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NAVAL UNDERWATER SYSTEMS CENTER
NEW LONDON LABORATORY
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TECHNICAL MEMORANDUM

DIELECTRIC PROPERTIES OF PIEZOELECTRIC POLYVINYLIDENE FLUORIDE (PVDF)

Date: April 16, 1984

Prepared by: Mark B. Moffett
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14. ABSTRACT
Measurements were made of the free dielectric constant and the dielectric loss tangent of piezoelectric polyvinylidene. fluoride (PVDF) at frequencies between 1 kHz and 5 MHz and temperatures between -28??C and +51??C. Samples of nonvoided and voided PVDF were tested. The dielectric constant increases with increasing temperature and/or decreasing frequency, ranging from about 3 to 11 f~r the voided material and from about 4 to 14 for the nonvoided material. The dielectric loss tangent (dissipation factor) ranged from about 0.02 at low frequencies and high temperatures to about 0.3 at high frequencies and high temperatures. The voided material was not as lossy as the nonvoided material with maximum values - 0.2 for the voided material and 0.3 for the nonvoided material. A slight temperature aging effect was observed; an increase of a few percent in dielectric constant resulted from cycling the samples to 51??C.

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ABSTRACT

Measurements were made of the free dielectric constant and the dielectric loss tangent of piezoelectric polyvinylidene fluoride (PVDF) at frequencies between 1 kHz and 5 MHz and temperatures between -28°C and +51°C. Samples of nonvoided and voided PVDF were tested. The dielectric constant increases with increasing temperature and/or decreasing frequency, ranging from about 3 to 11 for the voided material and from about 4 to 14 for the nonvoided material. The dielectric loss tangent (dissipation factor) ranged from about 0.02 at low frequencies and high temperatures to about 0.3 at high frequencies and high temperatures. The voided material was not as lossy as the nonvoided material, with maximum values ~ 0.2 for the voided material and 0.3 for the nonvoided material. A slight temperature aging effect was observed; an increase of a few percent in dielectric constant resulted from cycling the samples to 51°C.

ADMINISTRATIVE INFORMATION

This memorandum was prepared under Job Order No. A61404, "Broadband Hydrophone for Transient Measurements," Principal Investigator: Dr. M. B. Moffett, Code 3321, Associate Investigator: J. M. Powers, Code 3234, Sponsoring Activity: NAVMAT 05B (CAPT Z. L. Newcomb), NUSC IED Program, Program Manager: Dr. K. M. Lima, Code 101. The authors are located at the Naval Underwater Systems Center, New London, CT 06320.

I. INTRODUCTION

Piezoelectric polyvinylidene fluoride (PVDF) is a useful hydrophone-element material because of its shock resistance, low density, and low mechanical impedance.[1,2] However, the capacitance is about 100 times less than that of piezoelectric ceramics, so that capacitive coupling loss is more likely to be a problem with PVDF hydrophone designs than with conventional lead zirconate-titanate (PZT) hydrophones. Furthermore, PVDF is a much lossier material than PZT,[3-5] a fact which appears to preclude its use in underwater projectors.[6] The dielectric constant and dissipation factor of PVDF have been measured at room temperature by Pennwalt workers[7] to about 400 kHz. Callerame, Tancrell and Wilson,[3] interested in medical ultrasonic applications, extended the frequency range to 20 MHz. Further extension to 100 MHz was provided by Leung and Yung[4] and by Chen.[5] All of these measurements were made on nonvoided material at room temperature. Because of the importance of the temperature dependence, we decided to measure the dielectric constant and dissipation factor as a function of temperature as well as frequency. The advent of thick-film, voided PVDF[8] made it desirable to perform the same measurements on the newer material as well.

II. EXPERIMENT

Two samples of PVDF were tested: 1) a nonvoided 2.5 cm x 2.5 cm x 0.11 mm, nickle-aluminum electroded Kynar® element manufactured by Pennwalt Corporation,[7] and 2) a voided 6.3 cm x 6.3 cm x 0.64 mm element, serial number AC 47/3, manufactured by Thorn EMI, Limited.[8] The voided element had electroplated copper electrodes. The sample under test was held by a specially-made alligator clip making separate electrical contact with each electrode. A 3-ft length of RG-58C/U cable passing through a cloth insulator served as the electrical feedthrough into the temperature-controlled environment. The latter was provided by a Tenney Junior oven/refrigerator unit. All impedance measurements were made with a Hewlett-Packard 4192A low-frequency impedance analyzer with a 16095A probe fixture operated in a single-ended mode.

