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TECH. NOTE
CHEM.1215

TECH. NOTE
CHEM.1215

ROYAL AIRCRAFT ESTABLISHMENT
F A R N B O R O U G H , H A N T S

TECHNICAL NOTE No: CHEM.1215

**THE DIELECTRIC PROPERTIES OF SOME
RUBBER-BASED CORE MATERIALS
FOR SANDWICH RADOMES AND
THE EFFECTS OF MOISTURE,
TEMPERATURE AND FREQUENCY**

by

A.A.FYALL and J.H.SEWELL

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ROYAL AIRCRAFT ESTABLISHMENT, FARNBOROUGH

The dielectric properties of some rubber-based core materials for sandwich radomes and the effects of moisture, temperature and frequency

by

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RAE Ref: Chem.804/A.A.F.

SUMMARY

Values are given for the dielectric constant and loss tangent of several vulcanizates proposed as core materials for airborne sandwich type radomes. Dielectric properties were measured on dry materials and also on materials subjected, for varying periods of time, to tropical conditioning in a humidity chamber. The effect of variation of frequency on the dielectric properties was investigated.

A brief review is made of the effect of structure and composition on the dielectric properties of vulcanizates and the behaviour of the materials tested is explained on the basis of this review.

As the materials were not in their usual expanded form when measured their approximate dielectric properties in this expanded form are calculated.

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1 Introduction

1.1 General aspects of sandwich radomes

From the design viewpoint, the simplest type of radome is constructed from a single sheet of material. For transmitted radiation of 25 centimetres wavelength or above, such a design is feasible because the wall thickness permissible for the electrical functioning of the radome, allows the fabrication of an efficient structure. At radar frequencies, however, the thicknesses involved are mechanically unacceptable, but a slight modification in construction overcomes this difficulty. If a radome is constructed of two thin skins, so that the distance between them is a quarter of the radar wavelength, the reflections from the two skins, being of like phase at the point of reflection, are out of phase by π radians at any point outside the walls, and cancel out. Depending on the nature of the spacing material, a substantial gain in structural strength is obtainable.

Arrangements vary for separating the two skins in these "sandwich" type radomes. Mechanical spacers can be used but their excessive weight and the need to maintain critical tolerances on the spacing makes them impractical. In the construction of sandwich radomes to-day, the spacer takes the form either of an expanded or foamed plastic or of a honeycomb structure, usually made of glass fibres impregnated with a suitable resin. The foam type of spacer with which this report will be concerned is extremely light (\approx density 10 pounds/cubic foot) and satisfactorily stabilises the sandwich skins.

The main electrical requirements of a good core material are:-

- (a) that the dielectric constant should be as low as possible. This will ensure that the reflection at each interface will be reduced to a minimum and will increase the permissible thickness of the material;
- (b) that the loss angle should be as low as possible. The power losses and the subsequent range and performance of the radar equipment depend directly on the magnitude of the loss tangent;
- (c) that the water absorption of the material be kept as low as possible. The high dielectric constant of water (\approx 80 at radar frequencies) means that the presence of small amounts in the radome will seriously upset its performance.

1.2 The effect of the structure and composition of vulcanizates on their dielectric properties

Research has repeatedly shown that the behaviour of many elastic high polymers (e.g. rubber, polyvinyls, neoprenes, Bunas, etc.) placed in an alternating electric field is very similar to that of polar liquids. The phenomena of dielectric dispersion and absorption to which these substances give rise is generally explained by Debye's theory. This theory is based on the idea of dielectric viscosity, characterizing the resistance offered by the ambient medium to the rotation of the dipoles under the action of the electric field.

The dipoles responsible for dielectric losses in rubber are formed during vulcanization when sulphur becomes attached to the molecular chains of the polymer. According to Schallmach¹, only the intramolecular atoms of sulphur contribute to the dielectric absorption of rubber. The other ingredients of mixes, present in small quantities, influence the dielectric properties of vulcanizates only to the extent that they modify the combination of the sulphur with rubber².

On the other hand, fillers exert an important direct action on the dielectric properties of the mixes when the latter contain them in relatively large proportions as is frequently the case in practice. In this respect a distinction is usually made between mineral fillers such as whiting, which can be regarded as a dielectric, and strongly conductive fillers such as carbon blacks. In both cases the dielectric constant is increased by the presence of filler, but the cause of this increase depends on the magnitude of the electrical resistivity of the filler.

In mineral fillers, such as chalk, kaolin, etc., the dielectric constant of the particles is generally much higher than that of the surrounding rubber matrix. Thus the dielectric constant of the mix lies between that of the rubber and that of the filler.

With conductive fillers such as carbon blacks, it appears difficult to apply the foregoing theory to the rubber mix. The idea of dielectric constant applied to each particle has little physical significance in this case, nor is there any reason to regard the filler aggregate as homogeneous dielectric elements. One theory which has been suggested consists of treating each particle as a single conductor. Each pair of neighbouring particles then forms a microscopic condenser, the juxtaposition of which in series and in parallel with other such condensers, together produce the dielectric properties of the compound. On the other hand various authors have observed that the presence of carbon black in rubber mixes generally introduces large additional dielectric losses, which vary with the degree of dispersion of the black³. It has been shown that the total dielectric losses in a mixture can be higher than the total losses in the rubber matrix and those due to the direct conductivity of the mix⁴. Some phenomenon other than the conductivity of the black or the dielectric viscosity of the rubber therefore helps to increase the absorption. This has been attributed to the Maxwell-Wagner effect resulting from interfacial polarization between the rubber and the particles of black⁵. Such a hypothesis seems hardly tenable since it demands the attribution of a separate dielectric constant to each particle.

A variation in the dielectric viscosity of the polymer itself has been observed when mixtures of natural rubber or Butaprene contain carbon black, although this filler did not appear to influence the dielectric viscosity of mixtures based on neoprene⁵.

Although in this note no systematic study has been made of the effect of various constituents on the dielectric properties of the materials examined, the results obtained were compared on a broad basis with the results obtained elsewhere for similar mixes.

2 Description of samples

Five different series of core materials were tested. As the expanded material could not be produced in thicknesses suitable for test in the available apparatus, sheets of the unexpanded material were provided from which suitable specimens could be cut.

Series A was designated LM.724 and was a high softening point compound. The reference numbers were B5, B7, B8.

Series B was designated LM.1351 and was a medium softening point compound. The reference numbers were B6, B9 and B10.

Series C was designated H-10. The reference numbers were B11, B12 and B13.

Series D was designated HEE/SS/NA (without black) Hycar base. The reference numbers were B14, B15 and B16.

Series E was designated HEE/SS/NA Hycar base with carbon black. The reference numbers were B23 and B24.

3 Method of testing and conditioning of samples

The method of measurement was that of Hartshorn and Ward which is incorporated in the Tinsley - N.P.L. Radio Frequency Dielectric Test set. Details of the method and of the working of the set are given in a previous note⁶. This note also gives details of the size of sample, frequency, temperature and relative humidity control and of the conditioning of the samples.

