USEFUL RELATIONSHIPS BETWEEN DIELECTRIC PROPERTIES AND BULK DENSITIES OF GRANULAR AND POWDERED MATERIALS

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Abstract. When knowledge of the dielectric properties, or permittivity, of granular or powdered materials is important, as in dielectric heating applications or in the sensing of moisture content by radio-frequency or microwave instruments, some reliable relationship between the dielectric properties and the bulk density of the material is required. Linear relationships between functions of the permittivity and bulk density have been observed, and dielectric mixture equations have been explored to obtain useful relationships for determining the dielectric properties of granular and powdered materials. These findings are reviewed, useful permittivity-density relationships are identified, and an equation is provided that permits reliable calculation of the permittivity of a particulate material at any given bulk density, if the permittivity is known or measured at another known bulk density. The relationships are illustrated for pulverized coal, wheat, ground wheat, limestone, and a coal-limestone mixture. The useful mixture equation is valid for granular and powdered materials with permittivities in the same range as those for grain, coal and limestone.

Introduction

When the permittivity, or dielectric properties, of granular or powdered solid materials are important, some reliable relationship between the permittivity and the bulk density of the air-particle mixture is required. Examples of such applications include radio-frequency or microwave dielectric heating of these materials and the use of correlations between permittivity and water content of hygroscopic materials for sensing moisture content. Also, relationships between the permittivities of pulverized or granular samples of materials and those of the solid materials have long been of interest for use in determining the permittivities of the solids from measured permittivities of the particulate samples. Dielectric mixture equations have been developed and investigated for this purpose [1-3]. An extrapolation procedure, based on the linearity with density of functions of the real and imaginary parts of the permittivity of pulverized and granular materials, has also been used to obtain estimates of the permittivity of the solid material [4, 5]. Both of these methods, calculation by dielectric mixture equations and the extrapolation procedure, will be described in the following sections for estimating the permittivity of solids from measured permittivity data on granular or powdered samples of those materials.

Dielectric Mixture Equations

Many different dielectric mixture equations have been proposed to represent the effective permittivity of mixtures of dielectric materials, and the characteristics and performance of these equations for various applications have been discussed in the literature [6-11]. The complex permittivity relative to free space is represented here as $\varepsilon = \varepsilon' - j\varepsilon''$, where ε' is the dielectric constant and ε'' is the dielectric loss factor. Dielectric mixture equations for a two-phase mixture can be used to calculate the dielectric properties of a solid material from the dielectric properties of an air-particle mixture of air and pulverized or granular particles of the solid. To use the mixture equations, one needs to know the permittivity of the pulverized sample at its bulk density (air-particle mixture density), ρ, and the specific gravity or density of the solid material, ρ_2. The fractional part of the total volume of the mixture occupied by the particles (volume fraction), v_2, is then given by ρ / ρ_2.

Several well-known dielectric mixture equations have been studied to relate the permittivities of the solid material to the permittivities of granular or pulverized samples of the same material [5, 12-15]. Two of these equations are the Complex Refractive Index mixture equation [16-22],

$$(\varepsilon)^{1/2} = v_1(\varepsilon_1)^{1/2} + v_2(\varepsilon_2)^{1/2} \tag{1}$$

and the Landau & Lifshitz, Looyenga mixture equation [23, 24],

$$(\varepsilon)^{1/3} = v_1(\varepsilon_1)^{1/3} + v_2(\varepsilon_2)^{1/3} \tag{2}$$

where ε represents the complex permittivity of the mixture, ε_1 is the permittivity of the medium in which particles of permittivity ε_2 are dispersed, and v_1, and v_2 are the volume fractions of the respective components, where $v_1 + v_2 = 1$.

