Dielectric Investigation on Coals. I. Dielectric Properties of Japanese Coals

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(Received January 22, 1957)

Introduction

Little information exists in the literature of the subject concerning the dielectric constant of coals. In 1948 Bond, Griffith and Maggs[1] reported a dielectric study of coals containing absorbed water. Their emphasis was then placed on the freezing mechanism of the water below 0°C and no reliable measurement was made on the dielectric constant of coal itself*.

In 1955 van Krevelen and his co-workers[2] published a report on dielectric constants of low rank and bituminous coals. The coal samples treated in their study were reduced to fine powders and the dielectric constant of the powders was indirectly estimated by the use of an immersion method. As is known with certainty, the conditions of their measurement were not free from errors of various sources. For instance, no reliable data are to be obtained in the region of the anomalous dispersion which is expected to occur from the dielectric heterogeneity of the measured system[3]. They measured apparent dielectric constants of binary mixtures composed of coal powder and an immersion liquid such as diisopropyl ether. In order to secure accuracy of measured values it is desirable that the measurements be made covering a wide range of frequencies or of temperatures. But actually their measurements were limited to a single frequency of 1 Mc/s at room temperature.

Since 1953 a dielectric investigation[4,5]

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[1] R. L. Bond, M. Griffith and F. A. P. Maggs, *Disc. Faraday Soc.*, **3**, 29 (1948).

[2] M. P. Groenewege, J. Schuyer and D. W. van Krevelen, *Fuel*, **34**, 339 (1955).

* There are available other papers in which the dielectric method was employed to estimate the moisture content in coals. See, for example, W. Klempt, *Brennstoff-Chem.*, **34**, 294 (1953); H. Oikawa, *J. Coal Res. Inst.*, (Tokyo), **3**, 137 (1952).

[3] K. W. Wagner, *Arch. Elektrotech.*, **2**, 371 (1941); R. W. Sillars, *J. Inst. Elec. Engrs.*, **80**, 378 (1937).

[4] I. Miyasita, *Bull. Res. Inst. Appl. Elec.*, Hokkaido Univ., **5**, 123, (1953), **6**, 117 (1954).

[5] I. Miyasita, R. Miura and K. Higasi, This Bulletin **28**, 148 (1955).

TABLE I

CHEMICAL ANALYSIS OF COALS USED FOR DIELECTRIC STUDIES*

(Composition on moisture-free basis, wt. %)

Coal No.	Seam	Ash	Carbon	Hydrogen	Oxygen	Nitrogen	Sulfur
I	Moziri, No. 4	5.12	85.25	5.89	6.91	1.72	0.23
II	Moziri, No. 3	11.85	81.71	5.93	9.78	2.00	0.58
III	Moziri, No. 9	3.55	82.98	5.92	9.07	1.89	0.14
IIIa	Moziri, No. 11	8.83	82.07	5.92	9.86	1.81	0.34
IV	Akabira, No. 11	6.80	80.76	5.91	—	—	—
V	Mitui-Bibai, No. 3-4	2.94	80.47	5.87	—	—	—
Va	Mitui-Bibai, No. 4-4	3.87	80.12	5.78	—	—	—
Vb	Mitui-Bibai, No. 1-5	4.12	78.40	5.78	—	—	—
Vc	Mitui-Bibai, No. 1-3	5.00	80.62	5.71	11.55	1.88	0.24
VI	Tempoku (brown coal)	1.22	66.62	5.26	—	—	—

* The writers' thanks are due to Mr. Y. Zyomoto for providing the data of this Table.

on coals has been under way in the laboratory of the present writers. In the present article of this series detailed description will be presented of the results obtained for ten coal samples from Hokkaido mines, Japan. It may be worth while to point out that successful dielectric determination was done on coals themselves for the first time. Further, a wide range of frequencies between 50Mc/s and 300 c/s was used in the present measurement.

Material and Method

Samples were prepared from the following ten coals (See Table I). For the sake of convenience, they have been arranged in order according to the geological age of the coal bed concerned. Coals (I)—(Vc): Hokkaido bituminous coals. Coal (VI): Tempoku brown coal, Hokkaido mine. Analyses of the coal samples are given in Table I.

