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Properties of Barium-Magnesium Titanate Dielectrics

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Dielectrics having compositions in the system $BaTiO_3$-4MgO:TiO_2-TiO_2 were matured (less than 0.1% of absorption) at 1,275° to 1,425° C. Data are given for the dielectric constant K at a frequency of 1 mc/s and various temperatures from −60° to +85° C, and for Q, the reciprocal of the power factor, at 25° C and frequencies of 50, 1,000, and 20,000 kc/s and 3,000 mc/s. Values of K (1 mc/s and 25° C) ranged from 12 to 1,550 and those of Q from 9 to 10,000. Values of K decreased, and those of Q increased for several weeks after specimens were matured, when the content of BaO was greater than 30 percent and that of TiO_2 less than 50 percent. Partial restoration of the original values of K and Q resulted from heating these specimens at various temperatures for brief periods. Linear thermal expansion (25° to 700° C) ranged from 0.46 to 0.71 percent. A few specimens of barium-strontium titanate were tested for the effects of thermal history on the properties.

I. Introduction

This is the second paper pertaining to ceramic dielectrics composed of titanium dioxide and the oxides of the alkaline earth elements. Previous work, by the present investigators, on barium-strontium titanate dielectrics [1] [1] indicated the usefulness of these materials in the fields of electrical communications and instrumentation.

Some of the properties of dielectrics having compositions in limited portions of the system BaO-MgO-TiO_2 have been determined by other investigators. Wainer [2] found that the addition of magnesia to barium titanate resulted in high electrical losses. Low losses, however, were observed by Rieke and Ungewiss [3] on bodies with compositions in a portion of the system MgO-TiO_2 (9 to 57 percent of TiO_2). Thus it might be expected that portions of the ternary system would represent compositions of bodies with low losses, and other portions would indicate compositions of bodies with high losses. It was anticipated also that many of these bodies would have a positive or neutral temperature coefficient of dielectric constant because of the low temperature coefficient of magnesium titanate reported by Rosenthal [4].

[1] Figures in brackets indicate the literature references at the end of this paper.

II. Preparation of Specimens and Methods of Test

In the preparation of specimens having the computed compositions shown in figure 1, chemically pure magnesium carbonate was the source of MgO. The titania, grade TMO, and barium carbonate were from the same stocks used in the production of barium-strontium titanates [1].

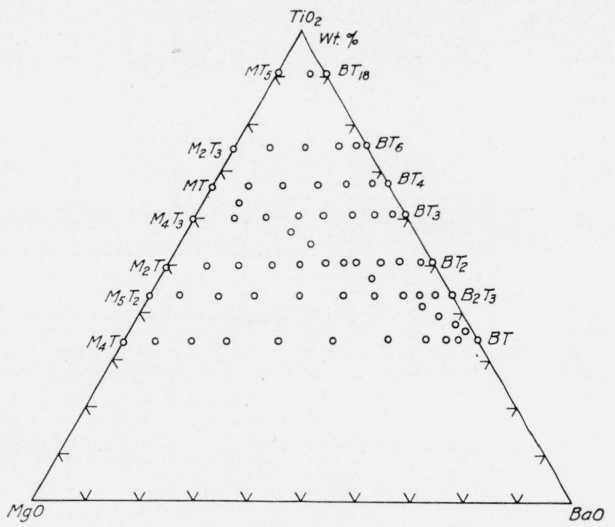

FIGURE 1. *Ternary diagram for system* BaO-MgO-TiO_2 *showing compositions studied.*

B=BaO; M=MgO; T=TiO_2.

In the first paper of this series, details are given of the method for producing mature specimens as indicated by less than 0.1 percent of absorption (water basis). The properties of the dielectrics were determined by the methods and equipment, with one exception, previously used. A crystal-controlled oscillator was installed in the apparatus for determining the effects of variation in temperature of the specimens upon the dielectric constant.

III. Results and Discussion

In table 1, data are given for the composition, heat treatment, absorption, shrinkage, dielectric constant (K), and Q-value (reciprocal of the power factor) of mature specimens.

TABLE 1. *Composition, heat treatment, absorption, shrinkage, dielectric constant, K, and Q, of bodies in the system* BaO-MgO-TiO_2