In addition to measurements made on all samples in the dry state over a range of frequencies from 210 Kc/s to 49 Mc/s, series A-D inclusive were tested at 210 Kc/s after exposure for varying periods to an atmosphere of high humidity at 40°C and subsequently to a dry atmosphere.

4 Results

Table I shows the effect of humidity and temperature on the dielectric properties of Series A. The values of dielectric constant and loss tangent are tabulated against time of wetting or drying. Table II, III and IV show similar results for Series B, C and D.

Table V shows the values of dielectric constant and loss tangent of the dry samples at various frequencies.

Figs.1 to 5 correspond to the values in Tables I-V.

Figs.6 to 8 show the variation with frequency of the electrical properties of a "dry" sample from each series, as outlined in Table V. Series C is not included, as only three measurements were obtained in the frequency range.

5 Discussion of Results

5.1 General

In no case was the original "dry" value of dielectric constant regained after cycling.

Series A. This series showed slow deterioration over 1300 hours and a return to the original values of $\tan \delta$ in approximately the same time.

Series B. This series showed deterioration over 1300 hours of wetting but regained a stable value of $\tan \delta$ on drying in less than half this time, although this value was slightly higher than in the original "dry" condition.

Series C. This series had very high values of dielectric constant and the original "dry" values were not regained after drying, although the loss tangent figures were practically unaffected by cycling.

Series D. This series showed a good rate of recovery of properties in the first 24 hours of drying (reduction of $\tan \delta \approx 20\%$).

From Figs.6 to 8 it will be seen that the values of ϵ' and $\tan \delta$ for all samples decreased with increasing frequency with the exception of Series C, in which the loss tangent increased with frequency, after passing through a minimum at 1.55 Mc/s.

5.2 Effect of structure and constituents

The constituents of Series C, D and E were known. Series C and E contained carbon black as a filler. In addition, Series C contained dibutyl phthalate as plasticiser. Series D was a mix similar to Series E but contained no carbon black.

Dixie 20, the carbon black used in Series C and E, is a semi-reinforcing furnace black with a stated average particle size of 800Å. The specific surface is 29 m²/gm and its specific resistance is 4.3 ohms-cm⁷. These characteristics show that the effect of Dixie 20 in a mix should be somewhat analagous to the behaviour of fine thermal black (average size 900Å) as investigated by Thirion and Chasset⁸. Since the amount of carbon in both series is only of the order of 10% by volume, then by comparison with the behaviour of a fine thermal mix of 10% by volume, the carbon should not have much effect, if any, on the dielectric properties of the mixes. From the results in Tables III, IV and V it will be seen that the losses for the loaded mixes are actually less than for the pure mix. A possible explanation is that the amount of carbon in the mixes is too small to affect the losses. Furthermore, the unloaded mix contains a greater concentration of sulphur than the loaded ones and consequently shows greater loss. The high dielectric constant of Series C is probably due to the polar nature of the dibutyl phthalate, which is present in a considerable quantity, in addition to the carbon filler.

The literature in general, indicates that losses due to the direct current conductivity of the carbon only occur at low frequencies and should disappear at radar frequencies. On the other hand, the increase in dielectric constant caused by the introduction of a conductive filler does not seem to disappear so readily at higher frequencies, the dielectric constant of the loaded material always being higher than that of the unloaded mix. Series C has a lower sulphur concentration than Series E (21.7% to 28.0%) and this is shown up by the fact that Series C samples have smaller losses than Series E. The foregoing comparisons indicate the trend followed by the samples, viz:-

- (a) the dielectric constants of Series C and E are higher than those of Series D, because the former contain conductive fillers;
- (b) the dielectric constant of Series C is higher than that of Series E, because of additional dipoles contributed by the dibutyl phthalate in the former and by the more polar Hycar which has been substituted by natural rubber in Series E.
- (c) the loss tangent results increase with increasing sulphur concentration, Series C having the lowest sulphur and the lowest loss tangent.
- (d) The effect of the carbon fillers on the loss tangent is probably small. It cannot be detected by comparison with the unfilled mix because the higher sulphur concentration increases the observed losses.

6 Calculation of the dielectric constants of expanded materials

As pointed out previously, the samples tested were unexpanded. Normally, the materials are used in the expanded form so estimates were made of the dielectric constants of the materials when expanded to a density of 10 pounds/cubic foot. The density of each samples was measured and a typical sample chosen from each Series.

The dielectric constant of the expanded materials were then calculated from the formula:-

$$\epsilon_{\text{(expanded)}} = \epsilon_{\text{(unexpanded)}}^V \epsilon_{\text{air}}^{1-V}$$

where ϵ = dielectric constant

and V = fractional volume of unexpanded material which would be required for one volume of the expanded material.

Table VI shows the dielectric constants of the unexpanded material and the calculated value for the expanded material. A comparative experimental figure is quoted for Series C. Although this figure was obtained at radar frequencies the agreement was good as the dielectric constant of the expanded rubber mixes does not vary a great deal with frequency⁹. No estimate can be made of the loss tangent of the unexpanded material.

The dielectric constant and loss tangent values for an approved core material are:-

$$\epsilon' = 1.175 \pm 0.025$$

$$\tan \delta \leq 0.0010.$$

Compared with these values, Series C would not be approved. Series A, B and D have the required dielectric constant values but could not be approved until the loss tangent values of the expanded materials had been determined.

These "expanded" values for dielectric constant are however calculated for the dry materials and if the wet material values were used, Series A, B and D might then fall into the "non-approved" category.

7 Conclusions

All specimens tested showed deterioration of dielectric properties on exposure to humidity and temperature, the loss tangents in the case of Series A and B increasing to 300% of their dry values, and in the case of Series C and D to 250%. Although in each case the loss tangent values were regained on drying, the dielectric constants all returned to values higher than the original.

The loss tangents of all the samples except those of Series C decreased with increasing frequency. The values for Series C increased with increasing frequency after reaching a minimum at 1.55 Mc/s.

The general characteristics of the materials are in keeping with previous reports on the dielectric properties of vulcanizates.

The formula used to estimate the dielectric constant of the expanded material seemed to give fairly good agreement with an experimental value.

