.463 and 0.397 for UO_2 and UN respectively. Eq. (14) corresponding to these values is also shown in Fig. 1. Poisson's ratio is a small quantity which depends on the ratio of E to G and is more prone to errors at high porosities (low values of V_L^*) where both E and G are small than for low porosity data (high values of V_L^*). This sensitivity is magnified at low values of Poisson's ratio due to the subtraction of two almost-equal terms. Thus the calculated values show large scatter at low values of V_L^*. Considering the above facts the agreement between Eq. (14) and the measured values can be considered to be extremely good.

The dependence of the ultrasonic velocity on relative density is usually expressed in terms of porosity, p where

$$p = 1 - \frac{\rho}{\rho_o}. \tag{15}$$

It has been shown [10,16,17] that ultrasonic velocity variation with porosity can best be described by the relation,

$$V_L^* = (1-p)^n, \tag{16}$$

where the empirical constant n is dependent on micro-structural features like pore geometry and its orientation in the material [16]. For low porosities the equation can be approximated by the linear relation,

$$V_L^* = 1 - np. \tag{17}$$

Eq. (17) has been shown to describe the ultrasonic velocity variation with porosity in UO_2 quite well [18]. Combining Eqs. (15) and (17):

$$\frac{\rho}{\rho_o} = \frac{V_L^* + n - 1}{n}. \tag{18}$$

Combining Eqs. (12), (14) and (18)

$$G^* = \frac{(V_L^* + n - 1)(1 - \eta_c V_L^*)}{n\gamma(v_o)} V_L^{*2}. \tag{19}$$

Eq. (19) shows that once the variation of longitudinal velocity with relative density or porosity is known, the shear modulus can be calculated provided that the properties of pore free or theoretically dense material are known from single crystal data or experimental measurements. Using Eqs. (9), (11), (14) and (18) similar expressions can be derived for E^* and K^* as well

$$E^* = \frac{(V_L^* + n - 1)}{n} \times \frac{(1 - \eta_c V_L^*)(1 + 2\eta_c V_L^*)}{\gamma(v_o)(1 + \eta_c V_L^*)(1 + v_o)} V_L^{*2}, \tag{20}$$

$$K^* = \frac{(V_L^* + n - 1)}{n} \frac{(1 + 2\eta_c V_L^*)(1 - v_o)}{(1 + v_o)} V_L^{*2}. \tag{21}$$

An expression for Poisson's ratio, v, can be derived form Eq. (14):

$$v = \frac{\eta_c V_L^*}{1 + \eta_c V_L^*}. \tag{22}$$

Eqs. (19)–(22) provide the complete set of equations which can be used to determine the change in elastic properties from the measurement of longitudinal ultrasonic velocity once the unirradiated values are known. It may be noted that these equations along with Eq. (17) can also be used to describe the elastic properties variation with porosity.

Table 1
Elastic constants and ultrasonic velocity values of pore free/theoretically dense material

Material	ρ_0 (gm/cc)	V_{Lo} (m/s)	V_{So} (m/s)	E_o (GPa)	G_o (GPa)	K_o (GPa)	v_0	Remarks
UO_2	10.96	5469.00	2823.91	230.0	87.4	208.1	0.316	VRH values from single crystal data [4]
UN	14.30	4890.00	2695.5	266.8	103.9	205.9	0.284	[5]

4. Data analysis and discussion

An analysis of data for Uranium Dioxide reported by Gatt et al. [6], Panakkal [5], Roque et al. [10] and data for Uranium Nitride reported by Padel and Novion [2] are presented below in terms of Eqs. (17)–(22). In analyzing these data elastic constants and ultrasonic velocity values of pore free material as given in Table 1 have been used. Sum of squares, Q, has been used as a measure of goodness of fit between data and the fitted equation. Q is given by

$$Q = 1 - \frac{\sum_{i=1}^{m}(S_{ci} - S_i)^2}{\sum_{i=1}^{m}(S_i - \overline{S})^2}, \quad (23)$$

where S_{ci} is the calculated ith value from the fitted equation, S_i is the measured value, $\overline{S}$ is the mean of the measured values and m is number of data points. For values of $Q > 0.90$ the fit is considered to be good.