Specimen designation	Proportion of end members of join		Composition weight			Heat treatment			Absorption	Shrinkage	Dielectric constant, K, at 25° C and—				Reciprocal, Q, of power factor at 25° C and—			
						No. 1 for 1 hr at—	No. 2				50 kc/s	1,000 kc/s	20,000 kc/s	3×10^{-6} kc/s	50 kc/s	1,000 kc/s	20,000 kc/s	3×10^{6} kc/s
	MgO: $5TiO_2$	BaO: $18TiO_2$	BaO	MgO	TiO_2		Temperature	Time										
	Weight percent	*Weight percent*	*Percent*	*Percent*	*Percent*	*°C*	*°C*	*hr*	*Percent*	*Percent*								
MT5	100.0	0.0	------	9.2	90.8	1,100	1,285	1	0.06	16.7	47	47	47	46	400	710	1,130	840
18BM6	32.6	67.4	6.5	2.9	90.6	1,100	1,275	1	.01	13.0	81	78	74	-------	102	42	33	-------
BT18	0.0	100.0	9.6	-------	90.4	1,100	1,275	1	.00	15.0	75	74	74	-------	260	1,000	7,000	-------
	$2MgO: 3TiO_2$	$BaO: 6TiO_2$																
M2T3	100.0	0.0	------	25.2	74.8	1,100	[a] 1,290	3	.00	10.7	18	18	18	16	34	350	3,000	1,800
6BM2	74.0	26.0	6.5	18.2	74.9	1,100	1,250	1	.01	14.5	26	25	23	21	135	50	30	550
6BM5	47.3	52.7	13.0	11.9	75.1	1,100	1,275	1	.00	14.1	48	45	44	34	42	15	12	430
6BM8	20.0	80.0	19.5	5.1	75.4	1,100	1,275	1	.00	14.7	70	64	48	-------	11	11	8	-------
6BM9	4.9	95.1	23.0	1.5	75.5	1,100	1,275	1	.00	14.9	53	52	48	-------	97	32	25	-------
BT6	0.0	100.0	24.3	-------	75.7	1,100	1,275	1	.01	14.0	46	45	44	-------	85	400	2,200	-------
	$MgO:TiO_2$	$BaO:4TiO_2$																
MT	100	0	------	33.5	66.5	1,100	[a] 1,100	3	.00	12.9	18	17	17	17	200	4,000	>8,000	2,400
4BM2	80	20	6.5	26.5	67.0	1,100	1,275	1	.00	15.4	28	27	25	25	125	28	19	110
4BM4	60	40	13.0	20.0	67.0	1,100	1,300	1	.01	17.4	41	39	30	33	52	11	9	130
4BM6	40	60	19.5	13.4	67.1	1,100	1,300	1	.01	14.8	55	49	37	-------	44	9	8	-------
4BM7	25	75	24.3	8.4	67.3	1,100	1,300	1	.00	15.9	60	53	40	-------	27	9	9	-------
4BM9	10	90	29.2	3.3	67.5	1,100	1,300	1	.00	13.8	46	44	39	-------	260	22	18	-------
BT4	0	100	32.4	-------	67.6	1,100	1,330	1	.03	16.9	34	34	35	33	>1,600	2,000	3,700	2,600
	$4MgO: 3TiO_2$	$BaO:3TiO_2$																
M4T3	100	0	------	40.2	59.8	1,100	1,365	1	.00	15.0	16	16	16	16	300	4,000	2,300	3,000
3BM2	80	20	7.8	32.2	60.0	1,100	1,315	1	.00	15.7	18	18	18	17	1,500	9,000	3,600	1,100
3BM5	50	50	19.5	20.1	60.4	1,100	1,300	1	.02	15.5	29	27	24	24	54	19	27	120
3BM6	40	60	23.4	16.1	60.5	1,100	1,300	1	.00	17.1	34	31	26	24	37	13	18	110
3BM7	25	75	29.3	10.0	60.7	1,100	1,300	1	.01	17.3	45	39	32	32	27	8	14	320
3BM8	15	85	33.2	6.0	60.8	1,100	1,260	1	.00	17.5	50	47	37	-------	62	8	10	-------
3BM9	5	95	37.1	2.0	60.9	1,100	1,260	1	.02	18.5	38	37	35	-------	160	50	38	-------
BT3	0	100	39.0	-------	61.0	1,100	1,260	1	.03	17.4	44	44	43	42	650	720	800	460

[a] Heat treated previously (see table 2).

TABLE 1. *Composition, heat treatment, absorption, shrinkage, dielectric constant, K, and Q, of bodies in the system* $BaO\text{-}MgO\text{-}TiO_2$—Continued