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Attached:-

Tables I to VI.
Drgs.Nos. Chem.1907 to 1914.
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TABLE I
Series A
The effect of humidity and temperature on the dielectric properties

Treatment	Time (Hours)	B5		B7		B8	
		ϵ'	$\tan \delta$	ϵ'	$\tan \delta$	ϵ'	$\tan \delta$
Drying over P_2O_5		-	-	3.675	0.0151	3.446	0.0155
		3.333	0.0152	3.440	0.0137	3.242	0.0130
		3.305	0.130	3.330	0.0132	3.210	0.0131
"Dry" value	0	3.305	0.0130	3.330	0.0132	3.210	0.0131
100% R.H. and 40°C	54	3.508	0.0207	3.581	0.0218	3.627	0.0223
	216	3.749	0.0403	3.913	0.0336	3.921	0.0435
	552	4.161	0.0511	-	-	-	-
	816	4.198	0.0495	4.587	0.0428	4.128	0.0524
	1200	4.420	0.0424	4.590	0.0432	4.710	0.0434
	1296	4.460	0.0428	4.610	0.0417	4.640	0.0440
Drying at 0% R.H. (i.e. over P_2O_5) at 40°C	24	4.204	0.0415	4.360	0.0410	4.330	0.0410
	96	3.890	0.0257	4.103	0.0330	4.035	0.0380
	336	3.542	0.0193	3.656	0.0185	3.695	0.0191
	504	3.492	0.0140	3.611	0.0146	3.588	0.0151
	720	3.399	0.0139	3.510	0.0136	3.523	0.0138
	1224	3.444	0.0132	3.694	0.0131	3.514	0.0134

TABLE II

Series B

The effect of humidity and temperature on the dielectric properties

Treatment	Time (Hours)	B6		B9		B10	
		ϵ'	$\tan \delta$	ϵ'	$\tan \delta$	ϵ'	$\tan \delta$
Drying over P ₂ O ₅		3.028	0.0183	3.054	0.0182	3.113	0.0169
		2.749	0.0147	3.074	0.0169	2.683	0.0139
		3.015	0.0169	3.075	0.0175	2.940	0.0162
"Dry" value	0	3.015	0.0169	3.075	0.0175	2.940	0.0162
100% R.H. and 40°C	54	3.022	0.0287	3.366	0.0334	3.119	0.0320
	216	3.651	0.0400	4.093	0.0519	3.382	0.0528
	552	3.650	0.0465	-	-	-	-
	816	4.058	0.0507	4.552	0.0592	3.859	0.0486
	1200	4.650	0.0488	4.950	0.0510	5.070	0.0536
	1296	4.710	0.0480	4.950	0.0490	5.020	0.0510
Drying over P ₂ O ₅ at 40°C	24	4.450	0.0440	4.610	0.0470	4.650	0.0460
	96	4.050	0.0386	4.111	0.0411	4.172	0.0402
	336	3.499	0.0173	3.514	0.0198	3.618	0.0194
	504	3.469	0.0183	3.514	0.0191	3.594	0.0192
	720	3.424	0.0177	3.537	0.0183	3.553	0.0182
	1224	3.494	0.0173	3.532	0.0187	3.596	0.0182

TABLE III

Series O

The effect of humidity and temperature on the dielectric properties

Treatment	Time (Hours)	B11		B12		B13	
		ϵ'	$\tan \delta$	ϵ'	$\tan \delta$	ϵ'	$\tan \delta$
Drying over P_2O_5		5.236	0.0148	5.234	0.0138	5.182	0.0148
		5.187	0.0128	4.912	0.0053	5.134	0.0135
		5.147	0.0118	5.047	0.0114	5.055	0.0121
"Dry" value	0	5.147	0.0118	5.047	0.0114	5.055	0.0121
100% R.H. and 40°C	54	5.292	0.0252	5.318	0.0262	5.324	0.0222
	216	6.662	-	6.311	-	6.423	0.0287
	816	7.339	0.0384	6.933	0.0383	7.110	0.0338
	1200	7.760	0.0320	7.680	0.0360	7.700	0.0340
	1296	8.310	0.0280	7.980	0.0290	8.270	0.0310
Drying over P_2O_5	24	7.750	0.0240	7.650	0.0240	7.660	0.0250
	96	7.463	0.0267	7.284	0.0272	7.143	0.0266
	336	6.259	0.0287	6.049	0.0298	6.019	0.0221
	504	5.824	0.0145	5.693	0.0145	5.741	0.0134
	720	5.706	0.0120	5.592	0.0120	5.704	0.0120
	1224	5.724	0.0112	5.550	0.0108	5.759	0.0114

TABLE IV
Series D

The effect of humidity and temperature on the dielectric properties

Treatment	Time (Hours)	B14		B15		B16	
		ϵ'	$\tan \delta$	ϵ'	$\tan \delta$	ϵ'	$\tan \delta$
Drying over P_2O_5		3.209	0.0291	2.657	0.0229	3.046	0.0271
		3.021	0.0272	2.658	0.0228	2.950	0.0268
		3.142	0.0272	2.681	0.0237	2.960	0.0261
'Dry' value	0	3.142	0.0272	2.681	0.0237	2.960	0.0261
100% R.H. and 40°C	54	3.149	0.0353	2.832	0.0290	3.161	0.0316
	816	3.265	0.0402	2.865	0.0341	3.236	0.0377
	1200	3.598	0.0567	3.261	0.0501	3.574	0.0543
	1226	4.030	0.0627	3.890	0.0670	3.520	0.0690
		4.170	0.0670	4.010	0.0600	4.204	0.0702
Drying over P_2O_5	24	4.070	0.0550	3.840	0.0540	3.990	0.0560
	96	3.747	0.0420	3.630	0.0460	3.728	0.0484
	336	3.516	0.0325	3.319	0.0306	3.454	0.0315
	504	3.427	0.0296	3.306	0.0285	3.426	0.0289
	720	3.411	0.0287	3.270	0.0270	3.390	0.0274
	1224	3.426	0.0277	3.290	0.0268	3.386	0.0279

TABLE V
Dielectric constants and loss tangents values of dry samples at different frequencies