Each of these equations can be used to calculate the complex permittivity of the solid material by substituting $1 - j0$ for ε_1, the complex permittivity of air, and solving for ε_2. The corresponding equations are:

Complex Refractive Index (1)

$$\varepsilon_2 = \left(\frac{\sqrt{\varepsilon} + v_2 - 1}{v_2} \right)^2 \tag{3}$$

Landau & Lifshitz, Looyenga (2)

$$\varepsilon_2 = \left(\frac{\varepsilon^{1/3} + v_2 - 1}{v_2} \right)^3 \tag{4}$$

Linear Extrapolation Technique

Essentially linear relationships between functions of the real and imaginary components of the complex permittivity of particulate materials such as pulverized coal, wheat, and whole-wheat flour and their bulk densities were identified previously [4, 25, 26]. These findings were based on earlier work in which Klein [27] observed the linearity of $\sqrt{\varepsilon'}$ with the bulk density of granular coal, and in which Kent [28], working with fish meal, found that ε' and ε'' were both quadratic functions of the bulk density of the fish meal as follows:

$$\varepsilon' = a\rho^2 + b\rho + 1 \tag{5}$$

$$\varepsilon'' = c\rho^2 + d\rho \tag{6}$$

where ρ represents the density of the air-particle mixture, a, b, c and d are constants for a given particulate material and ε' and ε'' have values of *1* and *0*, respectively, for air alone (ρ = 0). Equations (5) and (6) are equivalent to expressing the relative complex permittivity as a quadratic function of bulk density, $\varepsilon = a^* \rho^2 + b^* \rho + 1$, where the constants $a^* = a - jc$ and $b^* = b - jd$ are complex numbers.

The linearity of $(\varepsilon')^{1/2}$ with bulk density was confirmed with data on pulverized coal (Fig. 1) [25] and this relationship is expressed as

$$(\varepsilon')^{1/2} = m\rho + 1 \tag{7}$$

Equations (5) and (7) are equivalent if $a = m^2$ and $b = 2m$. Thus, measurement of the dielectric constant of a particulate material at one bulk density, along with the $(\rho = 0, \varepsilon' = 1)$ intercept, provides information on the dielectric constant at all densities, including that of the solid material if its density is known. Examining the expression for the loss factor (6), the square can be completed by adding a constant, e, to each side, and a linear function is obtained

$$(\varepsilon'' + e)^{1/2} = c^{1/2}\rho + e^{1/2} \tag{8}$$

where $e = d^2 / 4c$. Thus, to describe the density dependence of the complex permittivity of a particulate material, one needs to obtain the values for $a, b, c,$ and d. Measurement of the permittivity of the particulate material at a given density establishes slope m of (7), thus determining values for a and b. Measurements at a few additional densities are necessary for determination of c and d, as explained previously [12, 29]. It is interesting to note that (5) and (6) are consistent with the Complex Refractive Index mixture equation (1), when used for the real part of the complex permittivity. Since $v_2 = \rho / \rho_2$, where ρ is the density of the air-particle mixture, and ρ_2 is the density of the particles, this substitution in (1) yields the following for an air-particle mixture:

$$(\varepsilon')^{1/2} = \frac{(\varepsilon_2')^{1/2} - 1}{\rho_2}\rho + 1 \tag{9}$$

which is equivalent to (7), where $m = (\sqrt{\varepsilon_2'} - 1)/\rho_2$. In an analogous manner, it can be shown that the linearity of the cube root of the dielectric constant of an air-particle mixture with its density is consistent with the Landau and Lifshitz, Looyenga mixture equation (2) [4],

$$(\varepsilon')^{1/3} = \frac{(\varepsilon_2')^{1/3} - 1}{\rho_2}\rho + 1 \tag{10}$$

