Abstract—The open-ended, coaxial-line probe technique can be used to obtain estimates of the permittivities of some solid dielectrics over broad ranges of frequency by measurements on powdered or pulverized samples, but certain limitations must be recognized. Such a probe was used with a network analyzer to estimate the permittivities of coal and limestone from reflection coefficients measured on pulverized samples. The bulk density of the pulverized samples for the coaxial probe measurements was determined from auxiliary, single-frequency permittivity measurements on the samples at known bulk densities, and the permissivities of the solid materials over the frequency range from 0.2 GHz to 20 GHz were then estimated by computations based on the Landau and Lifshitz, Looyenga dielectric mixture equation and solid material densities.

Index Terms—Bulk density, coal, dielectric properties, limestone, microwave measurements, open-ended coaxial-line measurements, particulate materials, permittivity measurements.

I. INTRODUCTION

ROAD frequency-range RF and microwave complex permittivity measurements are of interest for many dielectric heating and nondestructive property sensing applications. The open-ended coaxial-line sensor has been considered for such measurements on lossy materials [1] and developed commercially for use with network analyzers for measurements on a wide range of materials [2], [3]. The advantages of the technique include convenience and easy sample preparation. It is very convenient for liquid samples. However, the accuracy of the technique varies with materials to be measured and the frequency at which measurements are needed. Accuracies are poorer on very low loss materials, and solids must have accurately machined plane surfaces to avoid errors caused by any appreciable air gaps. Nonhomogeneity of the dielectric will cause problems with granular or particulate materials. The probe technique was used to obtain estimates of the permittivities of compressed high-loss samples of stored-grain insects [4], but auxiliary single-frequency measurements by another technique were necessary to establish the sample bulk densities, and mixture equations were then used for estimating the properties of the insects.

Similar methods have been used in estimating the broad frequency-range permittivities of coal and limestone from measurements on powdered or pulverized samples of these materials. Measurements were taken to determine the differences in permittivities of the two materials for possible use in detecting rock dust concentration in coal mines, where rock dusting is required to keep the noncombustible dust content at or above 65% for explosion prevention [5]. Results of these measurements are reported in this paper.

II. MATERIALS AND METHODS

Pulverized samples of Pittsburgh coal and limestone (more than 90% calcium carbonate), with most of the particle diameters ranging from 5 μm to 100 μm, were furnished for the measurements by the Pittsburgh Research Center, U.S. Bureau of Mines. Moisture content, determined by drying at 105 °C for 24 h in an air oven, was 1.4% for the coal and 0.14% for the limestone.

Permittivity measurements were made on the powdered samples with a Hewlett-Packard 85070B open-ended coaxial-line probe and 8510B Microwave Network Analyzer. Calibrations and computations of permittivity were performed as described by Blackham and Pollard [3] who present a good review of prior studies and techniques employed by various investigators for reliable determination of permittivities from reflection coefficients at the plane of the open-ended, coaxial-line probe. The measurement system had previously been checked and found to provide reliable values for the permittivity of methanol over the frequency range from 0.2 GHz to 20 GHz [7].

The samples were confined in a Delrin sample cup that was held by an O-ring which sealed it into a Delrin water jacket used to maintain the samples at 25 ± 0.1 °C during the measurements (Fig. 1). A water and ethylene glycol solution was used in the temperature controller that circulated the fluid around the water jacket. The probe was rigidly mounted in a probe clamp on a vertical stand, and the sample cup and water jacket assembly rested on another movable support just below the probe. The network analyzer was calibrated with the probe connected through its special cable to the Test Set with measurements on air, a short circuit, and a triple-distilled-water sample at 25 °C, and test measurements were taken on air and water as a check to determine that correct values were obtained. The water jacket and sample cup containing the sample were then raised to insert the probe into the sample cup, insuring partial

1Mention of company or trade names is for purpose of description only and does not imply endorsement by the U.S. Department of Agriculture or The University of Georgia.

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Compression of the powdered sample in the Delrin sample cup to provide complete and stable contact between the probe and the powdered sample. Five to ten measurements of the permittivity were performed in succession, after which the sample cup and water jacket assembly was lowered to expose the probe, it was cleaned with a small camel-hair brush, and an air measurement was made to insure that the calibration had remained stable.

The network analyzer provided permittivity data at 51 frequencies at intervals on a logarithmic scale from 0.2 GHz to 20 GHz, but the bulk density of the powdered sample in the small region sensed by the probe was unknown. Even though the average bulk density of the sample in the sample cup can be determined from sample weight and dimensions, the local density at the tip of the probe can be much different for particulate samples [4]. The determination of this sample bulk density is critical to the estimation of the permittivities of the solid coal and limestone materials. The bulk density value was determined from auxiliary measurements at 11.7 GHz where sample bulk densities were accurately known and the relationships between permittivity and sample bulk densities could be well determined [4].

The required single-frequency auxiliary permittivity measurements were made at 11.7 GHz with an X-band measurement system [8] and the short-circuited waveguide method [9], [10]. Pulverized samples were weighed before they were placed into a 5 cm long, WR-90-waveguide, short-circuited sample holder for the measurements. Sample length (about 4 cm to 2 cm) 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.