Frequency	B5	B6	B7	B8	B9	B10	B11	B12	B13	B14	B15	B16	B23	B24
	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$	ϵ' $\tan \delta$
211 Kc/s	4.187	4.259	4.227	4.224	4.257	4.300	6.769	6.654	6.703	3.811	3.780	3.761	5.911	5.826
435 Kc/s	4.144	4.166	4.184	4.191	4.212	4.240	6.668	6.523	6.630	3.718	3.665	3.686	5.807	5.798
728 Kc/s	4.117	4.124	4.175	4.151	4.169	4.196	6.612	6.528	6.636	3.673	3.646	3.654	5.896	5.795
1.55 Mc/s	4.074	4.090	4.142	4.118	4.112	4.127	6.566	6.469	6.554				5.703	5.691
7.4 Mc/s	4.033	3.989	4.066	4.055	4.002	4.035	6.467	6.315	6.415				5.555	5.530
15.4 Mc/s	3.987	3.948	4.037	4.026	3.970	4.002	6.162	6.269	6.380				5.505	5.470
33 Mc/s	3.976	3.915	4.026	4.005	3.939	3.953	6.105	6.232	6.319				5.442	5.405
49 Mc/s	3.957	3.906	4.008	3.988	3.910	3.943	6.308	6.186	6.271				5.433	5.385
211 Kc/s	0.0134	0.0197	0.0138	0.0140	0.0194	0.0188	0.0121	0.0141	0.0121	0.0293	0.0343	0.0284	0.0236	0.0231
435 Kc/s	0.0131	0.0187	0.0133	0.0133	0.0201	0.0189	0.0116	0.0115	0.0113	0.0287	0.0289	0.0283	0.0235	0.0243
728 Kc/s	0.0125	0.0182	0.0124	0.0129	0.0188	0.0183	0.0114	0.0113	0.0112	0.0276	0.0276	0.0279	0.0221	0.0243
1.55 Mc/s	0.0124	0.0174	0.0127	0.0122	0.0178	0.0178	0.0112	0.0116	0.0118				0.0217	0.0224
7.4 Mc/s	0.00981	0.0155	0.0108	0.0108	0.0161	0.0158	0.0134	0.0135	0.0137				0.0187	0.0191
15.4 Mc/s	0.00902	0.0136	0.00944	0.00971	0.0138	0.0138	0.0169	0.0158	0.0161				0.0174	0.0179
33 Mc/s	0.00869	0.0128	0.00848	0.00856	0.0125	0.0122	0.0225	0.0217	0.0216				0.0153	0.0159
49 Mc/s	0.008371	0.0118	0.00798	0.00817	0.0116	0.0120	0.0259	0.0261	0.0259				0.0151	0.0157

TABLE VI

Calculated values for dielectric constant of expanded material

Series	Sample No.	Density lb/cu ft	Dielectric Constant (unexpanded)	Dielectric Constant (expanded) Calculated	Dielectric Constant (expanded) Experimental
A	B7	84.375	3.514	1.161	-
B	B9	83.125	3.542	1.161	-
C	B11	78.75	5.724	1.248	1.245 (R.R.E.)
	B12	78.75	5.550	1.243	
	B13	79.375	5.759	1.248	
D	B15	77.5	3.426	1.172	-

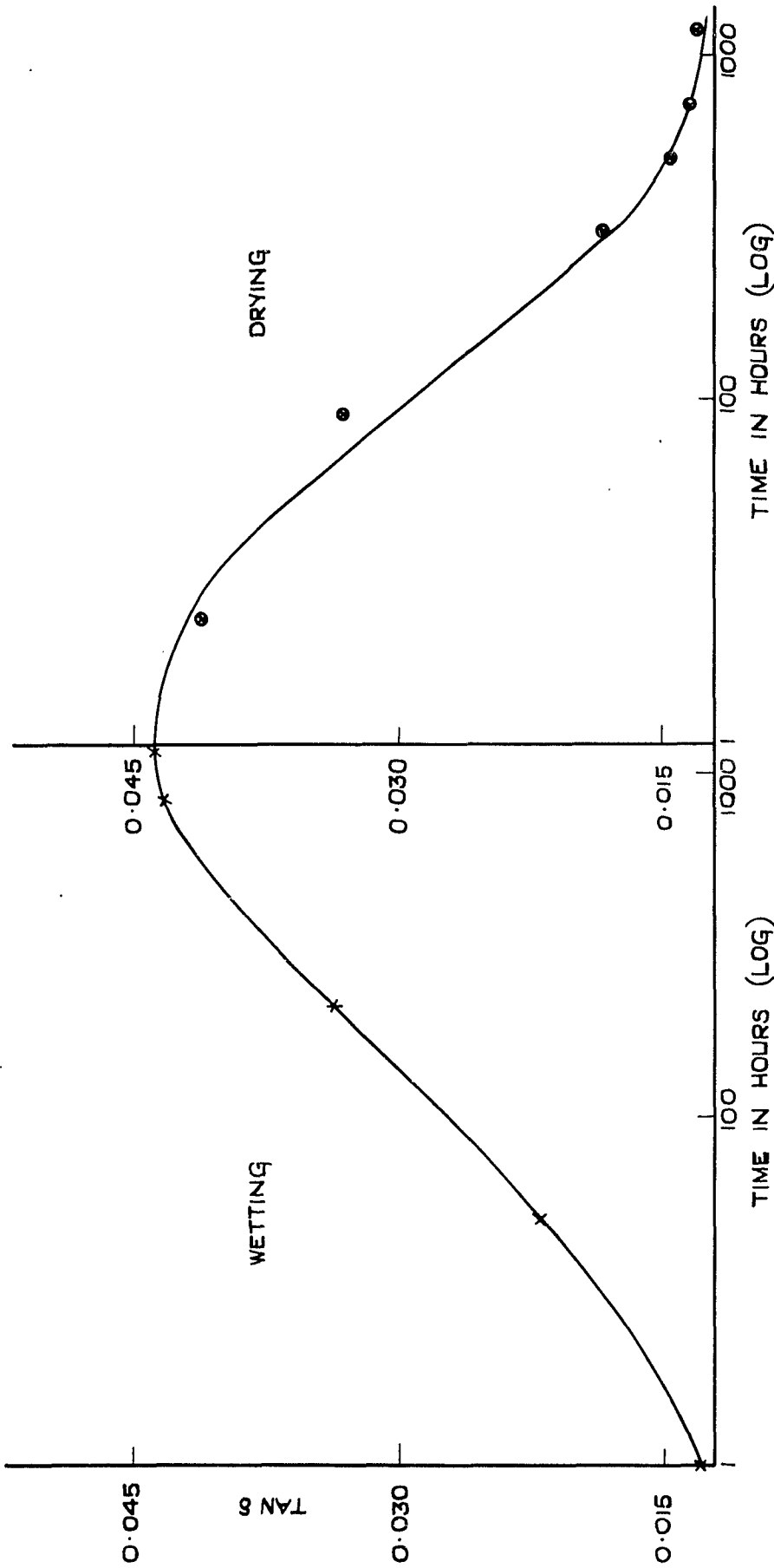


FIG. 1. VARIATION OF LOSS TANGENT FOR NO B7. (SERIES A) WITH TIME OF WETTING AND DRYING.