Specimen designation	Proportion of end members of join		Composition weight			Heat treatment			Absorption	Shrinkage	Dielectric constant, K, at 25° C and—				Reciprocal, Q, of power factor at 25° C and—			
						No. 1 for 1 hr at—	No. 2											
	$2MgO:TiO_2$	$BaO:2TiO_2$	BaO	MgO	TiO_2		Temperature	Time			50 kc/s	1,000 kc/s	20,000 kc/s	3×10^{-6} kc/s	50 kc/s	1,000 kc/s	20,000 kc/s	3×10^{6} kc/s
	Weight percent	*Weight percent*	*Percent*	*Percent*	*Percent*	*° C*	*° C*	*hr*	*Percent*	*Percent*								
M2T	100	0	------	50.2	49.8	1,100	[a] 1,380	1	.00	13.9	14	14	14	14	2,200	10,000	10,000	3,000
2BM1	85	15	7.3	42.7	50.0	1,100	[a] 1,310	1	.00	17.0	16	16	16	16	4,000	10,000	10,000	1,000
2BM2	73	27	13.2	36.6	50.2	1,100	[a] 1,310	1	.01	16.0	15	15	15	17	1,600	6,000	8,000	180
2BM4	60	40	19.6	30.1	50.3	1,100	1,285	6	.01	16.7	24	24	24	------	750	800	700	------
2BM5	50	50	24.5	25.1	50.4	1,100	1,260	3	.00	11.9	32	32	31	28	>1,200	1,100	700	47
2BM6	40	60	29.4	20.1	50.5	1,100	1,260	3	.02	12.7	31	31	32	------	220	410	270	------
2BM7	30	70	34.3	15.1	50.6	1,100	1,295	1	.06	17.9	33	33	33	34	1,400	580	360	53
2BM8	20	80	39.2	10.0	50.8	1,100	1,295	1	.03	18.3	32	32	32	33	2,000	740	500	80
2BM87	13	87	42.6	6.5	50.9	1,100	1,295	1	.00	16.1	39	39	39	37	1,400	660	390	75
2BM9	5	95	46.6	2.5	50.9	1,100	1,295	1	.00	18.1	111	110	110	------	130	88	47	------
BT2	0	100	49.0	------	51.0	1,100	1,290	1	.02	16.4	204	200	197	------	100	70	35	------
	$5MgO:2TiO_2$	$2BaO:3TiO_2$																
M5T2	100	0	------	55.8	44.2	1,245	1,415	1	.00	15.8	14	14	14	14	2,200	5,000	10,000	3,400
2B5M1	90	10	5.6	50.2	44.2	1,245	1,350	1	.00	12.6	16	16	16	16	>1,500	10,000	10,000	570
2B5M2	77	23	12.9	43.0	44.1	1,245	1,350	1	.00	16.9	18	18	18	16	>1,700	10,000	10,000	340
2B5M3	65	35	19.6	36.3	44.1	1,245	1,300	1	.00	14.9	26	26	26	26	800	680	800	60
2B5M5	50	50	28.1	27.9	44.0	1,100	1,300	1	.00	16.9	47	47	46	------	270	235	130	------
2B5M6	35	65	36.5	19.5	44.0	1,100	1,300	1	.00	16.3	117	116	116	------	770	630	245	------
2B5M7	25	75	42.1	14.0	43.9	1,100	1,300	1	.02	16.9	159	155	154	------	780	430	180	------
2B5M9	10	90	50.5	5.6	43.9	1,100	1,265	1	.00	16.3	207	204	201	------	480	260	130	------
2B5M95	5	95	53.3	2.8	43.9	1,100	1,300	1	.02	14.7	227	225	215	------	1,500	195	110	------
B2T3	0	100	56.1	------	43.9	1,250	1,300	1	.01	10.8	910	900	890	------	70	50	25	------
	$4MgO:TiO_2$	$BaO:TiO_2$																
M4T	100.0	0.0	------	66.9	33.1	1,245	1,425	1	.00	17.2	12	12	12	12	2,500	9,000	7,000	3,100
BM1	90.0	10.0	6.6	60.2	33.2	1,245	1,350	1	.00	15.7	15	15	15	15	>1,200	10,000	5,000	870
BM2	81.0	19.0	12.5	54.2	33.3	1,245	1,350	1	.01	15.7	21	21	21	20	1,100	2,000	2,000	90
BM3	70.4	29.6	19.4	47.1	33.5	1,245	1,350	1	.01	11.2	37	37	37	36	600	600	300	20
BM4	55.9	44.1	29.0	37.4	33.6	1,245	1,350	1	.00	12.8	120	120	120	------	140	100	55	------
BM6	40.7	59.3	39.0	27.0	34.0	1,100	1,350	1	.01	11.6	250	250	250	------	230	400	150	------
BM7	25.5	74.5	48.9	17.0	34.1	1,100	1,365	1	.00	15.6	550	550	550	------	800	500	200	------
BM8	14.8	85.2	56.0	10.0	34.0	1,100	1,385	1	.00	15.4	850	850	850	------	700	900	360	------
BM9	10.0	90.0	59.1	6.7	34.2	1,100	1,385	1	.00	15.9	1,050	1,050	1,050	------	500	800	300	------
BM95	5.0	95.0	62.4	3.3	34.3	1,100	1,385	1	.00	16.6	1,550	1,550	1,550	------	510	560	770	------
BT	0.0	100.0	65.7	------	34.3	1,245	1,385	2	.05	10.6	1,400 ±200	1,400 ±200	1,400 ±200	------	100	130	70	------
	$MgO:TiO_2$	$BaO:TiO_2$																
MB1	90.0	10.0	6.6	30.0	63.4	1,100	1,290	1	.01	15.5	18	18	18	21	3,000	860	1,700	800
MB19	80.7	19.3	12.7	27.0	60.3	1,100	1,290	1	.00	16.9	19	19	19	19	3,000	7,000	10,000	1,550
MB30	70.0	30.0	19.5	23.5	57.0	1,100	1,290	1	.00	17.0	21	21	21	21	3,000	6,000	10,000	1,200
MB37	62.6	37.3	24.5	21.0	54.5	1,100	1,275	1	.03	13.3	20	20	20	19	1,900	3,000	5,000	260
MB48	51.8	48.2	32.5	17.0	50.5	1,100	1,290	1	.01	15.1	41	41	40	------	750	750	400	------
MB59	40.3	59.7	39.0	13.5	47.5	1,100	1,290	1	.01	15.7	94	94	96	------	500	340	130	------
MB70	30.4	69.6	47.0	9.5	43.5	1,100	1,300	1	.01	15.9	200	200	200	------	600	250	110	------
MB79	20.9	79.1	51.8	7.0	41.2	1,100	1,315	1	.01	15.3	350	350	350	------	800	300	130	------
MB85	14.3	85.7	56.0	4.8	39.2	1,100	1,315	1	.01	17.2	540	540	540	------	2,000	300	180	------
MB91	8.6	91.4	60.0	2.8	37.2	1,100	1,300	1	.00	17.2	800	800	800	------	450	600	300	------
MB95	4.9	95.1	62.5	1.5	36.0	1,100	1,300	1	.00	16.2	1,080	1,080	1,080	------	400	600	300	------

[a] Heat treated previously (see table 2).

The data for a given composition are considered to be the most representative among those obtained from measurements of 4 to 10 specimens.

Measurements at 3,000 mc/s and 25° C, made with the coaxial wave-guide instrument, gave values of K and Q that may be in error by a few percent for specimens of high dielectric constant. In testing such specimens with this instrument, the accuracy of measurements is dependent largely upon the exact determination of the average diameter of the central hole through the specimen. The relation of composition of the specimens to approximate maturing temperature is shown in figure 2. No attempts were made to determine the range in temperature for the production of mature specimens made from the individual preparations. When the compositions were in the region of $2MgO:3TiO_2$, the specimens were difficult to mature. Despite systematic variations in the duration and final temperature of the heat treatments, these specimens had 0.2 to 0.8 percent of absorption. Reheating the specimens, however, to the same or higher temperatures was effective in reducing the absorption, as illustrated by the data in table 2. These compositions were near that of a eutectic in the system $MgO\text{-}TiO_2$[5].