4.1. Uranium dioxide

Fig. 2 shows the longitudinal ultrasonic velocity variation of irradiated UO_2 with porosity as reported by Gatt et al. (refer Fig. 12 in [6]). These values were measured using acoustic microscopy and micro-echography. Gatt et al. [6] have not reported velocity values directly. They were evaluated from the Young's and shear modulus data reported by them using the relations

$$E = \frac{3V_L^2 - 4V_S^2}{V_L^2 - V_S^2}\rho V_S^2, \quad (24)$$

$$G = \rho V_S^2. \quad (25)$$

The plot indicates a linear variation of velocity (unnormalised) with porosity and Eq. (17) was fitted to the data by regression analysis giving

$$V_L = 5469(1 - 1.701p)(\text{m/s}), \quad (26)$$

having $Q = 0.930$, indicating a good fit. Also plotted in Fig. 2 the longitudinal velocity data reported by three other researchers namely Roque et al. [10], Panakkal and Ghosh [7] and Boocock et al. [3]. In all cases data seem to agree with Eq. (26) quite well. This also possibly indicates the similarity in the micro-structure of the material investigated by these researchers.

Fig. 3 shows the change of the normalized shear modulus with V_L^* as reported by Gatt et al. (refer Fig. 12 and Eqs. (28) and (29) in [6]). Eq. (19) is also plotted in Fig. 3 with $n = 1.701$ (as obtained from Eq. (26)) giving $Q = 0.947$. The estimated values of G^* from V_L^* using Eq. (19) lie within ±6% of the measured values, which is good for a quantitative evaluation method. The normalized values of shear moduli as reported by Boocock et al. [3] have also been shown in Fig. 3. It again shows an excellent agreement with Eq. (19) indicating that velocity–porosity relation as given by Eq. (26) is equally applicable to his data. A more sensitive test for the validation of the correlation for Poisson's ratio given by Eq. (14), is a comparison between

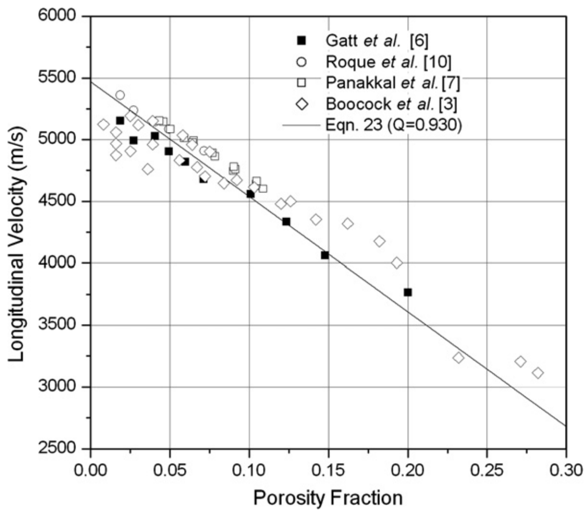

Fig. 2. Variation of V_L with porosity for irradiated UO_2.

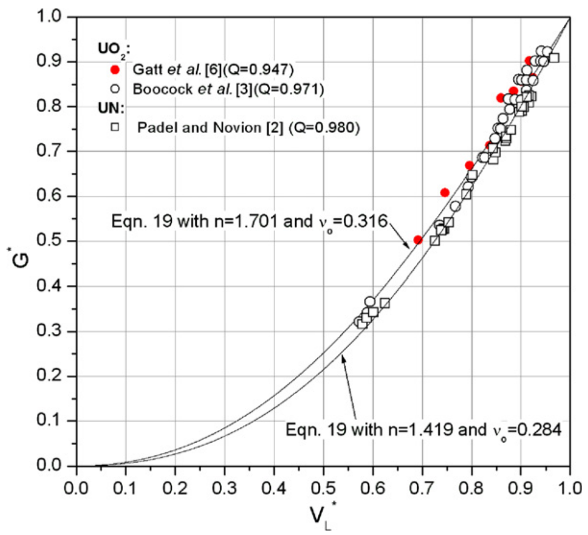

Fig. 3. Variation of G^* with V_L^* for UO_2 and UN.