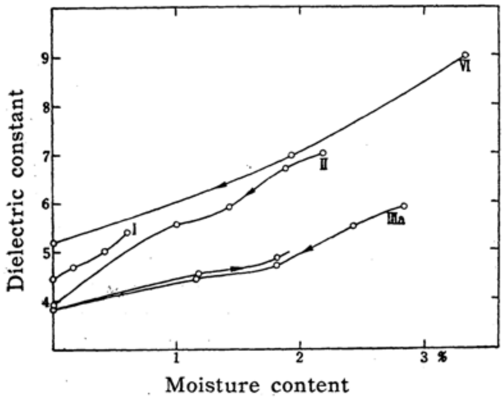

Fig. 1. Effect of dehydration on dielectric constants (at 1 Mc/s) of various coals. I, II, IIIa: Moziri bituminous coal; VI: Tempoku brown coal (cf. Table I). There is uncertainty as to the state of absolute drying.

Coal blocks which were rich in vitrain but had no cracks, were selected, carefully cut, ground, polished and finally made into the form of a circular plate of 5±0.1 mm. thick and 40 mm. in diameter. No sort of binding material such as Canada balsam was employed in the preparation of the disks.

The disks thus prepared were dried in a desiccator over phosphoric anhydride for three or four months. The presence of moisture in the sample usually results in obtaining a higher dielectric constant than expected (see Fig. 1). It is often accompanied by an anomalously large loss factor in the lower frequency region. Therefore, the complete removal of moisture was concluded to have been accomplished when the dielectric constant (especially measured in the lower frequency side) was low and became unchanged by further desiccation. Further check on this process of drying was made for one sample coal (IIIa) by making a comparison with another method of drying. The same material dried at 105°C in air for 28 hours gave little difference in dielectric constant.

The dielectric measurements were made with a Q-meter (Yokogawa QM-101) for the frequency range of 50 Mc/s to 50kc/s, and with an impedance bridge (Yokogawa BV-z-103A) for the range of 20kc/s to 300c/s. To attain a higher accuracy of the impedance bridge in the measurement of low-loss materials, a special device was necessary*. For this purpose, a combined unit of a narrow-band amplifier and a cathode-ray oscillograph was employed to detect the balance in the bridge circuit. Further, the source for the Q-meter was regulated with an automatic stabilizer.

Aluminium foils of 38 mm. diameter were pasted on the surfaces of the coal disk. They were connected with electrodes of the same 38 mm. diameter. The same electrodes were used both with the Q-meter and with the impedance bridge.

Readings of the Q-meter were taken when a resonance was established. After the test piece was removed resonace was regained by the use

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* Detailed description of this device will be reported elsewhere (I. Miyasita, to be published in *Bull. Inst. Appl. Elec.*, Hokkaido University).

TABLE II

VARIATION OF DIELECTRIC CONSTANT (ε') AND LOSS FACTOR (ε'') WITH FREQUENCY AT ROOM TEMPERATURE

Coal No.		300 c	1 kc	3 kc	10 kc	20 kc	100 kc	300 kc	1 Mc	10 Mc	50 Mc
I	ε'	4.56	4.55(4.54)	4.54	4.52	4.37	4.49	4.52	4.47	4.41	4.49
	ε''	—	0.0395	—	—	—	0.0346	0.0347	0.0351	0.0325	0.0318
II	ε'	4.51	4.37(4.37)	4.28	4.15	4.08	4.15	3.96	3.92	3.82	3.80
	ε''	—	0.0926	—	—	—	0.080	0.076	0.075	0.070	0.058
III	ε'	4.01	3.99(3.94)	3.97	3.96	3.86	3.92	3.88	3.88	3.86	3.83
	ε''	—	0.0334	—	—	—	0.0204	0.0173	0.0173	0.0189	0.021
IIIa(1)	ε'	4.25	4.19	4.14	4.12	4.05	3.99	3.90	3.85	3.78	3.74
	ε''	—	—	—	—	—	0.0585	0.0447	0.0424	0.0308	0.0278
IIIa(2)	ε'	4.07	3.98(3.85)	3.93	3.88	3.81	3.70	3.69	3.69	3.69	3.70
	ε''	—	0.0404	—	—	—	0.032	0.0216	0.0238	0.0181	0.0228
IV	ε'	3.76	3.75	3.75	3·66	3.66	3.66	3.64	3.66	3.61	3.46
	ε''	—	0.0325	—	—	—	0.0206	0.0169	0.0166	0.0169	0.0204
V	ε'	3.76	3.74	3.74	3.71	3.59	3.64	3.69	3.61	3.64	3.69
	ε''	—	0.0224	—	—	—	0.0128	0.0109	0.0124	0.012	0.014
Va	ε'	4.42	4.34(4.30)	4.30	4.20	4.11	4.12	4.10	4.10	4.08	4.14
	ε''	—	(0.0316)	—	—	—	—	—	—	—	—
Vb	ε'	4.5	4.42(4.25)	4.28	4.28	4.16	4.23	4.23	4.19	4.16	4.16
	ε''	—	(0.0442)	—	—	—	0.030	0.0266	0.0288	0.0268	0.0337
Vc	ε'	4.73	4.70	4.57	4.48	4.38	—	—	—	—	—
VI	ε'	5.65	5.56	5.5	5.4	5.28	5.35	5.35	5.20	5.10	5.25
	ε''	—	—	—	—	—	0.0815	0.0768	0.0856	0.101	0.137

