Measurement and Calculation of Powdered Mixture Permittivities

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***Abstract*—The permittivities of pulverized samples of coal and limestone were measured over a range of bulk densities at 11.7 GHz and 20 °C. The measured values were used, along with particle densities, determined by pycnometer measurements, and the Landau & Lifshitz, Looyenga (LLL) dielectric mixture equation to determine the solid material permittivities. Subsequently, similar measurements were taken on a 35%–65% coal–limestone mixture, and the LLL equation and the complex refractive index (CRI) dielectric mixture equation were used to calculate the permittivities of powdered coal, limestone, and the 35%–65% mixture at various bulk densities. Results compared to the measured permittivities showed that the LLL equation provided much better estimates than the CRI equation, with errors of 0.15% in predicting the dielectric constants of powdered coal, 1.4% in predicting the dielectric constants of powdered limestone, and 0.5% in predicting the dielectric constants of the coal–limestone mixture.**

***Index Terms*—Coal, dielectric mixture equations, dielectric properties, limestone, permittivity measurements, powders.**

I. INTRODUCTION

OFTEN there is need for reliable relationships between the permittivities of solid and powdered materials. For some materials, machining of samples to exact dimensions required for permittivity measurements is difficult, and measurements on pulverized samples are more easily performed. In some instances, as with mineral deposits in rocks, the materials must be pulverized for purification [1], and in other instances, the granular nature of the material makes measurements of the solid particles impractical [2], [3]. In these situations, the permittivities of the powdered or granular material can be measured, and the permittivities or dielectric properties of the solid materials can be closely estimated with the use of applicable dielectric mixture equations [4], [5].

In this paper, measurements of the permittivities ($\epsilon = \epsilon' - j\epsilon''$, where ϵ' is the dielectric constant and ϵ'' is the loss factor) of powdered coal and powdered limestone at microwave frequencies are described, and methods for determining the permittivities of the solid material are given. Use of appropriate dielectric mixture equations requires the determination of the material density, which is also described. From the bulk density of the powdered materials and the particle density, the volume fractions of the mixture components can be determined for use in dielectric mixture equations for calculation of the solid material permittivities.

Measurements on powdered mixtures of coal and limestone have been studied for potential use in rapidly measuring the proportions of limestone (rock dust) in coal mines for safety applications [6]. Federal regulations require that the noncombustible content of dust in coal mines must be maintained at levels of at least 65% for prevention of explosions. Therefore, rock dust is routinely distributed in coal mines to meet these requirements. Research has shown that proportions of limestone in coal–limestone dust mixtures can be effectively determined by measurements in a microwave resonant cavity, based on differences in both the dielectric constant and the dielectric loss factor between coal and limestone [7].

In connection with these studies, the permittivities of pulverized samples of pure coal, pure limestone, and a 35%–65% coal–limestone mixture, by weight, were measured over a range of bulk densities from loosely to tightly packed. Based on permittivities of coal and limestone and experimental determination of the densities, the expected permittivities of the powdered samples were calculated according to the complex refractive index (CRI) and the Landau & Lifshitz, Looyenga (LLL) mixture equations, and then compared with the measured permittivities of the same powdered materials.

II. PERMITTIVITY AND DENSITY MEASUREMENTS

Permittivity measurements on pulverized coal and limestone samples were made at 11.7 GHz and 20 °C with an X-band measurement system [8] and the short-circuited waveguide method [9]. The technique involved slotted-line (slotted-waveguide-section) measurements with the sample in a 5-cm long WR-90 short-circuited waveguide sample holder attached to the slotted section. Node positions and node widths were determined without and with the sample in the sample holder in contact with the short-circuit termination. Pulverized samples were weighed before they were placed into the vertically oriented sample holder for the measurements. Sample length was determined for each of a sequence of measurements at successively increasing sample bulk densities that were determined from sample weight, sample length, and waveguide cross-sectional area. Permittivities were calculated from sample length, node shift due to the sample presence, and standing wave ratios determined from the node-width measurements with and without the sample in the short-circuited sample holder [10].

Extensive measurements to evaluate errors in permittivity determination associated with various sources of error in the measurement and approximations in the calculations were documented earlier [10], and verification tests on materials of known

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permittivity showed that accuracies better than 0.5% were obtained in measurement of ϵ' [8]. Particle densities ρ_s, which correspond to solid material densities, were calculated from sample weights of 15 to 25 g and corresponding particle volumes that were determined by measurements with an air-comparison pycnometer [1], [6].