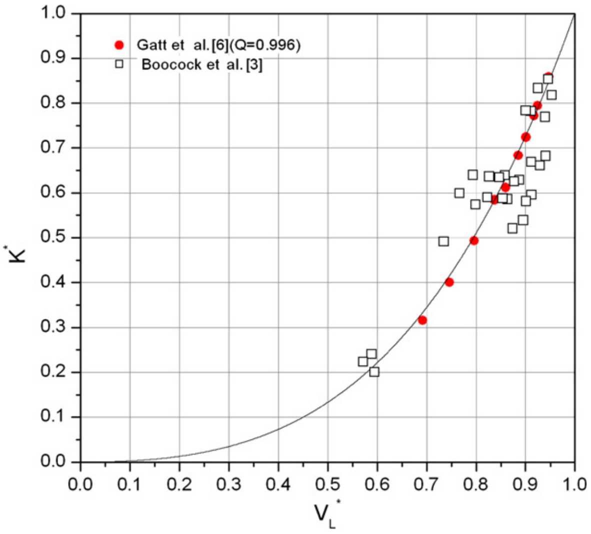

Fig. 4. Variation of K^* with V_L^* for UO_2.

the normalized bulk modulus K^* calculated from the measured values of E^* and G^* with the values predicted from Eq. (21). Fig. 4 shows K^* values for UO_2, estimated from the corresponding values of E^* and G^* plotted against normalized velocity, V_L^* for the data reported by Gatt et al. [6] and Boocock et al. [3] along with the plot of Eq. (21). The agreement between the data reported by Gatt et al. [6] and Eq. (21) speaks for itself having a value of $Q = 0.996$. For Boocock et al.'s data the closeness of the calculated points to the theoretical curve is remarkable considering the sensitivity of these calculations to the propagation of experimental errors in measurement of E and G. Eq. (20) along with Eq. (17) can be used to predict the variation of Young's modulus with porosity. This is shown in Fig. 5 where the predicted values have been compared with the data reported by Boocock et al. [3], Padel and Novion [2] and Gatt et al. [6]. Experimental values show quite a good agreement with the predicted ones with the value of $Q = 0.920$ for the data reported by Padel and Novion [2] and Gatt et al. [6].

4.2. Uranium nitride

The ultrasonic longitudinal velocity versus porosity data for UN as reported by Padel and Novion [2] is shown in Fig. 6. Eq. (17) was fitted to the data using pore free material velocity value as given in Table 1 yielding the relation

$$V_L = 4890(1 - 1.419p). \quad (27)$$

As can be seen from the figure, Eq. (27) agrees with the experimental data well having $Q = 0.934$. Also plotted in Fig. 6, ultrasonic longitudinal velocity values of UN reported by Whaley et al. [19] for 92.8%, 95%, 97% and 98.5% dense samples. These data points closely follow Eq. (27). As before, shear and bulk moduli were calculated from Eqs. (19) and (21) respectively using $n = 1.419$ and are plotted in Figs. 3 and 7 respectively. In both cases measured experimental values show close agreement with the theoretically predicted values. Whaley et al. [19] have reported Young's and shear moduli values of