of an auxiliary condenser when the two electrodes came to be the same distance apart. Dielectric constant and loss factor of the sample were obtained by simple calculations from the readings at the resonance points. Procedure with the impedance bridge was essentially the same as those described in the usual text book.

The range of errors in the measured quantities is largely dependent upon the frequency of the measurement. Broadly speaking the Q-meter and the impedance bridge, after the improvement above noted are believed to give errors of less than ±2% in dielectric constant, while the loss factors are more sensitive in respect to conditions of the measurement.

As none of the test pieces prepared from coal samples was a perfectly circular disk with two perfectly flat surfaces, error arising from the irregularity of the shape demanded special attention. Regarding edge effects, Prof. Miura of the electrical engineering dept. of this University kindly took the trouble to examine the error involved. Further, two sorts of calibration experiments were conducted: first, with a test piece of variable edge; second, comparison with Hartshorn's[6] procedure regarding this correction. Error of the edge irregularity in the dielectric constant was inferred thus to be only a small percent, or less than 5%.

Concerning the unflat surfaces, the use of aluminium foil was believed to help diminish the principal error, which arises from the possible air gap between the electrodes and the test piece. Both the inhomogeneity of the sample and the existence of ash and moisture provided other difficulties, of which the latter is more important. Every effort was made to eliminate errors arising from all these effects, but they were not always estimable. Probably one may assume that the maximum error in the absolute value in dielectric constant amounts to about 5% or near to it. Loss factors in general are more liable to errors than dielectric constants, but they are less significant in the interpretation of the results.

Measurements were carried out at room temperature. For some samples, (IIIa) (Va) and (V), measurements were also made at 100°C, but no appreciable difference was found in the dielectric constant values.

Experimental Results

Results of the measurement are collected in Table II. Values in parentheses are those redetermined after the lapse of a few months (in a desiccator). In sample (IIIa) two test pieces prepared from the same coal specimen were examined (Compare IIIa(1) and IIIa (2) in Table II). One gave a higher value in dielectric constant than the other by the amount 0.04—0.29. The choice of the two pieces was not made at random—they were selected from blocks of entirely different appearance but the same specimen. Therefore this deviation of less than ±10% may suggest the range of maximum deviation in the dielectric constant which arises from any individual difference of the sample.

Perhaps the most remarkable feature in this table is that, over a wide range of frequency, observed dielectric constants show values of less than 5, while loss factors have small values below 0.1. Only one exception is found in the brown coal (VI).

There are available no refractivity data for Japanese coals. But according to our microwave measurement on Mayazi coal (83.14% carbon), its high frequency dielectric constant was 3.37[7].

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[6] L. Hartshorn and W. H. Ward, *Proc. Inst. Elec. Engrs.*, **79**, 597 (1936).

[7] K. Higasi, I. Miyasita and Y. Ozawa, this Bulletin, **30**, 546 (1975).

Further it is reported that the refractive index for European coals amounts to 1.7–2.0[8]. Therefore it may be reasonable to suppose that the observed dielectric constants should be only slightly larger than the square of the corresponding refractive index, the difference being less than 2[5]. These points will be discussed fully in subsequent papers.

Discussion

To begin with, the dielectric constants measured at two frequencies, 1 kc/s and 1 Mc/s, are plotted against the carbon content (dry, ash-free) of the coal. For the sake of comparison the dielectric constants at 1 Mc/s given by van Krevelen[2] are plotted in the same figure. Full discussion on this problem will be given when ample data on a variety of coals are provided from the investigation on coal powders.