The effects of systematically varying the composition of the specimens upon the values of K and Q may be obtained from the data in table 1. For example, when the content of

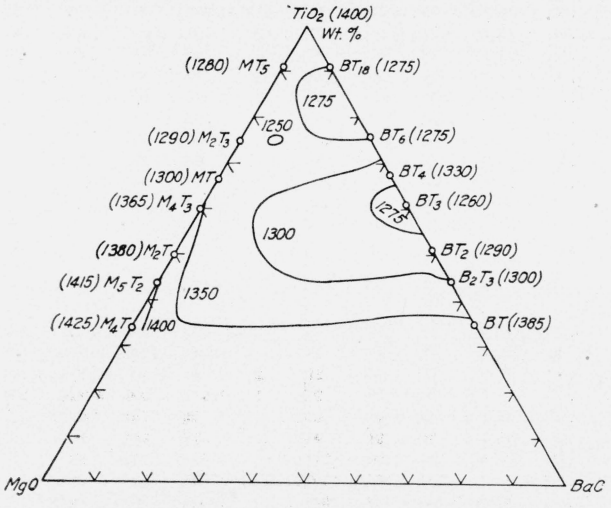

FIGURE 2. *Approximate maturing temperature (° C) after calcining treatment.*

B=BaO; M=MgO; T=TiO_2.

TABLE 2. *Effect of consecutive heat treatments at the same or higher temperatures upon the absorption of some specimens*

Specimen designation	Heat treatment		Absorption
	Temperature	Time	
	° C	hr	Percent
M2T3	1,275	3	0.54
	1,285	3	.53
	1,290	3	.00
MT	1,290	3	.22
	1,300	3	.00
M2T	1,350	1	1.02
	1,370	1	0.87
	1,380	1	.00
2BM1	1,310	1	.15
	1,310	1	.00
2BM2	1,310	1	.40
	1,310	1	.01

TiO_2 is maintained at a given percentage within the range 60 to 90 and MgO is substituted for BaO, the values of Q are affected more than those of K. Substituting a few percent (3 to 8) of MgO for BaO causes a rapid decrease in the values of Q from several hundred to 8 or 22 at a frequency of 1 mc/s and 25° C. Further substitution of MgO for BaO results in a gradual increase in the values of Q, which become high (350 to 4,000) when the substitution is complete.

A similar substitution of MgO for BaO causes slight increase in the relatively low values of K (34 to 74), followed by a decrease to the values of K (16 to 47) characteristic of specimens with compositions in this portion of the system $MgO\text{-}TiO_2$ (60 to 90 percent of TiO_2).

For specimens having compositions in the remainder of the system investigated, 33 to 60 percent of TiO_2, the values of both K and Q are affected greatly by the substitution of MgO for BaO. In this region of compositions, there is a continuous decrease in the values of K from several hundred, characteristic of the barium-titanate specimens, to the low values of 12 to 14 typical of the specimens of the magnesium titanates. The variation in Q-values is typified by a gradual rise and fall followed by a rapid rise to the high values (5,000 to 10,000) characteristic of the magnesium titanate specimens with a content of TiO_2 less than 60 percent.

Some of the specimens having a relatively high content of BaO exhibited changes in the values of K and Q with time after the final heat treatment.

The data on K and Q in table 1, however, were obtained when such specimens had aged for 6 months or more, and no further changes in these properties were observed.

The stability of the dielectric constant and power losses with respect to time was determined by remeasuring the values of K and Q at 1 mc/s and 25° C for specimens stored 6 months at room temperature. Specimens containing more than 30 percent of BaO and less than 50 percent of TiO_2 had lower values of K and higher values of Q than when freshly prepared, as shown by the data in table 3. The changes in K and Q for specimen BM6 are illustrated in figure 3. These changes are reversible, because a partial restoration of the original values of K and Q occurred when the specimens were reheated to 600° or 700° C. Even a moderate heat treatment to approximately 100° C will increase the value of K of a specimen that previously had come to equilibrium at 25° C. For example, specimen MB91 exhibited a decrease in K from 897 to 808 in 6 months but when reheated to 85° C for 15 minutes and maintained at 25° C for 6 hours, the value of K was 873.

TABLE 3. *Changes in K and Q of some specimens, at 25° C and 1 mc/s, after 6 months*

Specimen designation	K			Q		
	After 1 day	After 6 months	Change	After 1 day	After 6 months	Change
			Percent			*Percent*
MB48	34	34	0	450	450	0
MB59	79	76	−4	118	191	62
MB70	267	229	−14	156	316	103
MB79	343	322	−6	295	590	100
MB85	536	499	−7	235	455	94
MB91	897	808	−10	450	630	40
MB95	1,183	1,084	−8	450	670	49
BT	1,650	[a] 1,530	−7	74	[a] 107	45
BM3	38	37	−3	330	495	50
BM4	125	120	−4	79	100	27
BM6	256	228	−11	405	730	80
BM7	706	558	−22	220	470	114
BM8	1,040	848	−18	480	900	57
BM9	1,270	1,050	−17	440	790	80
BM95	1,770	1,550	−12	350	565	61
2B5M3	26	26	0	1,450	1,600	10
2B5M5	46	46	0	235	310	32
2B5M6	129	116	−10	250	630	152
2B5M7	185	157	−15	250	460	84
2B5M9	231	204	−12	146	253	73
2B5M95	260	225	−13	162	195	20

[a] After 2 years.

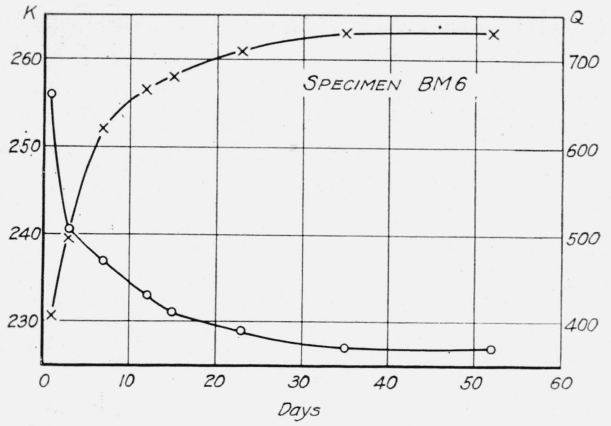

FIGURE 3. *Changes in values of K and Q with time after the maturing heat treatment.*

○=K; X=Q.