III. DETERMINATION OF SOLID MATERIAL PERMITTIVITIES

Results of the permittivity measurements and sample bulk density determinations on the coal, limestone, and 35%–65% coal–limestone mixture are shown for the dielectric constant ϵ' in Fig. 1, where the linearity with bulk density of the cube root of the dielectric constant of an air-particle mixture, as noted previously [5], [11], is consistent with the LLL dielectric mixture equation. This equation can be stated as follows for a two-phase mixture:

$$(\epsilon)^{1/3} = v_1(\epsilon_1)^{1/3} + v_2(\epsilon_2)^{1/3} \tag{1}$$

where subscripts 1 and 2 refer to the air and the solid particulate material, respectively, and v represents the volume fraction occupied by a component of the mixture. For the two-phase (air–particle) mixture, $v_1 + v_2 = 1$, and the permittivity of air is $1 - j0$. Solving (1) for ϵ_2 in terms of ϵ (relative complex permittivity of the mixture) provides an expression from which the permittivities of the solid material particles can be calculated, where $v_2 = v_s$, the volume fraction occupied by the solid material

$$\epsilon_s = \epsilon_2 = \left[\frac{\epsilon^{1/3} + v_2 - 1}{v_2}\right]^3 = \left[\frac{\epsilon^{1/3} + v_s - 1}{v_s}\right]^3. \tag{2}$$

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 LLL equation can be used with confidence in calculating the complex permittivity of the solid material from the permittivities of the powdered samples [6]. The mean values of the solid material permittivities calculated with the LLL mixture equation for the permittivity measurements at each bulk density gave $4.21 - j0.156$ for the coal and $7.41 - j0.063$ for the limestone used in these measurements. The respective solid-material densities for the coal and limestone from the air-comparison-pycnometer measurements were 1.48 and 2.75 g/cm^3 [6].

IV. COMPUTATION OF COAL AND LIMESTONE MIXTURE PERMITTIVITIES

Two dielectric mixture equations that have been observed to provide good results in work with pulverized coal and similar materials are the CRI and the LLL equations [5]. The latter is expressed in (1), and the former can be expressed as

$$(\epsilon)^{1/2} = v_1(\epsilon_1)^{1/2} + v_2(\epsilon_2)^{1/2}. \tag{3}$$

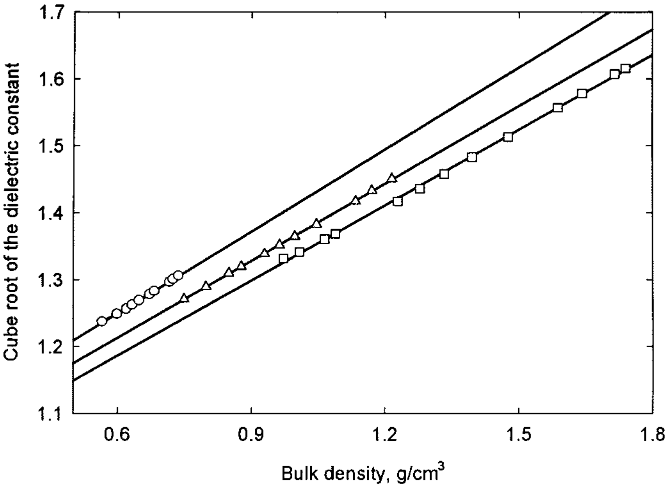

Fig. 1. Linear relationships between the cube roots of the dielectric constants of pulverized samples and their bulk densities at 20 °C and 11.7 GHz. O—coal, △—35%–65% coal–limestone mixture, □—limestone.

Therefore, (1) and (3) have been used to calculate the relative complex permittivity values for the pure coal, pure limestone, and 35%–65% coal–limestone mixture samples for comparison with the measured values over range of 10–13 different bulk densities for these powdered samples. Equations (1) and (3) 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 $4.21 - j0.156$ and $7.41 - j0.063$, respectively, as given in the previous section. The volume fractions can be determined as $v_s = \rho/\rho_s$.

V. RESULTS AND DISCUSSION

Comparisons of the calculated and measured permittivities for the coal, limestone, and 35%–65% coal–limestone mixture are presented in Tables I–III for six bulk densities spanning the range of densities at which measurements were taken on each of the powdered materials. For the coal samples, shown in Table I, the LLL equation (1) 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 CRI equation (3) overestimated the permittivity with an error of about 4% for the dielectric constant and larger errors for the loss factor.