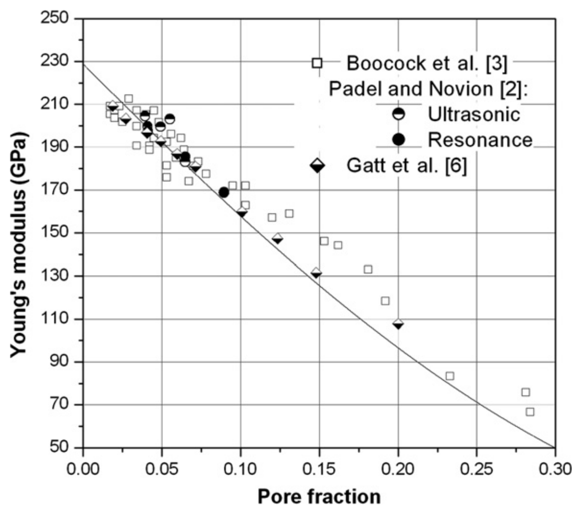

Fig. 5. Variation of E with porosity for UO_2.

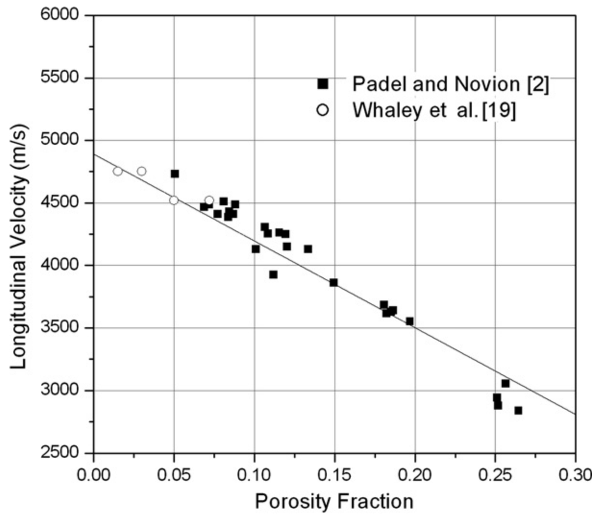

Fig. 6. Variation of V_L with porosity for UN.

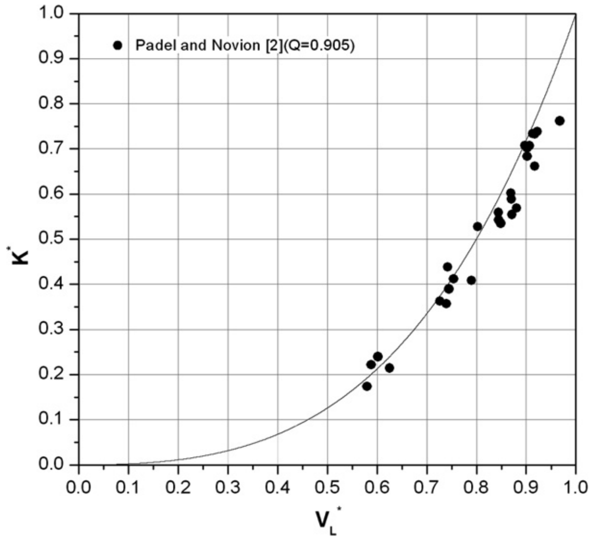

Fig. 7. Variation of K^* with V_L^* for UN.

polycrystalline UN as reported by several investigators [20–22] for specimens having different pore fraction. These values are shown in Fig. 8. To compare them with the present theory Eqs. (19) and (20) are also plotted in the figure with V_L^* replaced by Eq. (27) so that E and G can be expressed as a function of porosity fraction. The agreement between the two can be considered extremely good considering the fact data source in each case is different.

It may be noted that all the predicted elastic properties have been calculated based on the assumption that the Poisson's ratio velocity relation

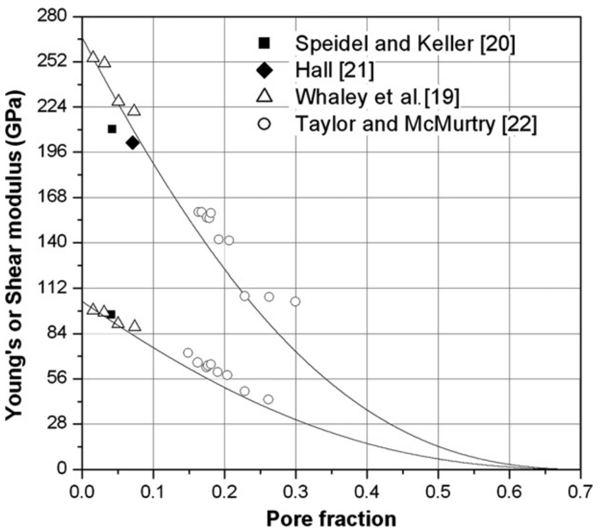

Fig. 8. Variation of E and G with porosity for UN.