In determining the stability of barium-magnesium titanates, some specimens were included from another investigation [1] on dielectrics with compositions in the system BaO-SrO-TiO_2, table 4. These data show that significant changes, mostly increases, in Q-values occurred with most of the specimens. The significant changes in K were decreases that occurred when the specimens contained more than about 30 percent of BaO and less than approximately 55 percent of TiO_2. In this group of specimens, the changes in K and Q were a maximum for specimens with about 40 percent of BaO.

Although decreases in Q-values may be attributed to moisture adsorption, it is very improbable that moisture also caused the values of Q to increase. Possibly slow changes in the crystalline structure, such as inversion or variation in the amount of solid solution, are associated with the instability of the specimens.

Although the dielectric constant and power losses of some specimens are affected by the thermal history, the respective values of K and Q become constant for given specimens when maintained at a constant temperature for several months. Thus, the thermal history of some titanates is one out of a number of factors that contribute to variations in the dielectric constant and power losses.

Changes in the temperature of the specimens affect the dielectric constant. The data in table 5, for specimens having compositions in the system BaO-MgO-TiO_2, were obtained by

TABLE 4. *Changes in K and Q of some specimens, with compositions in the system* BaO-SrO-TiO_2, *after storage for more than 6 months*

[Measured at 1 mc/s and 25° C]

Specimen designation	Composition weight			Storage period	K			Q		
	BaO	SrO	TiO_2		After 1 day	After storage	Change	After 1 day	After storage	Change
				Months			%			%
S2T3		46.3	53.7	19	198	198	0	2,600	5,800	+55
BS22	13.2	33.1	53.7	20	151	151	0	650	700	+7
BS23	18.1	28.2	53.7	20	150	150	0	680	590	−13
BS25	23.7	22.6	53.7	19	147	146	[a]−0.6	1,450	1,750	+21
BS26	28.0	18.3	53.7	17	133	132	[a]−.7	540	560	+4
BS271	32.8	13.5	53.7	23	153	150	−2	850	1,080	+26
BS279	36.7	9.6	53.7	23	203	191	−6	475	700	+47
BS28	40.5	5.8	53.7	23	255	234	−8	172	266	+55
BS29	43.5	2.8	53.7	23	214	205	−4	102	135	+32
BT2.2	46.3		53.7	23	95	93	−2	75	85	+13
ST		56.4	43.6	7	258	256	[a]−0.8	9,000	9,000	0
B2S1	6.2	50.2	43.6	20	255	257	[a]+.8	1,300	287	−78
B2S2	13.2	43.2	43.6	20	283	286	[a]+.9	860	770	−10
B2S3	18.1	38.3	43.6	19	309	309	0	1,900	1,360	−28
B2S4	23.7	32.7	43.6	19	374	372	[a]−0.5	1,030	900	−13
B2S5	28.1	28.2	43.6	18	451	447	−1	780	1,100	+41
B2S6	33.7	22.6	43.6	18	660	622	−6	520	700	+26
B2S72	40.5	15.7	43.8	18	950	845	−11	187	315	+68
B2S83	46.6	9.6	43.9	18	1,070	985	−8	100	145	+45
B2S9	50.5	5.6	43.9	25	895	832	−7	66	80	+18
B2S95	53.3	2.8	43.9	25	820	787	−4	52	60	+15
B2T3	56.1		43.9	29	900	844	−6	38	43	+13
SB9	6.2	50.8	43.0	7	300	295	−2	4,700	4,800	[a]+2
SB50	32.8	28.2	39.0	7	872	862	−1	7,000	7,200	[a]+3

[a] Changes are within experimental error in determinations.

measuring the dielectric constant at 1 mc/s and at 10-deg. intervals from −60° to +85° C. Because the temperature at each interval was maintained constant for 15 minutes only before measurements were made, equilibrium values of K were not found for specimens with high content of BaO. For stable specimens, the average values of temperature coefficient of K, last column of table 5, are considered to be not better than ±10 ppm. or 5 percent, whichever is greater. Approximately half of these values are within the range +120 to +500 ppm. Where no values are given, computations of the coefficient of K were not made, because large irregularities appear in the curves for values of K plotted against temperature. In order to illustrate the variation of K resulting from changes in temperature and composition, figures 4, 5, and 6 were constructed for the temperatures −60°, 0°, and 60° C, respectively. These diagrams contain isodielectric-constant lines derived from the data in table 5.

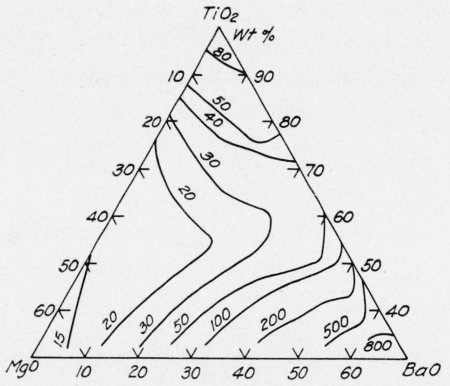

FIGURE 4. *Constant K with varying composition at 1 mc/s at −60° C.*

The most frequently observed value for the dielectric constant of $MgTiO_3$ was 17 (table 1), although values from 15 to 18 were found. Other investigators have reported values of 17 [6] and 14 [7]. Along the join $MgTiO_3$-$BaTiO_3$, the values of K change gradually from 17 for Mg-