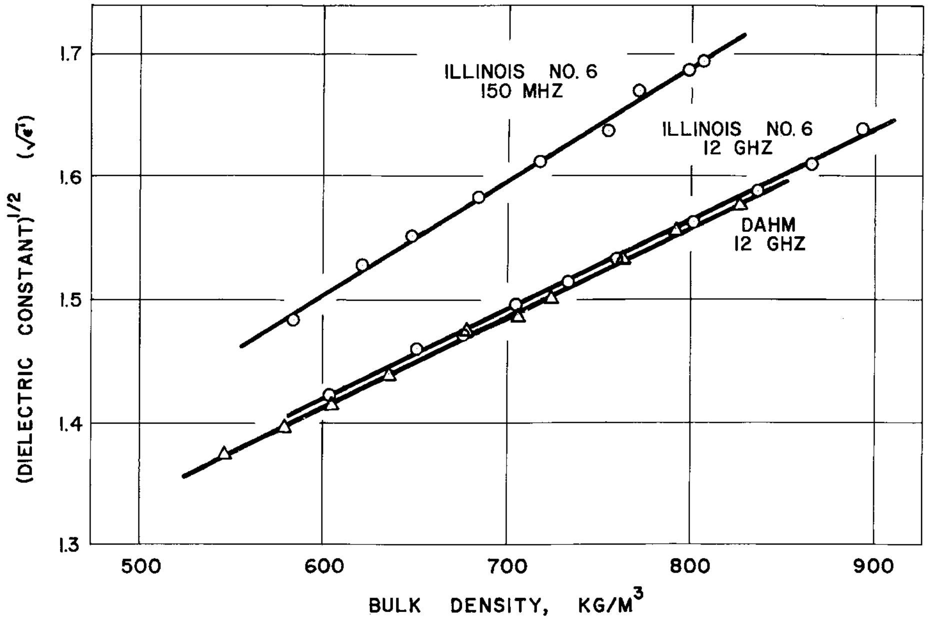

Fig. 1. Linear relationships between the square root of the dielectric constant of pulverized coal samples and the bulk density measured at indicated frequencies [25].

Experimental Data for Wheat

Both components of the permittivity are shown in Fig. 2 for a measurement sequence at different densities for whole-wheat flour of 10.9% moisture at 11.67 GHz [26], illustrating the quadratic behavior for both the dielectric constant and loss factor in accordance with (5) and (6). The same data are presented in Fig. 3, showing the linearity of the square roots and the cube roots of the dielectric constant with bulk density as defined in (7), (9), and (10). The linearity of the square root of the loss-factor function is also illustrated for these data in Fig. 4.

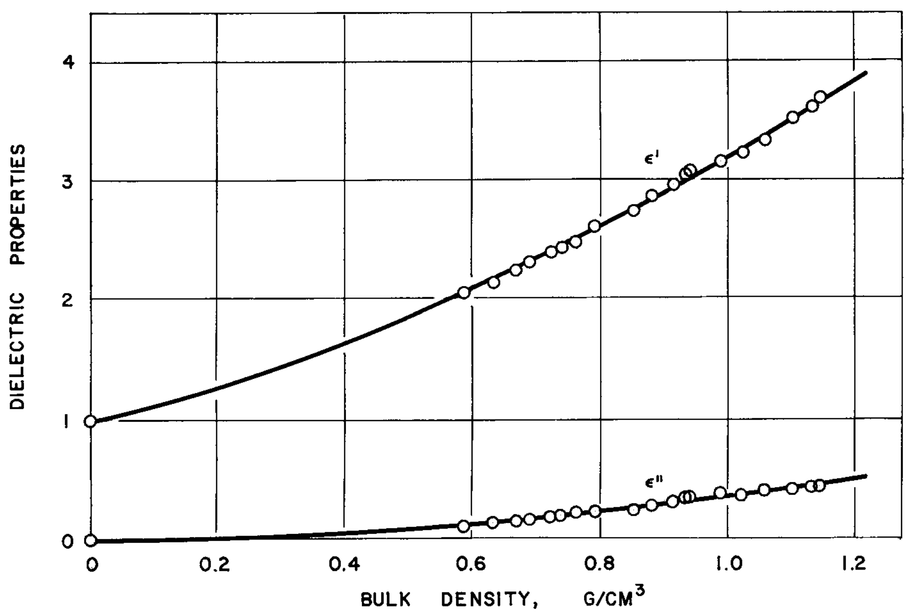

Fig. 2. Dielectric constant and loss factor of whole-wheat flour of 10.9% moisture at 11.67 GHz as functions of the bulk density of the flour [26].