For the limestone samples, shown in Table II, which have higher dielectric constants and lower loss factors than the coal, (1) again provided much closer estimates than (3), with (3) overestimating the dielectric constant by about 10% except at the highest densities where the error is somewhat less. As expected, the performance of both equations in estimating measured properties of the 35%–65% coal–limestone mixture, shown in Table III, 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 LLL 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%

TABLE I
Comparison of Measured and Calculated Permittivities of Pulverized Coal at 11.7 GHz at Indicated Bulk Densities. Landau and Lifshitz, Looyenga Equation (1), Complex Refractive Index Equation (3)

Bulk density	Volume fraction	Measured permittivity		Calculated permittivity (1)		Calculated permittivity (3)	
ρ (g/cm^3)	v_s	ϵ'	ϵ''	ϵ'	ϵ''	ϵ'	ϵ''
0.565	0.382	1.89	0.035	1.88	0.035	1.96	0.041
0.598	0.404	1.95	0.037	1.95	0.038	2.03	0.044
0.632	0.427	2.01	0.039	2.01	0.041	2.10	0.047
0.671	0.453	2.09	0.043	2.09	0.044	2.18	0.051
0.716	0.484	2.18	0.051	2.18	0.049	2.28	0.055
0.736	0.497	2.23	0.054	2.23	0.051	2.32	0.058

TABLE II
Comparison of Measured and Calculated Permittivities of Pulverized Limestone at 11.7 GHz at Indicated Bulk Densities. Landau and Lifshitz, Looyenga Equation (1), Complex Refractive Index Equation (3)

Bulk density	Volume fraction	Measured permittivity		Calculated permittivity (1)		Calculated permittivity (3)	
ρ (g/cm^3)	v_s	ϵ'	ϵ''	ϵ'	ϵ''	ϵ'	ϵ''
0.972	0.353	2.36	0.011	2.38	0.010	2.59	0.013
1.088	0.396	2.57	0.012	2.60	0.012	2.83	0.015
1.278	0.465	2.96	0.014	2.99	0.016	3.24	0.019
1.395	0.507	3.26	0.017	3.25	0.018	3.51	0.022
1.642	0.597	3.93	0.026	3.85	0.024	4.11	0.028
1.739	0.632	4.22	0.031	4.10	0.027	4.36	0.031

TABLE III
Comparison of Measured and Calculated Permittivities of Pulverized Coal–Limestone Mixture (35%–65%, by Weight) at 11.7 GHz at Indicated Bulk Densities. Landau and Lifshitz, Looyenga Equation (1), Complex Refractive Index Equation (3)

Bulk density	Volume fraction	Measured permittivity		Calculated permittivity (1)		Calculated permittivity (3)	
ρ (g/cm^3)	v_s	ϵ'	ϵ''	ϵ'	ϵ''	ϵ'	ϵ''
0.749	0.352	2.05	0.016	2.07	0.019	2.20	0.024
0.849	0.399	2.25	0.022	2.25	0.023	2.40	0.028
0.929	0.436	2.40	0.026	2.41	0.027	2.56	0.032
0.997	0.468	2.54	0.031	2.54	0.030	2.70	0.035
1.133	0.532	2.85	0.039	2.83	0.036	3.00	0.042
1.215	0.570	3.05	0.045	3.02	0.041	3.18	0.046

powdered coal–limestone mixture with an average of 0.5%. Corresponding errors of prediction for the CRI equation ranged as high as 4.6%, 10.2%, and 7.2% for the coal, limestone, and 35%–65% coal–limestone mixture, respectively, with mean values over the full range of densities of 4.3%, 7.7%, and 6.1%, respectively.

In using (1) and (3), which express the mixture permittivity relationships for a two-component mixture, the 35%–65% coal–limestone powdered mixture was considered as a combined or equivalent material, with a solid material density determined by pycnometer measurements as 2.13 g/cm^3. As stated, the solid coal and solid limestone densities were determined by the same method as 1.48 and 2.75 g/cm^3, respectively. Using the latter two densities, one can calculate the solid density of an equivalent material consisting of 35% coal and 65% limestone, by weight, from the following relationship:

$$\frac{1}{\rho_m} = \frac{0.35}{\rho_c} + \frac{0.65}{\rho_l} \tag{4}$$

where ρ_m, ρ_c, and ρ_l represent the solid particle densities for the mixture, the coal, and the limestone, respectively. Solving (4) for ρ_m, with the above values for the solid coal and limestone densities gives 2.11 g/cm^3. This compares with 2.13 g/cm^3 determined by pycnometer measurements and is within the 1% accuracy expected for this type of measurement.