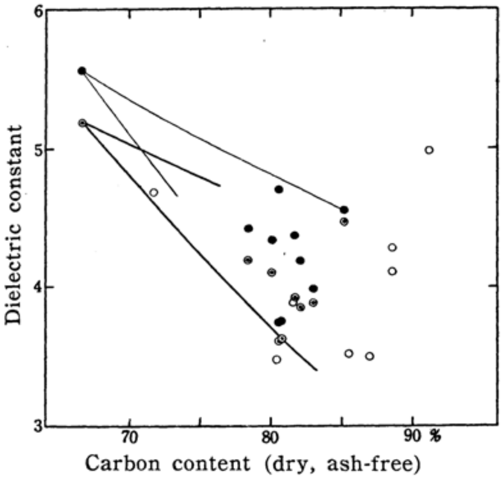

Fig. 2. Dielectric constant of coal and carbon content.

◉ 1 Mc } Present research
● 1 kc }
○ 1 Mc Krevelen's data for strongly dried coal.

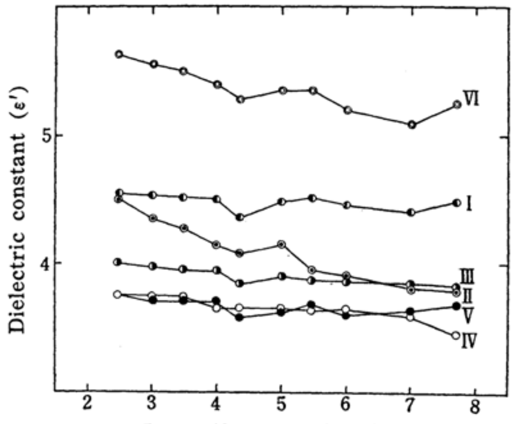

Fig. 3. Variations of dielectric constants of various coals with frequency. I, II, III: Moziri coal; IV: Akabira coal; V: Mituibibai coal; VI: Tempoku coal (cf. Table I).

In the present article attention will be focussed on the frequency dependence of dielectric constants and loss factors. These quantities, when directly obtained from coals themselves, are free from errors due to dielectric inhomogeneity. At a glance at the ε' curves in Fig. 3, one is impressed by the fact that the curves for (I), (III), (IV), (V) are nearly flat and run almost parallel. That is, they are non-dispersive in the whole range of the measured frequencies 300 c/s – 50 Mc/s. Slight increases at the lower frequency side are found in the curves of (II) and (VI), but they are not very pronounced.

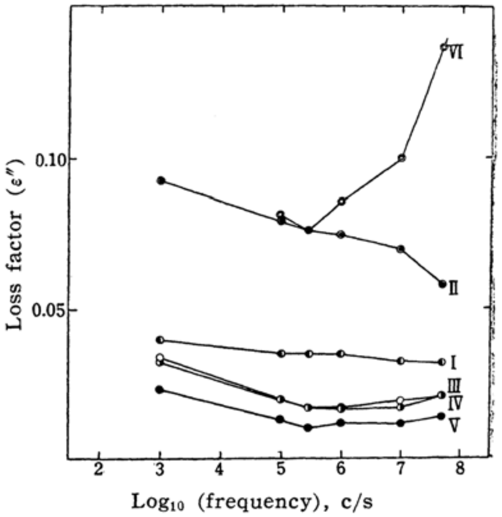

Fig. 4. Variations of loss factors of various coals with frequency. Coal No. I, II, III, etc: ditto (cf. Table I).

The curves for loss factors (Fig. 4) are of interest in this connection. It will be confirmed from loss factor that there is no anomalous dispersion in the samples of (I), (III), (IV) and (V). Samples (II) and (VI) have larger loss factors than others, but the frequency dependence is entirely different between the two. The loss factor ε'' of (II) gradually decreases at the higher frequency side, while that of (VI) sharply increases. This may suggest that the mechanism of dielectric absorption is different in the above two cases. From among the data not reproduced in Figs. 3 and 4, the samples of (IIIa2) and

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[8] D. W. van Krevelen, *Brennstoff-Chem.*, **34**, 167 (1953).