Particle densities, \( \rho_s \), which correspond to solid material densities, were calculated from pulverized sample weights of 15 g to 25 g and corresponding particle volumes that were determined by measurements in a Beckman Model 930 Air-Comparison Pycnometer [11]. Initial pycnometer measurements on pulverized coal samples revealed that the coal was compressible, as indicated by continual drift of the pycnometer pressure null indicator for several minutes after the sample was placed under 2 atmospheres of air pressure. Therefore the 1–1/2–1 atmosphere mode of operation was used, so the air-displacement measurement could be determined at a pressure of 1 atmosphere. Pycnometer measurements on limestone samples were made in the 1-to-2 atmosphere compression mode, because both methods gave the same particle volume values, and the 2-atmosphere mode has better sensitivity for the null pressure determination.

Once the permittivity data had been obtained from the probe measurements over the 0.2 GHz to 20 GHz range, the sample bulk densities had been determined from the auxiliary X-band data at 11.7 GHz (the density at which the dielectric constant from the probe and X-band results agreed), and the particle densities (solid material densities) were determined by pycnometer measurements, the estimated permittivities for the solid materials could be determined by computations based on the Landau and Lifshitz, Looyenga complex dielectric mixture equation. Temperature dependence of the dielectric constant for these materials is negligible for the comparison of values from X-band measurements at 20 °C and coaxial-line probe measurements at 25 °C.

It was shown earlier [12]–[14] that the linearity with bulk density of the cube root of the dielectric constant of an air-particle mixture, \( \epsilon' \) (the real part of the relative complex permittivity, \( \epsilon = \epsilon' - j\epsilon'' \), where \( \epsilon'' \) is the dielectric loss factor), is consistent with the Landau and Lifshitz, Looyenga dielectric mixture equation, which can be stated as follows for this two-phase mixture:

\[
(\epsilon')^{1/3} = v_a(\epsilon_a)^{1/3} + v_s(\epsilon_s)^{1/3}
\]  

(1)

where subscripts \( a \) and \( s \) 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, \( v_a + v_s = 1 \), and the permittivity of air is \( 1 - j\sigma_0 \). Solving (1) for \( \epsilon_s \) in terms of \( \epsilon \) (relative complex permittivity of the mixture) and \( v_s \), the volume fraction occupied by the solid material, provides an expression from which the permittivity of the particulate material can be calculated

\[
\epsilon_s = \left[ \left( \epsilon^{'1/3} + v_s - 1 \right) \right]^{3} \vspace{10pt}
\]

(2)
The necessary value for \( v_s \) can be obtained from the bulk density \( \rho \) of the mixture and the density \( \rho_s \) of the solid particulate material, since \( v_s = \rho / \rho_s \).

III. RESULTS

The cube roots of the dielectric constants of the pulverized coal and limestone samples determined by the X-band measurements at 11.7 GHz [6] are shown as functions of sample bulk density in Figs. 2 and 3. The relationship is expressed as

\[
(\varepsilon')^{1/3} = a + b\rho.
\]  

Linear regression analyses (Table I) showed very high coefficients of determination \( (r^2) \), and the intercepts are very close to the theoretical value of 1, which, for zero bulk density, is the dielectric constant of air. Since the point \( (\rho = 0, \varepsilon' = 1) \) is a valid data point, it was included in the regression calculations. When the linear regression of the cube root of the dielectric constant on bulk density provides an \( r^2 \) value so nearly 1 and the zero–bulk-density intercept is so close to the value of 1, the Landau and Lifshitz, Looyenga dielectric mixture equation can be used with confidence to estimate the permittivity of the solid material. The linear extrapolation of \( (\varepsilon')^{1/3} \) to the density of the solid material is illustrated for the coal measurements in Fig. 2 and for the limestone measurements in Fig. 3. Values provided by the regression equations for the dielectric constants of the two material samples are given in Table I. The mean values of the solid material permittivities shown in Table I were calculated by (2) from the Landau and Lifshitz, Looyenga mixture equation predictions for the permittivity measurements at each of the ten bulk densities illustrated for coal in Fig. 2 and at each of the thirteen bulk densities in Fig. 3 for limestone.

Results of the computations from the coaxial-probe measurements are illustrated for the dielectric constant of coal in Fig. 4. Similar plots for the dielectric constant of limestone and the loss factors of coal and limestone were fitted with second- and third-order polynomial equations, and the results for the dielectric constants are summarized in Fig. 5. Resulting estimates for the loss factor were judged unreliable for reasons to be discussed later.

IV. DISCUSSION AND CONCLUSIONS

Permittivity values obtained by the X-band waveguide measurements for the pulverized coal are in reasonable agreement with those reported for other measurements on pulverized coal samples [15]–[20] when differences in frequency, moisture content, and density are taken into account. Values obtained check extremely well with those reported earlier for Pittsburgh No. 8 run-of-mine coal at the same frequency and similar densities [19].