It is also interesting to estimate the permittivities of the powdered mixtures in terms of three components where, by the LLL mixture equation

$$(\epsilon_m)^{1/3} = v_1(\epsilon_1)^{1/3} + v_2(\epsilon_2)^{1/3} + v_3(\epsilon_3)^{1/3}. \tag{5}$$

Letting subscripts 1, 2, and 3 refer to the three components of the mixture, air, coal, and limestone, respectively, the permittivity of the mixture ϵ_m can be calculated in terms of the known permittivities of the components and their volume fractions. The volume fraction for the 35%–65% coal–limestone equivalent material in the mixed powder sample of bulk density ρ_b is given by $v_{eq} = \rho_b/\rho_{eq}$, where ρ_{eq} is the solid density of the equivalent combined coal–limestone material, which

TABLE IV
COMPARISON OF PERMITTIVITIES CALCULATED WITH TWO-COMPONENT MIXTURE EQUATION (1) AND THREE-COMPONENT MIXTURE EQUATION (5) WITH MEASURED PERMITTIVITIES OF 35%–65% PULVERIZED COAL–LIMESTONE MIXTURE AT 11.7 GHz

Bulk density (g/cm^3)	Measured permittivity		Calculated permittivity (1)		Calculated permittivity (5)	
	ϵ'	ϵ''	ϵ'	ϵ''	ϵ'	ϵ''
0.749	2.05	0.016	2.07	0.019	2.09	0.022
0.798	2.15	0.020	2.16	0.021	2.19	0.024
0.849	2.25	0.022	2.25	0.023	2.28	0.027
0.877	2.30	0.023	2.31	0.024	2.34	0.028
0.929	2.40	0.026	2.41	0.027	2.44	0.030
0.963	2.47	0.028	2.48	0.028	2.51	0.032
0.997	2.54	0.031	2.54	0.030	2.58	0.034
1.046	2.64	0.034	2.65	0.032	2.69	0.037
1.133	2.85	0.039	2.83	0.036	2.88	0.041
1.170	2.94	0.042	2.91	0.038	2.97	0.044
1.215	3.05	0.045	3.02	0.041	3.07	0.046

was determined by measurement, as mentioned, as 2.13 g/cm^3. Since $v_1 + v_2 + v_3 = 1$, $v_{eq} = v_2 + v_3$, and $v_1 = 1 - v_{eq}$. The solution of (5) for ϵ_m still requires the determination of the values for v_2 and v_3, knowing that their sum is v_{eq}.

For a unit volume, the mass of the mixture $m_m = m_2 + m_3$, where m_2 is the mass of coal particles in the unit volume, and m_3 is the mass of limestone particles in that same volume. For the 35%–65%, by weight, mixture, $m_2 = 0.35m_m$, and $m_3 = 0.65m_m$. Thus, $v_2 = 0.35(\rho_{eq}/\rho_2)v_{eq}$, and $v_3 = 0.65(\rho_{eq}/\rho_3)v_{eq}$, which permits the calculation of the needed volume fractions v_1 and v_2.

The permittivities calculated by the two-component mixture equation (1) and those calculated by the three-component equation (5) are compared with the measured permittivities in Table IV for all 11 different densities for which permittivity measurements were obtained. Both the dielectric constants and loss factors calculated with the three-component mixture equation were somewhat larger than the measured values and somewhat larger than the values predicted by the two-component mixture calculation as well. Errors in the dielectric constant, compared to the measured values, averaged 1.5% for the three-component equation compared to 0.5% for the two-component equation. Thus, for this application, use of the two-component equation seems preferable, and it is also a bit simpler to use. However, this example illustrates that the three-component LLL mixture equation can be used to provide reasonable estimates for the permittivity of such powdered mixtures.

VI. CONCLUSIONS

These tests have shown that the Landau & Lifshitz, Looyenga (LLL) dielectric mixture equation can be used to provide accurate estimates of the permittivities of powdered materials at known bulk densities, when the permittivities of the solid constituents and their densities are known. Accuracy of prediction by the equation for the dielectric constant was higher for the coal (0.15% error), which had a permittivity of $4.21 - j0.156$, than it was for limestone (1.4% error), which had a permittivity of $7.41 - j0.063$. The error of prediction for the 35%–65% mixture was 0.5%. The complex refractive index dielectric mixture equation had poorer performance in estimating the measured permittivity values for the powdered samples with errors several times greater than those of the LLL equation.

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