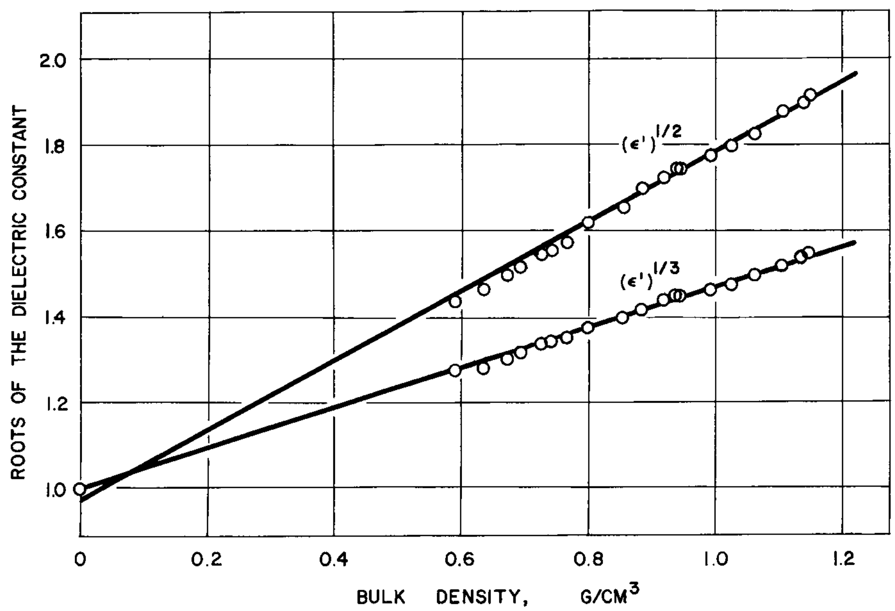

Fig. 3. Straight-line relationship between the square root and cube root of the dielectric constant of whole-wheat flour at 10.9% moisture and 11.67 GHz and the bulk density of the flour [26].

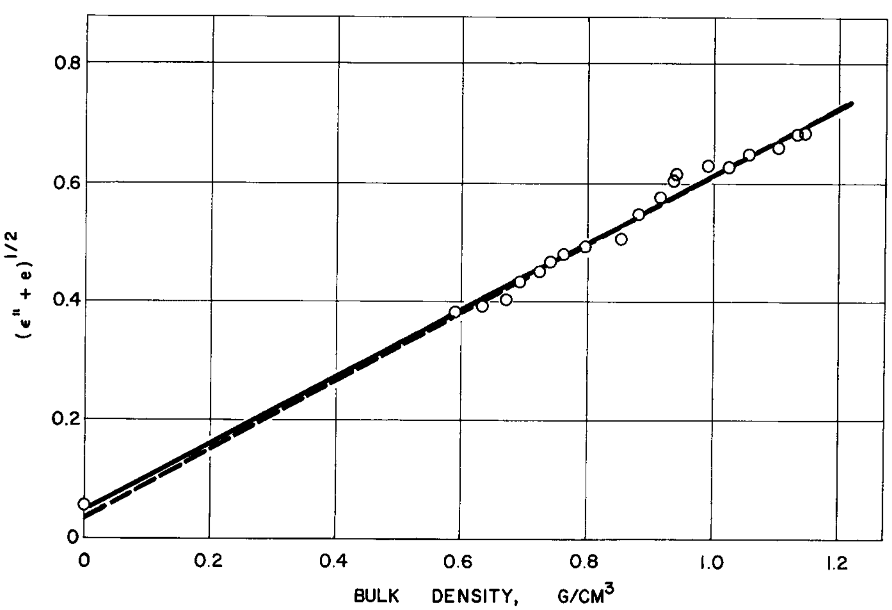

Fig. 4. Straight-line relationship between the function of the loss factor of whole-wheat flour of 10.9% moisture at 11.67 GHz and the bulk density of the flour. Solid line is the regression line with the $(0,\sqrt{e})$ intercept included, and the dashed line is the regression line with the intercept excluded from the data set [26].

As shown in Fig. 3, the cube-root relationship of (10) provides a slightly better fit than the square-root relationship of (9).

Results of similar sequences of measurements over a range of densities are shown in Figs. 5 and 6 for whole-wheat flour (ground wheat) and whole-kernel hard red winter wheat, respectively. These were different wheat samples and the measurements were taken at different frequencies. However, for these two different particle sizes of similar material, results showed that both the square root and cube root relationships fit the data reasonably well. The cube-root relationship, however, was a bit superior to the square-root relationship in fitting the