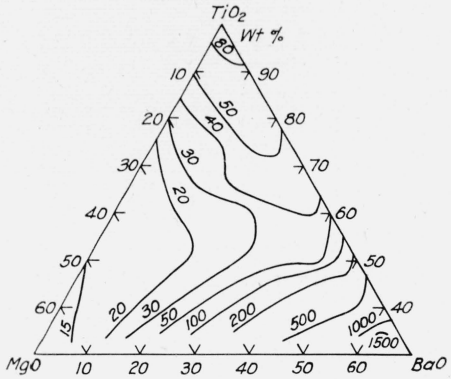

FIGURE 5. *Constant K with varying composition at 1 mc/s at 0° C.*

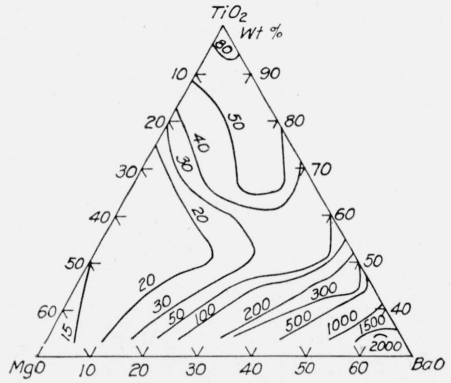

FIGURE 6. *Constant K with varying composition at 1 mc/s at 60° C.*

TiO_3 to about 1,400 for $BaTiO_3$. The specimens with compositions in this join exhibit no peak values of K within the temperature range $-60°$ to $+85°$ C in contrast to the peaks observed previously for specimens with compositions in the join $SrTiO_3$-$BaTiO_3$ [1].

The effects of variation in frequency and composition on the ranges in values of Q, measured at 25° C, are illustrated in figures 7, 8, and 9 for frequencies of 50, 1,000, and 20,000 kc/s, respectively. The upper central portions of these diagrams show that, as the frequency is raised, there is a considerable enlargement in the area for compositions with very low ranges in values of Q. Changes in frequency had the least effect upon the ranges in values of Q for specimens with relatively high content of MgO and low content of BaO and of TiO_2, as shown in the lower left regions of figures 7, 8, and 9.

With increasing frequency, 50 kc/s to 3,000

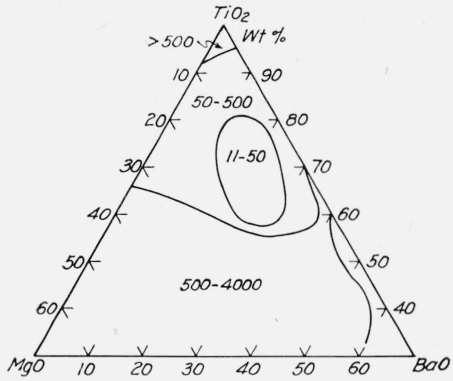

FIGURE 7. *Ranges in Q-values with varying composition at 25° C at 50 kc/s.*

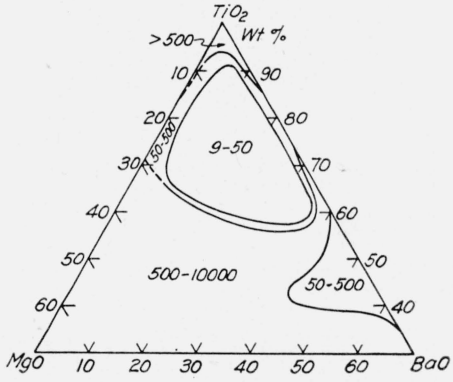

FIGURE 8. *Ranges in Q-values with varying composition at 25° C at 1,000 kc/s.*

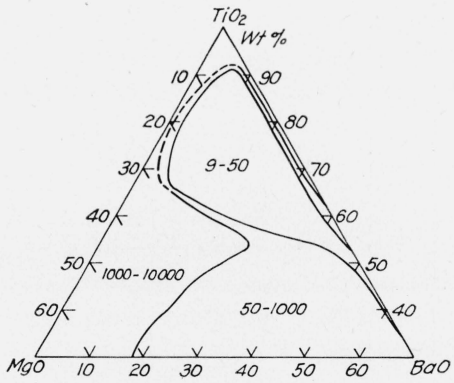

FIGURE 9. *Ranges in Q-values with varying composition at 25° C at 20,000 kc/s.*

mc/s, the low values of K (16 to 78) tend to decrease when the content of TiO_2 ranges from 60 to 90 percent. For specimens having less than 60 percent of TiO_2, the dielectric constant tends to remain constant at all frequencies used.

TABLE 5.—*Dielectric constant, at 1 mc/s, from −60° to 85° C, and average temperature coefficient of dielectric constant*