One can note from Table I that the \( \varepsilon'_s \) values predicted by the linear regression equations agree better with the mixture equation prediction when the intercept is closer to the value 1. The mixture equation prediction of the dielectric constant is equivalent to that provided by a straight line through the point \( (0, 1) \) and the selected single point defined by the cube root of the measured dielectric constant at any particular bulk density. The intersection of that straight line with the vertical line at the solid material density \( \rho_s \) gives the estimate for \( \varepsilon'_s \). Thus, if the point from the measured \( (\rho, \varepsilon') \) value is above the regression line (see Fig. 2 for example), the estimated \( \varepsilon'_s \) value will be high, and if the measured point is below the regression line, the estimated value for \( \varepsilon'_s \) will be low.

For the measurements reported on these samples, the mean values of the permittivities calculated by the Landau and Lifshitz, Looyenga mixture equation, taken over all measured permittivity and bulk density points, should provide the most reliable estimates for \( \varepsilon'_s \), and they also provide values for the loss factor, which is of greater interest than the dielectric constant for most dielectric heating applications. For coal and limestone, the estimates over all data points for \( \varepsilon'_s \) ranged from 4.20 to 4.26 and 7.21 to 7.70, respectively, with mean values as listed in Table I. Corresponding ranges for \( \varepsilon''_s \) were 0.15 to 0.17 and 0.05 to 0.07, respectively [6].

Results of the extension of the permittivities obtained from the coaxial probe measurements on the powdered samples to
TABLE I
ESTIMATED PERMITTIVITIES OF SOLID MATERIALS FROM X-BAND WAVEGUIDE MEASUREMENTS ON PULVERIZED SAMPLES AT 20 °C AND 11.7 GHz

<table>
<thead>
<tr>
<th>Material</th>
<th>Density, g/cm³</th>
<th>Linear Regression</th>
<th>ε' predicted by linear regression (mean values)</th>
<th>ε' by mixture eqn. (mean values)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Intercept</td>
<td>Slope</td>
<td>Coeff. determn.</td>
</tr>
<tr>
<td>Coal</td>
<td>1.48</td>
<td>1.0003</td>
<td>0.4152</td>
<td>0.9999</td>
</tr>
<tr>
<td>Limestone</td>
<td>2.75</td>
<td>0.9877</td>
<td>0.3561</td>
<td>0.9982</td>
</tr>
</tbody>
</table>

Fig. 4. Dielectric constants of solid coal, calculated from open-ended coaxial-line probe measurements on pulverized Pittsburgh coal, at 25 °C, fitted with third-order polynomial regression. \( r^2 = 0.95 \).

Fig. 5. Dielectric constants of solid Pittsburgh coal and solid limestone estimated from coaxial-line probe measurements by complex computation based on Landau and Lifshitz, Looyenga mixture equation, and density of the solid materials. Coal, \( r^2 = 0.95 \). Limestone, \( r^2 = 0.84 \).

Those for the solid material by way of the mixture equation computations indicate that the dielectric constants of both the coal and the limestone decrease with increasing frequency between 0.2 GHz and 20 GHz, with the limestone showing little dispersion below 3 GHz (Fig. 5).

The broad-frequency-range loss factors of both coal and limestone estimated by this technique were erratic and three to ten times greater than those obtained from the X-band short circuited waveguide measurements at 11.7 GHz (Table I) and were, therefore, unreliable. Other measurements have shown that the loss factor decreases with increasing frequency for coal [19] and for limestone [21]. Loss factor values obtained from the X-band measurements agree well with previously reported data [19]-[21]. The low accuracy of the open-ended coaxial probe technique for materials of such low loss (\( ε'' \) about 0.04 and 0.02, respectively, for the pulverized coal and limestone samples at 11.7 GHz [6]) is most likely responsible for the unsatisfactory results in estimating loss factors by this technique from the open-ended coaxial-line probe measurements on pulverized samples.

Problems of resonance due to reflections from the dielectric interfaces at the sample and sample cup boundaries were also observed, as may be noted at the higher frequencies in Fig. 4. These might be expected because of the low-loss nature of the pulverized materials. Other precautions can be taken to eliminate or minimize such disturbances [1], and resulting measurements could be improved with further measurements and refinement of the techniques. However, the open-ended coaxial probe technique is not suitable for reliable determination of the loss factors of such low-loss samples. The insensitivity at the lower end of the frequency range is also evidenced by the scatter in the dielectric constant data in Fig. 4. Instability of the calibration is also a problem in working with such low-loss and low-permittivity materials.

Values of the permittivity of coal and limestone are sufficiently different to justify further studies aimed at determining rock dust content in coal and rock dust mixtures by dielectric sensing techniques. Further measurements by more reliable techniques of coal and rock dust samples and of mixtures of different proportions will be necessary. Consideration of complicating factors, such as equilibrium moisture contents, and permittivity-density relationships for the mixtures will also be required to better assess the potential for sufficiently accurate determinations of rock dust content in coal and rock dust mixtures.

REFERENCES


Stuart O. Nelson (SM’72–F’98), for a photograph and biography, see this issue, p. 126.

Philip G. Bartley, Jr., for a photograph and biography, see this issue p. 125.