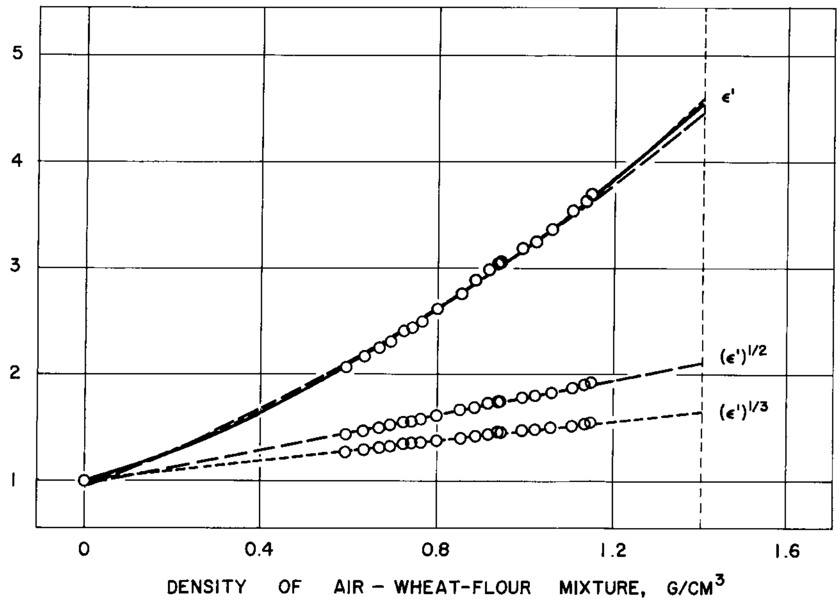

Fig. 5 Density dependence of the dielectric constant and its square root and cube root for whole-wheat flour at 11.7 GHz and 22 °C. Solid lines are regression lines for second-order polynomial regression for the dielectric constant and linear regression for the square root and cube root. Short- and long-dashed lines for the dielectric constant are predictions from the regression of the cube root and square root of the dielectric constant, respectively [4].

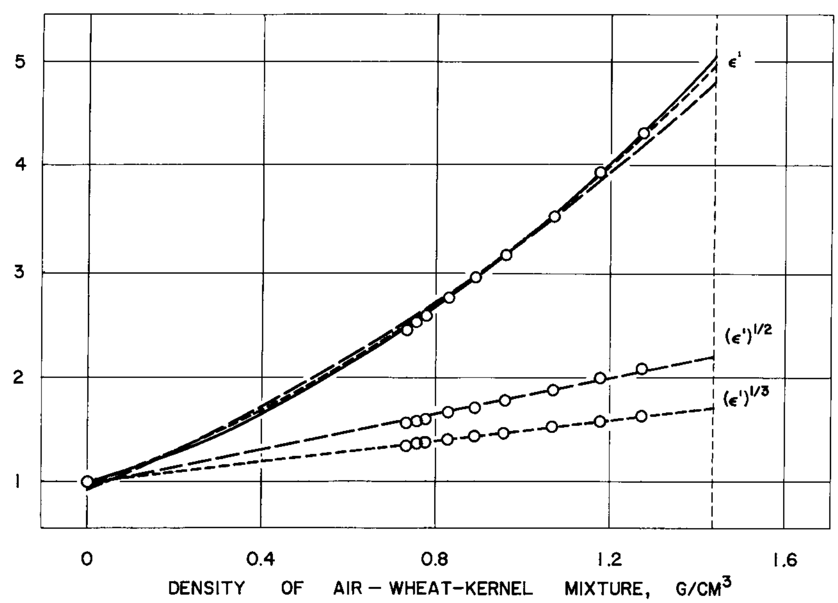

Fig. 6. Density dependence of the dielectric constant and its square root and cube root for whole-kernel wheat at 9.4 GHz and 24 °C. Solid lines are regression lines for second-order polynomial regression for the dielectric constant and linear regression for the square root and cube root. Short- and long-dashed lines for the dielectric constant are predictions from the regression of the cube root and square root of the dielectric constant, respectively [4].

data for both the whole-kernel wheat and the ground wheat. In these studies, the permittivity data were extrapolated to the densities of the flour and grain particle densities, as determined by air comparison pycnometer measurements [26], to obtain estimates of the permittivities of the solid materials at their densities as indicated by the vertical dashed lines in Figs. 5 and 6.

Experimental Data for Coal and Limestone

Results of permittivity measurements and sample bulk density determinations on pulverized samples of coal, limestone and a 35%-65% coal-limestone mixture [30] are shown for the dielectric constant ε' in Fig. 7, where the linearity with bulk density of the cube root of the dielectric constant of an air-particle mixture, as noted previously [4, 13], is consistent with the Landau & Lifshitz, Looyenga dielectric mixture equation (2).