Specimen designation	Values of K at (°C)—																Average temperature coefficient of K
	−60	−50	−40	−30	−20	−10	0	+10	20	30	40	50	60	70	80	85	
																	ppm
MT_5	48.8	48.4	48.0	47.7	47.4	47.1	46.9	46.7	46.5	46.3	46.1	46.0	45.8	45.7	45.5	45.5	−500
18BM6	76.6	76.9	77.1	77.3	77.4	77.5	77.7	77.7	77.8	77.8	77.8	77.8	77.8	77.8	77.9	77.9	+120
M_2T_3	15.35	15.38	15.41	15.43	15.47	15.48	15.53	15.54	15.59	15.63	15.66	15.68	15.70	15.73	15.75	15.77	180
6BM2	21.45	21.7	21.95	22.2	22.45	22.65	22.9	23.15	23.35	23.6	23.7	23.8	23.9	24.0	24.1	24.15	810
6BM5	35.8	37.0	38.2	39.3	40.4	41.3	42.3	43.2	44 0	44.9	45.7	46.3	47.0	47.7	48.4	48.7	2,050
6BM8	49.5	51.2	53.5	55.2	57.6	59.8	62.0	63.5	65.0	67.0	68.4	70.2	72.4	74.7	77.0	78.3	3,100
6BM9	47.9	48.3	48.8	49.4	49.9	50.4	50.9	51.4	51.7	52.1	52.3	52.6	52.9	53.1	53.3	53.4	740
MT	15.90	15.93	15.96	15.99	16.02	16.05	16.09	16.12	16.16	16.20	16.22	16.23	16.25	16.27	16.28	16.29	150
4BM2	22.8	23.3	23.9	24.5	25.0	25.5	25.9	26.2	26.6	26.9	27.2	27.6	27.9	28.2	28.5	28.7	1,560
4BM4	28.6	29.6	30.8	31.9	33.0	34.1	35.3	36 3	37.4	38.6	39.5	40.3	41.0	41.7	42.3	42.6	2,650
4BM6	35.9	37.2	38.6	40.2	41.8	43.3	45.0	46.7	48.5	50.1	51.4	52.8	54.0	55.2	56.0	56.4	3,000
4BM7	38.3	39.8	41.5	43.0	44.7	46.4	48.1	49.8	51.5	53.2	55.0	56.5	58.0	59.3	60.5	61.3	3,170
4BM9	38.7	39.4	40.0	40.6	41.3	41.9	42.6	43.2	43.7	44.3	44.8	45.2	45.6	46.0	46.3	46.5	1,240
MB1	20.5	20.8	21.0	21.2	21.3	21.5	21.7	21.9	22.1	22.4	22.6	22.9	23.2	23.5	23.7	23.8	1,030
MB19	18.5	18.5	18.6	18.6	18.6	18.7	18.7	18.8	18.8	18.9	19.0	19.0	19.1	19.1	19.2	19.2	260
MB30	20.3	20.4	20.4	20.5	20.5	20.5	20.5	20.6	20.6	20.6	20.7	20.7	20.8	20.9	21.0	21.1	270
MB37	20.0	20.0	20.1	20.1	20.1	20.1	20.2	20.2	20.2	20.3	20.3	20.3	20.4	20.4	20.4	20.4	140
MB48	32.1	32.5	32.9	33.2	33.5	33.9	34.1	34.3	34.3	34.3	34.5	34.8	35.0	35.2	35.6	35.7	----------
MB59	65	67	69.5	72	74	76.5	79	80.5	81	82.5	84.5	87	91	94.5	100	104	----------
MB70	182	193	205	217	230	243	256	253	252	256	263	275	290	310	337	365	----------
MB79	255	275	295	320	340	360	390	400	410	425	440	475	500	550	640	705	----------
MB85	350	370	400	430	470	510	580	600	610	630	670	730	820	950	1,160	1,360	----------
MB91	530	570	630	680	750	820	910	900	890	900	930	1,020	1,150	1,350	1,660	1,900	----------
MB95	680	750	850	950	1,100	1,250	1,300	1,250	1,200	1,200	1,200	1,370	1,700	2,250	3,200	3,900	----------
M_4T_3	16.3	16.3	16.3	16.4	16.4	16.4	16.4	16.5	16.5	16.5	16.5	16.5	16.6	16.6	16.7	16.7	170
3BM2	17.0	17.1	17.2	17.2	17.3	17.3	17.4	17.5	17.5	17.6	17.6	17.7	17.7	17.7	17.8	17.8	320
3BM5	22.9	23.2	23.4	23.7	24.0	24.5	24.8	25.1	25.5	25.8	26.2	26.6	27.0	27.3	27.8	28.0	1,400
3BM6	26.0	26.5	27.0	27.5	28.1	28.6	29.1	29.7	30.3	31.0	31.7	32.3	33.0	33.6	34.1	34.4	1,940
3BM7	30.3	31.7	33.1	34.0	34.7	35.6	36.6	37.5	38.3	39.1	40.2	41.4	42.5	43.7	44.6	45.2	2,700
3BM8	36.0	36.5	37.7	39.8	41.5	42.8	44.0	44.9	45.9	46.9	47.9	48.8	49.6	50.3	51.0	51.4	2,400
3BM9	34.9	35.4	35.8	36.2	36.6	36.8	37.0	37.1	37.2	37.3	37.5	37.7	37.8	38.0	38.1	38.2	480
M_2T	13.75	13.8	13.8	13.8	13.85	13.9	13.9	13.9	13.95	14.0	14.0	14.0	14.1	14.1	14.1	14.1	190
2BM1	15.9	15.9	16.0	16.0	16.1	16.1	16.2	16.2	16.2	16.3	16.3	16.4	16.4	16.5	16.6	16.6	300
2BM2	16.6	16.7	16.7	16.8	16.8	16.9	16.9	17.0	17.0	17.0	17.1	17.1	17.2	17.2	17.3	17.3	300
2BM4	23.0	23.1	23.2	23.3	23.4	23.5	23.6	23.7	23.7	23.8	23.9	24.0	24.0	24.1	24.2	24.3	380
2BM5	27.5	27.6	27.8	28.0	28.1	28.3	28.4	28.5	28.6	28.7	28.7	28.8	28.9	29.0	29.2	29.3	440
2BM6	30.1	30.2	30.4	30.5	30.7	30.8	31.0	31.1	31.2	31.3	31.4	31.5	31.7	31.9	32.0	32.1	440
2BM7	31.6	32.0	32.3	32.5	32.8	33.0	33.1	33.2	33.3	33.5	33.6	33.8	34.0	34.2	34.4	34.5	620
2BM8	31.1	31.3	31.5	31.7	32.0	32.1	32.2	32.3	32.4	32.5	32.7	32.8	33.0	33.2	33.3	33.4	490
2BM87	34.6	34.9	35.1	35.4	35.6	35.7	35.8	35.9	36.1	36.3	36.5	36.7	36.9	37.1	37.3	37.5	550
2BM9	87	90	93	96	99	102	105	107.5	110	111.5	113.5	115.5	117.5	120	122	123	2,300