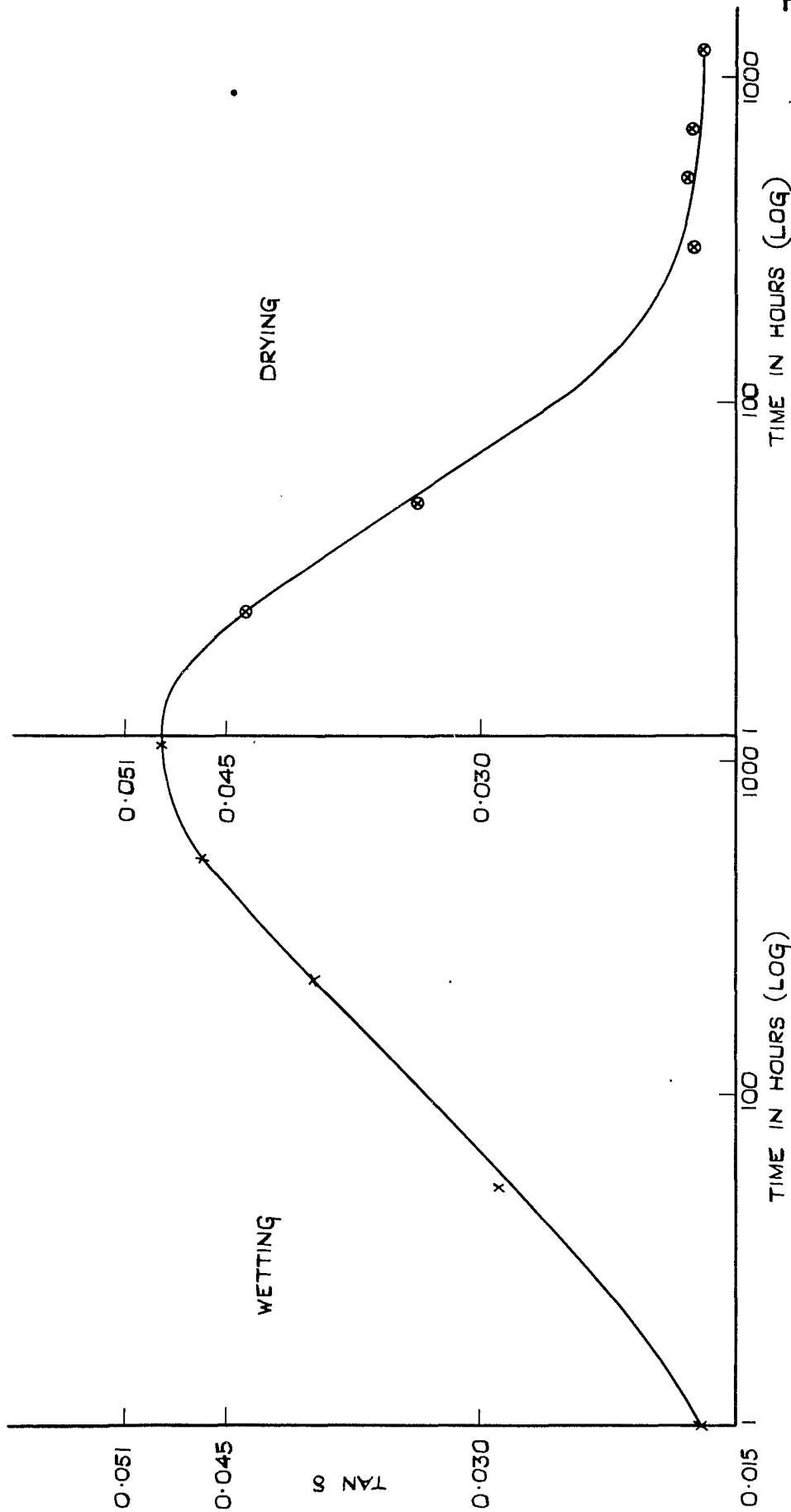


FIG. 2. VARIATION OF LOSS TANGENT FOR B6. (SERIES B) WITH TIME OF WETTING AND DRYING.

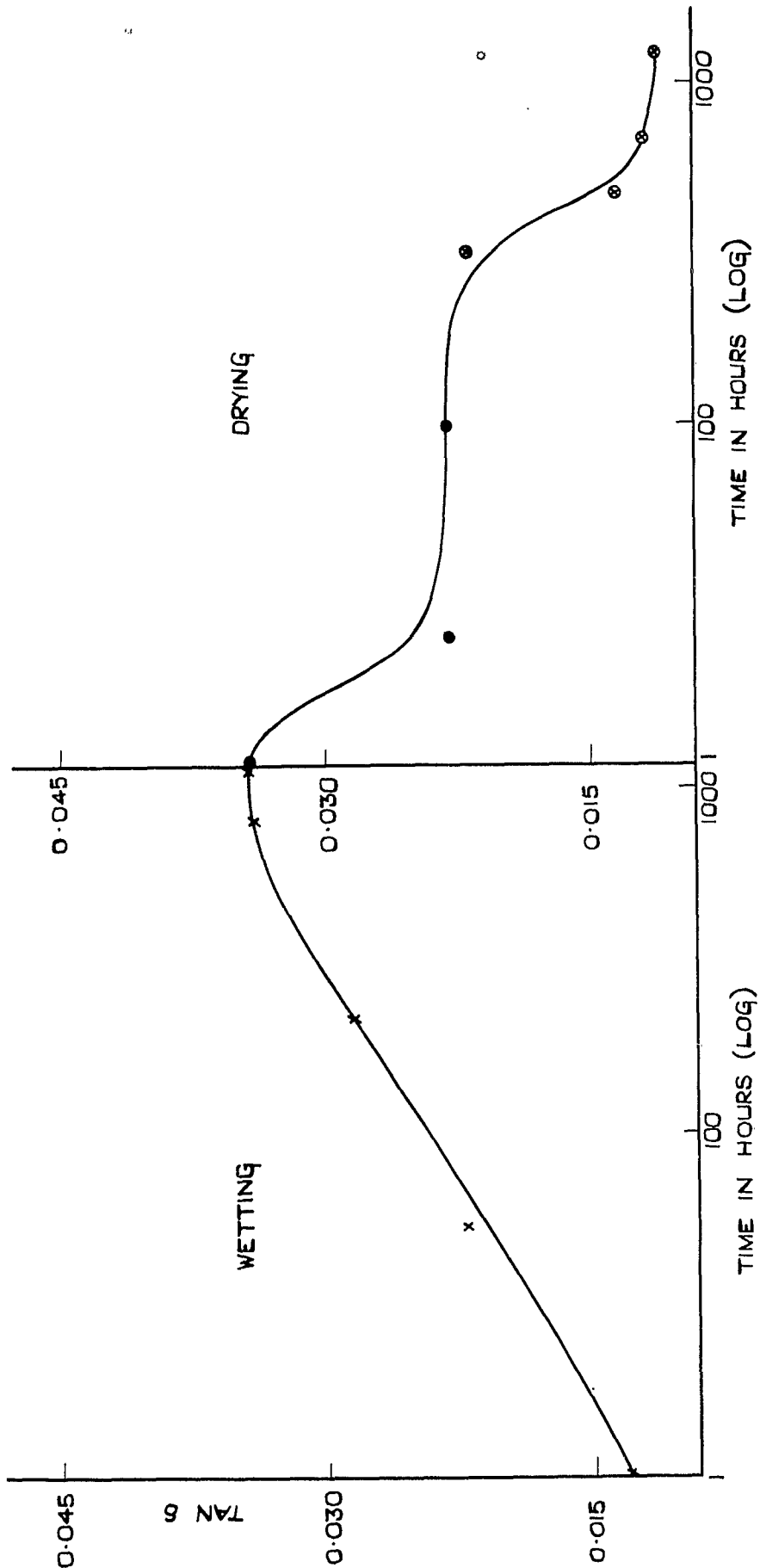


FIG. 3. VARIATION OF LOSS TANGENT FOR B 13. (SERIES C)
WITH TIME OF WETTING AND DRYING.