The necessary value for v_s can be obtained if the bulk density ρ of the mixture and the density ρ_s of the solid particulate material are known, since $v_s = \rho / \rho_s$. For such linear relationships between the cube roots of the dielectric constants and the bulk density, The Landau & Lifshitz, Looyenga equation can be used with confidence in calculating the complex permittivity of the solid material from the permittivities of the powdered samples [31]. The mean values of the solid material permittivities calculated with the Landau & Lifshitz, Looyenga mixture equation for the permittivity measurements at each bulk density were 4.21– *j*0.156 for the coal and 7.41– *j*0.063 for the limestone used in these measurements. The respective solid-material densities for the coal and limestone from air-comparison-pycnometer measurements were 1.48 and 2.75 g/cm^3 [31].

The performance of (1) and (2) were compared for calculating the complex relative permittivity values for the pure coal, pure limestone, and a 35%-65% coal-limestone mixture, and calculated results were compared with measured values. Equations (1) and (2) require only the permittivities of the solid-material constituents of the air particle mixture and the volume fractions occupied by each constituent. Permittivities of the coal and limestone particles are 4.21 – *j*0.156 and 7.41 – j 0.063, respectively, as already mentioned and the volume fractions were determined as $v_s = \rho / \rho_s$.

Comparisons of the calculated and measured permittivities for the coal, limestone, and 35%-65% coal-limestone mixture revealed that for the coal samples, the Landau & Lifshitz, Looyenga Equation (2) estimated the measured permittivity values very well. The dielectric constant was given extremely well by the computation, and the loss factor values were also good. The Complex Refractive Index Equation (1) overestimated the permittivity with an error of about 4% for the dielectric constant and larger errors for the loss factor.

For the limestone samples, which have higher dielectric constants and lower loss factors than the coal, (2) again provided much closer estimates than (1), with (1) overestimating the dielectric constant by about 10%. As expected, the performance of both equations in estimating measured properties of the 35%-65% coal-limestone mixture, was intermediate between their performance on the coal and limestone samples.

Over the full range of densities measured for each material, the error of prediction for the Landau & Lifshitz, Looyenga Equation ranged from 0 to 0.6% for the dielectric constant of the powdered coal with an average of 0.15%, from 0 to 2.9% for the powdered limestone with an average of 1.4%, and from 0 to 1.2% for the 35%-65% powdered coal-limestone mixture with an average of 0.5%. Corresponding errors of prediction for the Complex Refractive Index Equation ranged as high as 4.6%, 10.2% and 7.2% for the coal, limestone, and 35%-65% coal-limestone mixture, respectively.

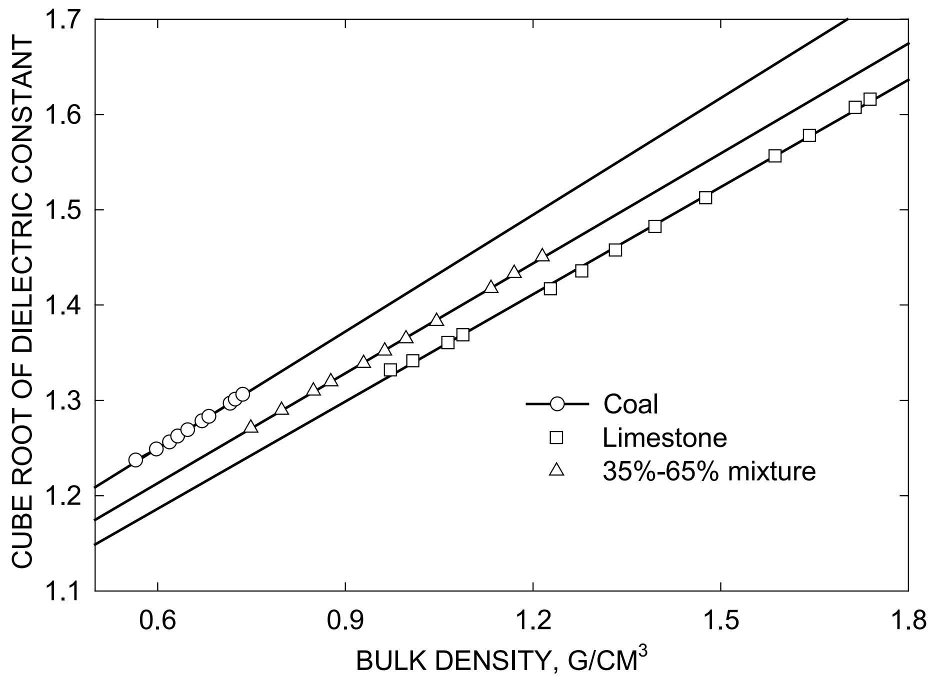

Fig. 7. Linear relationships between the dielectric constants of pulverized coal and limestone and a 35%-65% coal-limestone mixture and bulk densities of the samples at 11.7 GHz and 20 °C [32].