given by Eq. (14) is true. Thus the close agreement between the predicted elastic properties and the experimental values confirms the validity of the proposed relation.

5. Conclusion

A new correlation between Poisson's ratio and ultrasonic longitudinal velocity has been proposed and shown to agree with the experimental data on uranium dioxide and uranium nitride extremely well. The elastic property values predicted based on this equation using physical acoustics theory agrees with the experimental data within ±6%. Considering the error involved in experimental determination of these elastic moduli, the fit can be considered quite good for the purpose of quantitative non-destructive evaluation of elastic properties. Moreover, it is established that ultrasonic longitudinal velocity solely can be used for the determination of elastic properties of fuel material.

Acknowledgements

The first two authors would like to acknowledge the financial support received from Board of Research in Nuclear Sciences, India for this work. The authors also thank Director, CG&CRI for his permission to publish this paper.

References

[1] V. Roque, B. Cros, D. Baron, P. Dehaudt, J. Nucl. Mater. 277 (2000) 211.
[2] A. Padel, C.D. Novion, J. Nucl. Mater. 33 (1969) 40.
[3] J. Boocock, A.S. Furzer, J.R. Mathews, AERE Report no. 2565, 1972.
[4] J.B. Wachtman, M.L. Wheat, H.J. Anderson, J.L. Bates, J. Nucl. Mater. 16 (1965) 39.
[5] J.P. Panakkal, IEEE Trans. Ferroelectr. Freq. Control 38 (1991) 161.
[6] J.-M. Gatt, Y. Monerie, D. Laux, D. Baron, J. Nucl. Mater. 336 (2005) 145.
[7] J.P. Panakkal, J.K. Ghosh, J. Mater. Sci. Lett. 3 (1985) 835.
[8] J.P. Panakkal, J.K. Ghosh, P.R. Roy, J. Phys. D: Appl. Phys. 17 (1984) 1791.
[9] A.P. Roberts, M. Teubner, Phys. Rev. E 51 (1995) 4141.
[10] V. Roque, D. Baron, J. Bourgoin, J.-M. Saurel, J. Nucl. Mater. 275 (1999) 305.
[11] J.P. Panakkal, H. Willems, W. Arnold, J. Mater. Sci. 25 (1990) 1397.
[12] O. Yeheskel, O. Tevet, J. Test. Eval. 28 (2000) 189.
[13] O. Yeheskel, J. Test. Eval. 32 (2004) 17.
[14] G. Ondracek, Z. Werkstofftech 9 (1978) 31.
[15] Y.H. Zhao, G.P. Tandon, G.J. Weng, Acta Mech. 76 (1989) 105.

[16] A.K. Maitra, K.K. Phani, J. Mater. Sci. 29 (1994) 4415.
[17] L.P. Martun, D. Dadon, M. Rosen, J. Am. Ceram. Soc. 79 (1996) 1281.
[18] K.K. Phani, S.K. Niyogi, J. Mater. Sci. Lett. 5 (1986) 427.
[19] H.L. Whaley, W. Fulkerson, R.A. Potter, J. Nucl. Mater. 31 (1969) 345.
[20] E. Speidel, D.K. Keller, Battelle Memorial Institute (USA) Report, BMI - 1633 (1963).
[21] A.R. Hall, UKAEA Report, AERE-R5650 (1967).
[22] K. Taylor, C. McMurtry, Oak Ridge Operations (USA) Report, ORO-400 (1961).