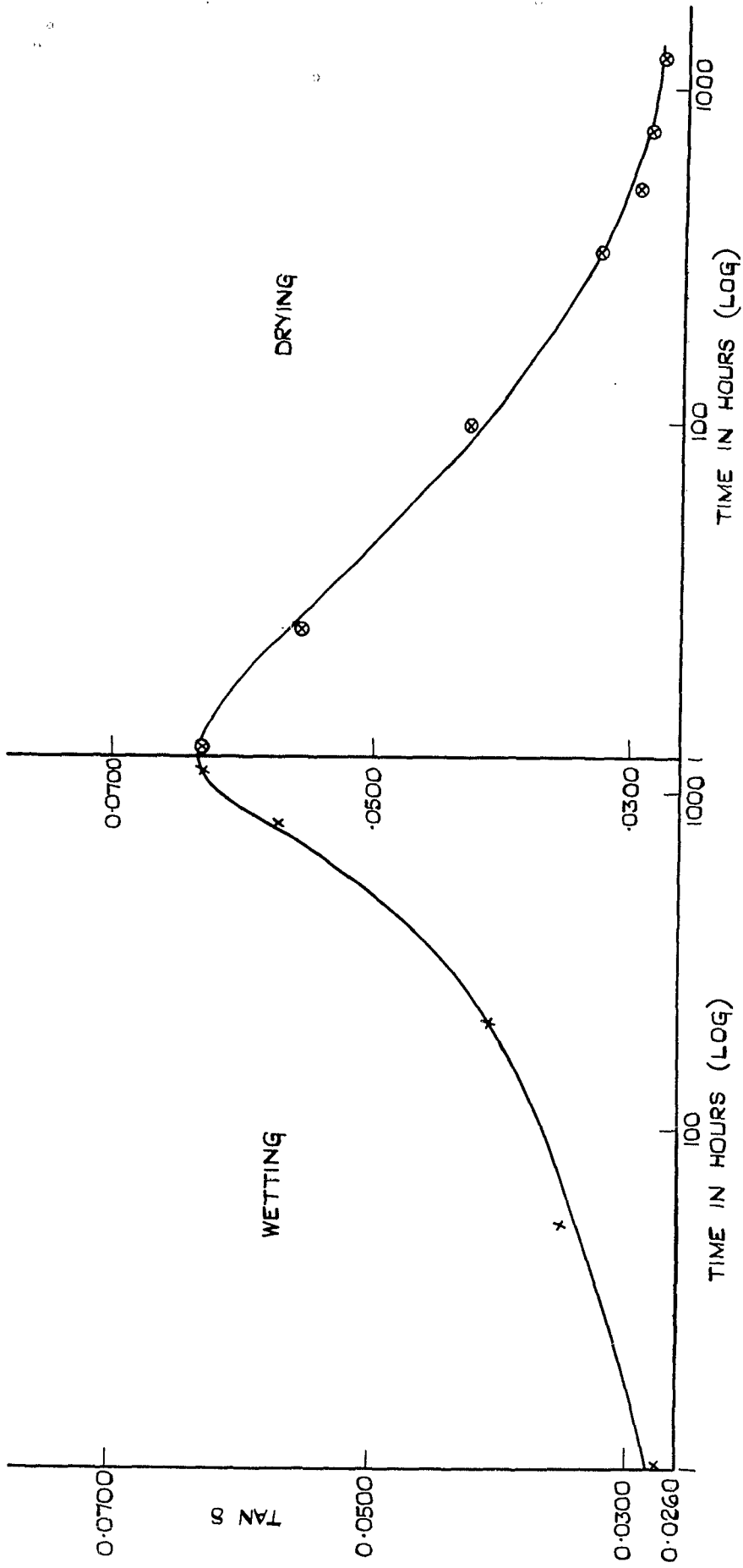


FIG. 4. VARIATION OF LOSS TANGENT FOR B14. (SERIES D)
WITH TIME OF WETTING AND DRYING.