Discussion and Conclusions

Applications of these techniques have been discussed in detail for coal [4, 5, 13, 25, 29], wheat and wheat flour [4, 14, 15, 26], plastics [12, 13], and coal and limestone and their mixtures [31, 32]. As a result of these studies, the Landau & Lifshitz, Looyenga equation generally provided the best relationship between the permittivities of the solid and particulate materials. Therefore, a relationship based on this equation can be recommended for correcting permittivities of granular and powdered materials, with permittivities in the same range as these materials, for changes in bulk density as follows:

$$\varepsilon_b = \left[\left((\varepsilon_a)^{1/3} - 1\right)\rho_b / \rho_a + 1\right]^3 \qquad (11)$$

where ε_a is the complex permittivity of the air-particle mixture at a given density ρ_a, and ε_b is the permittivity of the mixture at a different density ρ_b. Use of complex calculation provides values for both the dielectric constant and loss factor.

References

[1] C. J. F. Bottcher, "The dielectric constant of crystalline powders.," *Recueil des Travaux Chimiques des Pays-Bas.*, vol. 64, pp. 47-51, 1945.

[2] D. C. Dube, "Study of Landau-Lifshitz-Looyenga's formula for dielectric correlation between powder and bulk," *Journal of Physics D: Applied Physics*, vol. 3, pp. 1648-1652, 1970.

[3] D. C. Dube, R. S. Yadava, and R. Parshad, "A formula for correlating dielectric constant of powder & bulk," *Indian Journal of Pure & Applied Physics*, vol. 9, pp. 719-721, 1971.

[4] S. O. Nelson, "Observations on the density dependence of the dielectric properties of particulate materials," *Journal of Microwave Power*, vol. 18, no.2, pp. 143-152, 1983.

[5] S. O. Nelson, "Use of dielectric mixture equations for estimating permittivities of solids from data on pulverized samples," in *Physical Phenomena in Granular Materials*, vol. 195, *Materials Research Society Symposium Proceedings*, G. D. Cody., T. H. Geballe, and P. Sheng, Eds. Pittsburgh, PA: Materials Research Society, 1990, pp. 295-300.

[6] L. K. H. Van Beek, "Dielectric behavior of heterogeneous systems," in *Progress in Dielectrics*, vol. 7, J. B. Birks, Ed. London: Heywood Books, 1967, pp. 69-114.

[7] W. R. Tinga, "Mixture laws and microwave-material interactions," in *Dielectric Properties of Heterogeneous Materials*, vol. Pier 6, *Progress in Electromagnetics Research*, A. Priou, Ed. New York, Amsterdam, London, Tokyo: Elsevier, 1992, pp. 1-40.

[8] G. Banhegyi, "Comparison of electrical mixture rules for composites," *Colloid & Polymer Science*, vol. 264, pp. 1030-1050, 1986.

[9] A. Sihvola, *Electromagnetic Mixing Formulas and Applications*. London, UK: Institution of Electrical Engineers, 1999.

[10] C. Grosse and J. L. Greffe, "Permittivite statique des emulsions," *Journal de Chimie Physique*, vol. 76, no.4, pp. 305-327, 1979.

[11] J. L. Greffe and C. Grosse, "Static permittivity of emulsions," in *Dielectric Properties of Heterogeneous Materials*, vol. 6, *Progress in Electromagnetics Research*, A. Priou, Ed. New York, Amsterdam, London, Tokyo: Elsevier, 1992, pp. 41-100.

[12] S. O. Nelson and T.-S. You, "Relationships between microwave permittivities of solid and pulverised plastics," *Journal of Physics D: Applied Physics*, vol. 23, pp. 346-353, 1990.