(Vb) are to be classified in the same group as (II) from the frequency dependence of the loss factor.

There are two types of causes which produce dielectric dispersion. One is the existence of movable dipoles in the system. In certain polymers and organic crystals dipoles are not completely blocked from rotation, producing distinct dispersion phenomena[9]. In Fig. 5, dielectric behavior of phenol-formaldehyde resin[10] is compared with that of Tempoku brown coal. It is generally accepted that the dielectric losses of this plastic are mainly determined by the rotation of the polar OH groups of the structure. Similarly Tempoku coal may have some polar groups such as OH group which have freedom of rotation to a certain extent. However, there is not any of the coals which exhibits dispersive charactor so clearly as Tempoku coal (Figs. 3 and 4). Most of the bituminous coals behave like non-polar substances. It seems reasonable to suppose, therefore, that in these coals the polar groups are in a state of non-rotation with the electric field applied from outside.

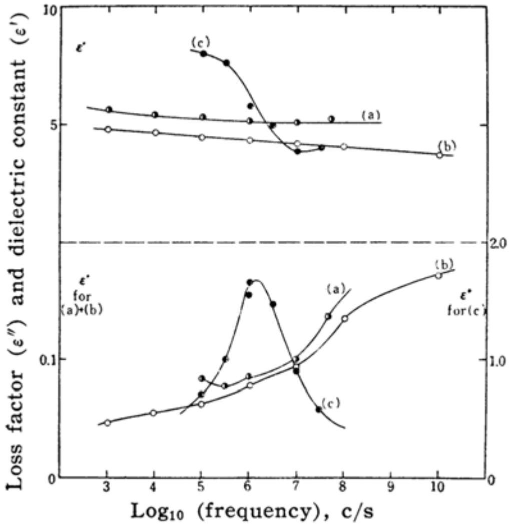

Fig. 5. Two types of dielectric dispersion.
(a) Tempoku brown coal.
(b) Phenol-formaldehyde resin.
(c) A mixture of coke-paraffin wax.

The other important cause of dielectric dispersion is the dielectric heterogeneity of the system. For the sake of illustration, the dielectric properties of a mixture of coke-paraffin wax[11] are shown in curves (c) in Fig. 5. A peak in the loss factor is observed at a particular frequency when a semi-conductive particle of coke is surrounded by paraffin medium with good insulating power. Indeed, the direct current conductivity of coke itself is calculable with some precision from that frequency by the use of Wagner-Sillar's theory[3,11].

It is of some interest in this connection that Ellis[12] advanced a proposition that there exist units of graphite structure in bituminous coals. If such structure corresponds to particles of semi-conductive nature, the situation may not be essentially different from that of a coke-paraffin wax system. As far as the coals treated in the present experiment are concerned, no such dispersion phenomenon was observed as would support Ellis's view on the graphite micellar structure*.

Lastly, let attention be directed to the increase of loss factor at the lower frequency region as observed in curve II in Fig. 4. There are two reasons for believing that this increase is due to the trace of water in the system. First, this sort of increase in loss factor becomes very pronounced in all the materials which are not completely dried. Second, the phenomenon itself can be theoretically accounted for by Sillar's calculation[3] on the assumption that in cracks of the coal there is some trace of water particles of elongated shape.

Summary

In a wide frequency range covering 300 c/s to 50 Mc/s dielectric constant and loss factor of ten Japanese coals were measured at room temperature. The result provides a view that Japanese coals are almost non-polar in effect and embrace no units of graphite structure of the sort proposed by Ellis.

The writers wish to thank Prof. G. Takeya and Assit. Profs. R. Miura and M. Kugo for friendly help and stimulating discussions.

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[9] C. P. Smyth, "Dielectric Behavior and Structure," Chap. V, McGraw-Hill Book Co. (1955).

[10] L. Hartshorn, L. V. L. Parry and E. Rushton, *Proc. Inst. Elec. Engrs.*, **100**, 23 (1953).

[11] Y. Zyomoto and K. Higasi, to be published in this Bulletin. Further see Y. Zyomoto and K. Higasi, *Kagaku*, **26**, 638 (1956).

[12] C. Ellis, *Proc. Roy. Soc.* (London), **212A**, 1 (1952).

* As the nature of Japanese coals is regarded to be particular as compared with European coals, this does not exclude the possibility that British bituminous coals have such graphite units.