M_5T_2	14.4	14.4	14.4	14.45	14.5	14.5	14.5	14.55	14.6	14.6	14.6	14.6	14.65	14.7	14.7	14.7	160
$2B_5M_1$	15.8	15.9	15.9	16.0	16.0	16.0	16.0	16.1	16.1	16.1	16.1	16.1	16.2	16.2	16.2	16.3	220
$2B_5M_2$	17.3	17.3	17.4	17.4	17.5	17.5	17.5	17.5	17.6	17.6	17.6	17.6	17.7	17.7	17.8	17.8	200
$2B_5M_3$	24.2	24.4	24.6	24.9	25.0	25.1	25.2	25.3	25.4	25.6	25.8	26.1	26.4	26.6	26.9	27.1	----------
$2B_5M_5$	38.9	39.8	41.0	42.0	43.2	44.2	45.2	45.6	45.9	46.8	47.3	48.2	49.4	50.9	52.7	53.7	----------
$2B_5M_6$	89	95	101	106	112	118	122	125	126	130	135	143	153	167	188	200	----------
$2B_5M_7$	132	141	151	158	167	173	178	182	183	188	195	245	213	227	243	255	----------
$2B_5M_9$	172	181	190	197	205	208	210	210	211	217	225	232	240	250	260	266	----------
$2B_5M_{95}$	182	190	200	208	218	224	229	231	233	240	250	260	270	284	300	310	----------
M_4T	12.35	12.35	12.4	12.4	12.4	12.4	12.4	12.4	12.45	12.5	12.5	12.5	12.5	12.55	12.55	12.6	140
BM1	15.4	15.4	15.4	15.4	15.45	15.45	15.5	15.5	15.5	15.5	15.6	15 6	15.6	15.65	15.7	15.7	130
MB2	20.4	20.5	20.7	20.9	21.1	21.1	21.2	21.2	21.3	21.5	21.7	22.0	22.3	22.4	22.5	22.5	680
BM3	32.7	33.4	34.0	34.5	35.0	35.5	36.0	36.1	36.4	37.1	38.1	39.3	40.6	42.2	44.1	44.9	----------
BM4	79	82	86	92	98	106	114	115	115	120	126	133	144	158	179	185	----------
BM6	160	170	185	195	210	225	240	240	245	257	270	287	305	330	370	402	----------
BM7	370	400	440	480	520	560	600	630	630	650	690	760	830	1,010	1,260	1,350	----------
BM8	550	590	650	700	760	820	860	880	900	950	1,030	1,130	1,300	1,650	2,100	2,240	----------
BM9	630	690	760	830	920	1,000	1,080	1,100	1,100	1,180	1,270	1,400	1,640	2,250	2,760	2,790	----------
BM95	960	1,040	1,150	1,250	1,380	1,530	1,630	1,640	1,640	1,720	1,850	2,100	2,600	3,350	3,440	3,350	----------

Increasing frequency causes more irregular variations in Q-values than in those of K. For example, specimens with compositions in the system MgO-TiO_2 tend to exhibit higher values of Q at frequencies of 1 and 20 mc/s than at those of 50 kc/s and 3,000 mc/s. This trend is shown also by the barium titanates with a content of TiO_2 from 67 to 90 percent. The opposite tendency, lower values of Q at intermediate frequencies than at the extremes of frequency, is exhibited by specimens having compositions on some of the joins (60 to 67 percent of TiO_2) between magnesium titanates and barium titanates (specimens 4BM2, 4BM4, 3BM5, and 3BM6 in table 1).

The percentage of linear thermal expansion was fairly high, except the lower value of M_2T_3, despite a wide variation in composition (table 6). These dielectrics would be cracked by local heating to high temperatures. Consequently, preheating at a slow rate would be necessary in order to solder connections to the metal-coated dielectrics.

TABLE 6. *Linear thermal expansion*

Specimen designation	Temperature range from 25° C to —						
	100° C	200° C	300° C	400° C	500° C	600° C	700° C
	Percent	*Percent*	*Percent*	*Percent*	*Percent*	*Percent*	*Percent*
MT_5	0.05	0.13	0.21	0.31	0.40	0.50	0.60
6BM5	.06	.15	.26	.36	.46	.57	.67
MT	.06	.15	.26	.36	.46	.58	.70
MB37	.06	.15	.26	.36	.48	.57	.71
M_2T	.06	.15	.25	.34	.44	.55	.67
M_2T_3	.04	.10	.19	.26	.33	.39	.46

IV. Summary

Dielectrics having compositions indicated by points in the system $BaTiO_3$-$4MgO$:TiO_2-TiO_2 can be prepared from mixtures of titanium dioxide with barium and magnesium carbonates. Mature specimens, less than 0.1 percent of absorption, result from dry-pressing these calcined mixtures and heating the disks thus formed to various temperatures within the range 1,250° to 1,425° C.

The dielectric constant, K, of matured specimens varies from 12 (high content of MgO) to several hundred (high content of BaO). Most of the specimens have positive temperature coefficients of K. The Q-values range from 8 to 10,000 (high content of either MgO or TiO_2). The values of K and Q are affected by the thermal history of specimens that have a content of BaO greater than 30 percent and a content of TiO_2 less than 50 percent. The dielectric constant decreases and Q-values increase for several weeks after these specimens receive the final heat treatment. Although reheating causes a reversal of these changes in K and Q, a decrease of K and an increase of Q again occur with time. After remaining at a constant temperature for a few months, these specimens have constant values of K and Q.

Relatively high values of linear thermal expansion were obtained with specimens that varied widely in composition.

V. References

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WASHINGTON, February 16, 1948.