[13] S. O. Nelson, "Estimation of permittivities of solids from measurements on pulverized or granular materials," in *Dielectric Properties of Heterogeneous Materials*, vol. PIER 6, *Progress in Electromagnetics Research*, A. Priou, Ed. New York, Amsterdam, London, Tokyo: Elsevier, 1992, pp. 231-271.

[14] S. O. Nelson, "Correlating dielectric properties of solids and particulate samples through mixture relationships," *Transactions of the ASAE*, vol. 35, no.2, pp. 625-629, 1992.

[15] S. O. Nelson, "Dielectric properties of agricultural products -- Measurements and Applications," *IEEE Transactions on Electrical Insulation*, vol. 26, no.5, pp. 845-869, 1991.

[16] A. Beer, *Einleitung in die Hohere Optik*. Braunschweig: Druck und Verlag von Friedrich Vieweg und Sohn. p. 35, 1853.

[17] J. C. Philip, "Das dielectrische Verhalten flussiger Mischungen, besonders verdunnter Losungen," *Zeitschrift fur Physikalische Chemie*, vol. 24, pp. 18-38, 1897.

[18] H. H. Lowry, "The significance of the dielectric constant of a mixture," *Journal of the Franklin Institute*, vol. 203, pp. 413-439, 1927.

[19] J. R. Birchak, C. G. Gardner, J. E. Hipp, and J. M. Victor, "High dielectric constant microwave probes for sensing soil moisture," *Proceedings of the IEEE*, vol. 62, no.1, pp. 93-98, 1974.

[20] A. Kraszewski, "Prediction of the dielectric properties of two-phase mixtures," *Journal of Microwave Power*, vol. 12, no.3, pp. 216-222, 1977.

[21] P. N. Sen, C. Scala, and M. H. Cohen, "A self-similar model for sedimentary rocks with application to the dielectric constant of fused glass beads," *Geophysics*, vol. 46, no.5, pp. 781-795, 1981.

[22] L. C. Shen, W. C. Savre, M. M. Price, and K. Athavael, "Dielectric properties of reservoir rocks at ultra-high frequencies," *Geophysics*, vol. 50, no.4, pp. 692-704, 1985.

[23] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media, pp. 45-47*, vol. 8: Pergamon Press, Oxford, England, 1960.

[24] H. Looyenga, "Dielectric constants of heterogeneous mixtures," *Physica*, vol. 31, pp. 401-406, 1965.

[25] S. O. Nelson, S. R. Beck-Montgomery, G. E. Fanslow, and D. D. Bluhm, "Frequency dependence of the dielectric properties of coal -- Part II," *Journal of Microwave Power*, vol. 16, no.3&4, pp. 319-326, 1981.

[26] S. O. Nelson, "Density dependence of the dielectric properties of wheat and whole-wheat flour," *Journal of Microwave Power*, vol. 19, no.1, pp. 55-64, 1984.

[27] A. Klein, "Microwave determination of moisture in coal -- comparison of attenuation and phase measurement," *Journal of Microwave Power*, vol. 16, no.3&4, pp. 289-304, 1981.
[28] M. Kent, "Complex permittivity of fish meal: a general discussion of temperature, density, and moisture dependence," *Journal of Microwave Power*, vol. 12, no.4, pp. 341-345, 1977.
[29] S. O. Nelson, "Estimating the permittivity of solids from measurements on granular or pulverized materials," in *Microwave Processing of Materials*, vol. 124, *Materials Research Society Symposium Proceedings*, W. H. Sutton, M. H. Brooks, and I. J. Chabinsky, Eds. Pittsburgh, PA: Materials Research Society, 1988, pp. 149-154.
[30] S. O. Nelson, "Measurement and calculation of powdered mixture permittivities," *IEEE Transactions on Instrumentation and Measurement*, vol. 50, no.5, pp. 1066-1070, 2001.
[31] S. O. Nelson, "Determining dielectric properties of coal and limestone by measurements on pulverized samples," *Journal of Microwave Power and Electromagnetic Energy*, vol. 31, no.4, pp. 215-220, 1996.
[32] S. O. Nelson, "Measurement and calculation of powdered mixture permittivities," *IEEE Transactions on Instrumentation and Measurement*, vol. 50, no.5, pp. 1066-1070, 2001.