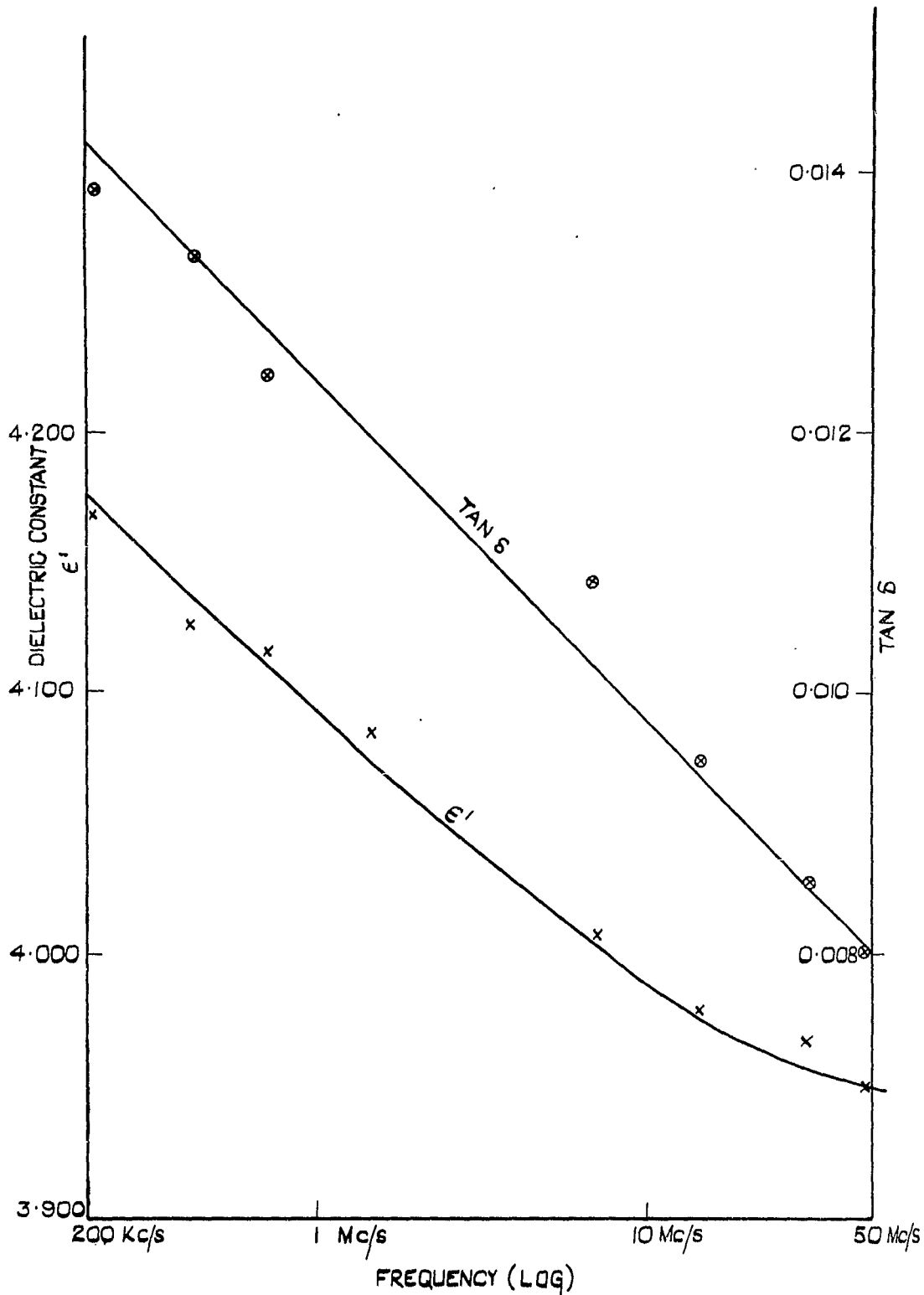


FIG. 5. VARIATION OF DIELECTRIC CONSTANT & LOSS TANGENT WITH FREQUENCY FOR SAMPLE NO B7. (SERIES A)

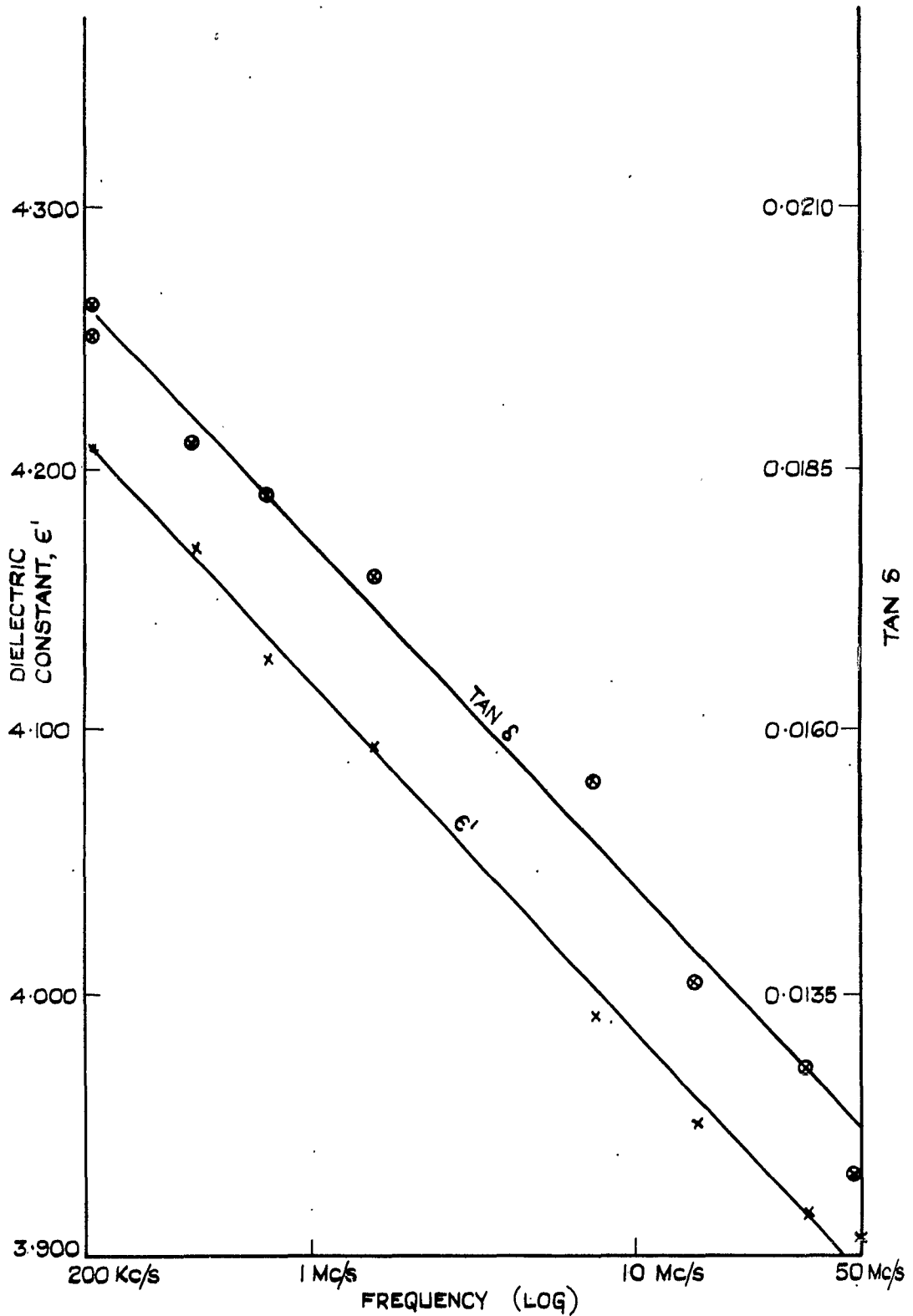


FIG. 6. VARIATION OF DIELECTRIC CONSTANT & LOSS TANGENT WITH FREQUENCY FOR SAMPLE N^o B6. (SERIES B)

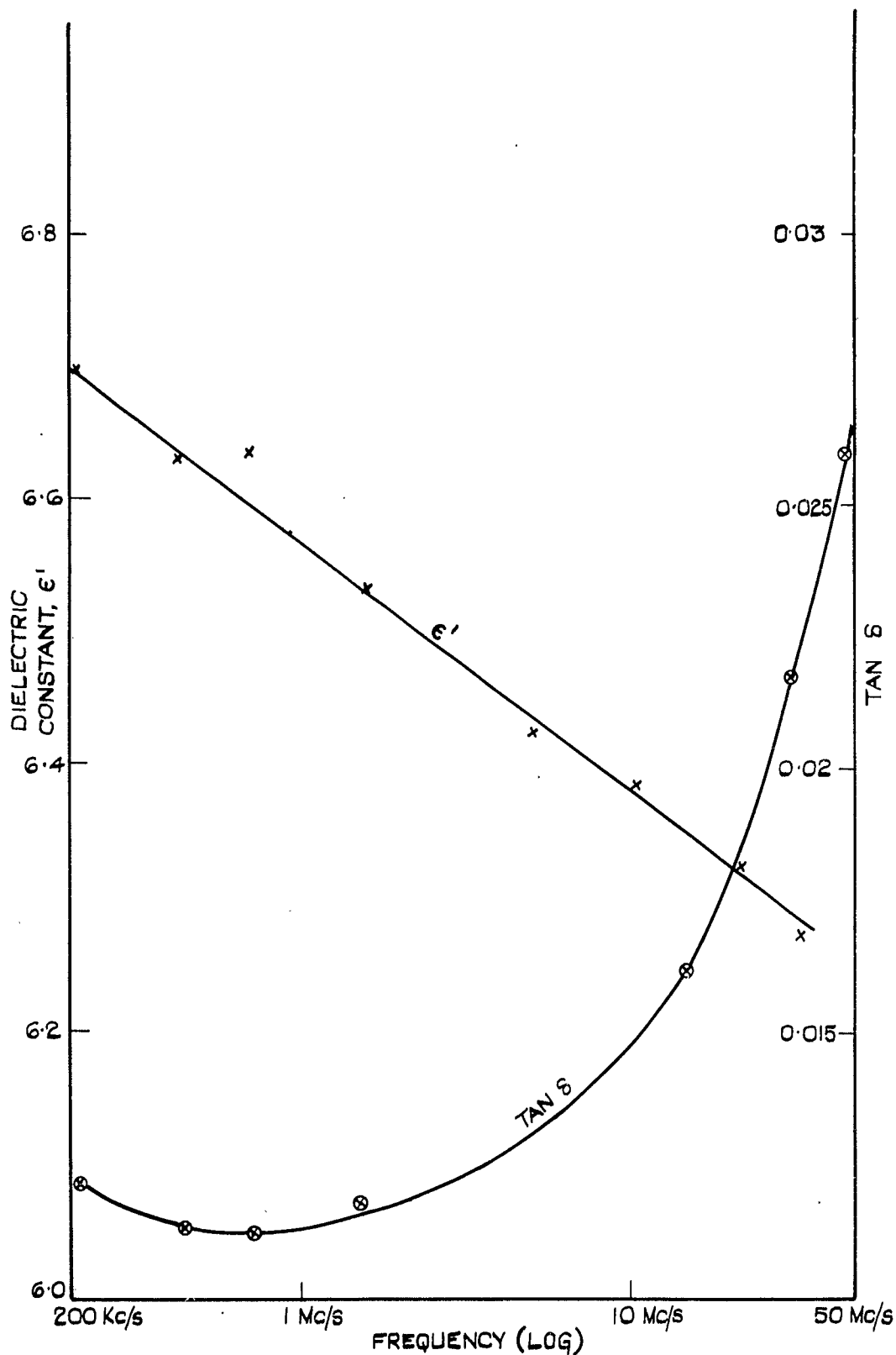


FIG. 7. VARIATION OF DIELECTRIC CONSTANT & LOSS TANGENT WITH FREQUENCY FOR SAMPLE NO B13. (SERIES C.)

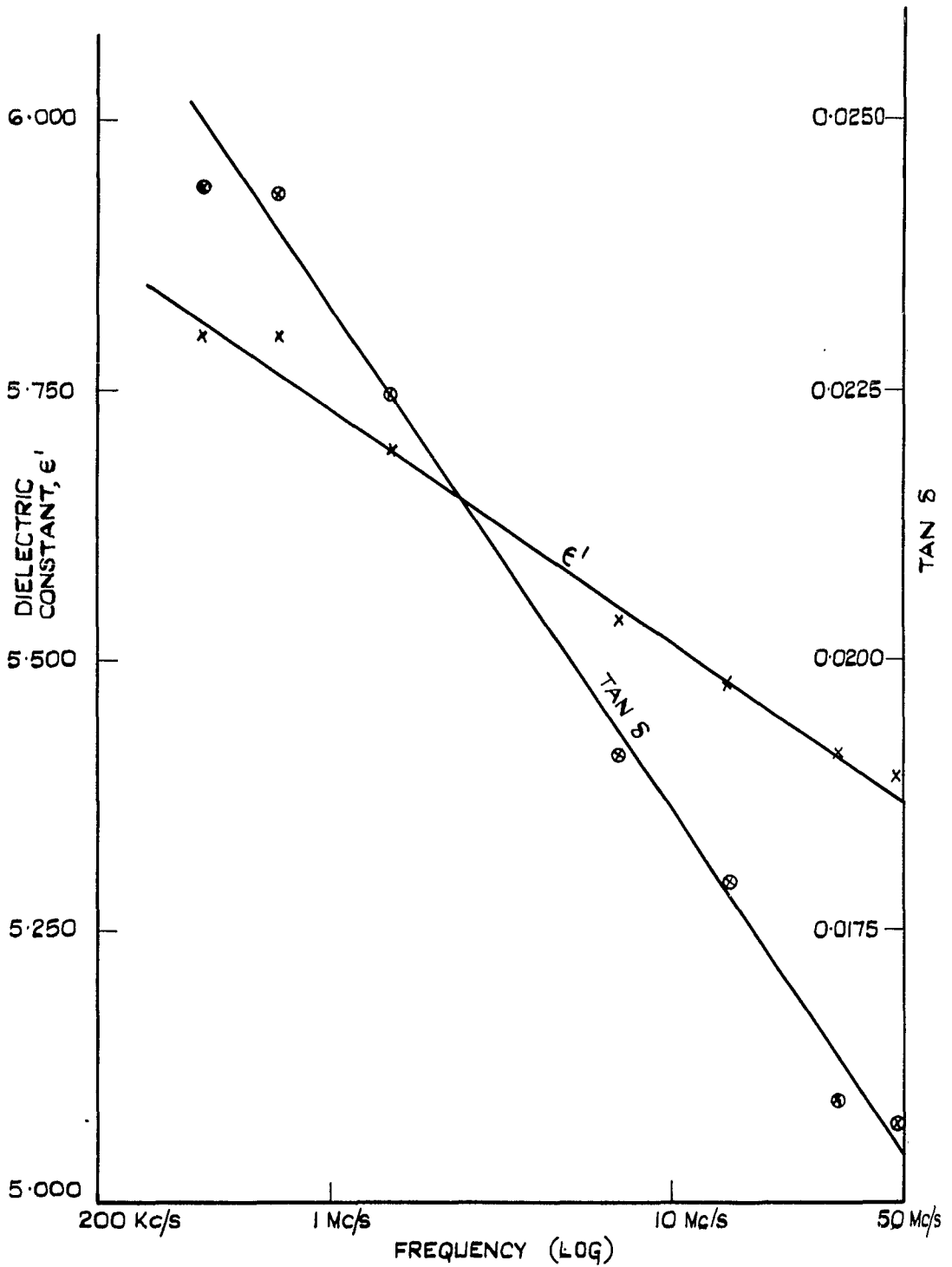


FIG. 8. VARIATION OF DIELECTRIC CONSTANT & LOSS TANGENT WITH FREQUENCY FOR SAMPLE N° B24. (SERIES E)



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