The impedance of the connecting cable was accounted for after measurement of the impedances, Z_{sc} and Z_{oc}, with the alligator clip contacts short-circuited and open-circuited, respectively. If the cable is assumed to behave as a uniform transmission line, the element impedance, Z, can be calculated from the measured impedance, Z_{in} (with element and cable in place), as follows:

$$Z = Z_{oc}\,(Z_{in} - Z_{sc})/(Z_{oc} - Z_{in}) \tag{1}$$

The element capacitance is then

$$C = \mathrm{Im}(Z^{-1})/2\pi f, \tag{2}$$

where f is the frequency. The dissipation factor is

$$D = \mathrm{Re}(Z^{-1})/\mathrm{Im}(Z^{-1}) = -\mathrm{Re}(Z)/\mathrm{Im}(Z). \tag{3}$$

The (free) dielectric constant, $\varepsilon_{33}^{T}/\varepsilon_0$ was determined from the capacitance, C, as

$$\varepsilon_{33}^{T}/\varepsilon_0 = C/C_0 = Ct/\varepsilon_0 A \qquad (4)$$

where C_0 is the capacitance of an air gap dimensionally equal to the element size, ε_0 is the permittivity of free space, 8.85pF/m, t is the element thickness and A the element area. C_0 was 50.3pF for the nonvoided element and 54.9pF for the voided one.

The samples were subjected to the temperature history depicted in Figure 1. The material was considered to be "virgin" during the first 22°C to 31°C to -27°C cycle, but a rise in capacitance of several percent was noted after cycling (and holding for about 2 hrs) to 51°C and so data obtained after that point are for "aged" material.

III. RESULTS

The element capacitances are given in Table I. After division by C_0 (see Eq. 4), the dielectric constants of Table II and Figures 2-5 were obtained. It can be seen that the dielectric constant decreases monotonically with increasing frequency and with decreasing temperature. In fact, over the range of frequencies and temperatures in this study, the dielectic constant changed by a factor of more than three. The dielectric constant of the nonvoided material was generally higher than that of the voided material.

The dielectric loss tangents are given in Table III and Figures 6-9. There is a broad maximum in the loss tangent which shifts upward in frequency as the temperature is raised. At low temperatures, the voided material exhibited a sharp peak in loss factor at about 1 MHz. (It should be noted that since most of the measurements were made in a 1-2-5 frequency sequence, there is a possibility that other sharp peaks of this type existed but escaped unnoticed between the large frequency steps.) A similar "resonance" has been observed by workers at Thorn EMI.[9]

IV. DISCUSSION

The dielectric constant and dissipation factor curves of Figures 2-9 have the general shape of a dielectric relaxation process. However, each of these curves changes too slowly with frequency to be described by a single relaxation time, i.e., a multiplicity of relaxation times must be involved. Another way of seeing this result is that since the relaxation time, τ, of a conductive dielectric is given by[10] $\tau = \varepsilon/\sigma$, where σ is the electrical conductivity, and since we can express the conductivity, σ, as $2\pi f\varepsilon D$, then the relaxation time, τ, would be given by $(2\pi fD)^{-1}$. In other words, to be describable by a single (constant) relaxation time, τ, the dissipation factor, D, would have to be inversely proportional to frequency. This is clearly not the case, as can be seen in Figures 6-9.

V. CONCLUSIONS

The free dielectric constant of nonvoided PVDF ranges from about 4 to 14 over the -28°C to 51°C temperature range and 1 kHz to 5 MHz frequency range, with a value of approximately 12 at 1kHz and room temperature. (This compares reasonably well with literature values.[7,11]) The voided material has a lower dielectric constant, ranging from about 3 to 11 over the temperature and frequency ranges considered here.

The voided material is not as lossy as the nonvoided material. The maximum dissipation factor was about 0.2 for the voided material and 0.3 for the nonvoided sample. At room temperature and 1 kHz, the dissipation factor was about 0.012 for each material.

After cycling to 51°C, the dielectric constant was elevated a few percent, but the dissipation factor was essentially unchanged.

VI. ACKNOWLEDGMENTS

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T(°C) \ f(kHz)	1	2	5	10	20	50	100	200	500	1000	2000	5000	
31	616.7	613.7	609.6	605.0	591.4	575.6	566.6	546.9	509.4	468.8	420.1	350.8	
22	602.7	599.7	592.6	586.0	580.4	549.6	532.6	504.0	454.7	408.9	362.5	304.7	virgin
11	576.7	571.7	562.6	551.0	537.4	495.6	471.6	435.0	383.0	342.8	306.2	265.5	nonvoided
2	552.7	543.7	526.6	507.0	481.4	433.6	402.6	365.1	321.2	290.4	264.9	236.7	element
-8	501.7	482.7	451.6	423.0	392.4	348.6	320.6	294.1	265.3	246.8	230.2	212.5	$C_o = 50.3$
-17	418.7	395.7	362.6	337.0	313.4	283.6	265.7	249.1	230.4	219.1	208.9	196.2	pF
-27	318.7	302.7	281.6	266.0	252.4	237.6	225.7	216.1	205.4	199.2	192.5	183.9	
51	681.7	676.7	670.6	665.0	663.4	634.6	630.6	620.9	600.9	577.5	543.5	474.2	
41	656.7	653.7	647.6	643.0	644.4	617.6	604.6	594.9	569.1	537.5	493.0	412.0	aged
31	639.7	635.7	629.6	624.0	617.4	593.6	579.6	562.9	525.3	485.5	435.1	361.3	nonvoided
22	619.7	614.7	607.6	600.0	591.4	562.6	543.6	517.0	467.6	421.7	372.0	311.3	element
11	587.7	582.7	572.6	560.0	543.4	505.6	477.6	440.0	385.9	343.8	306.2	263.6	$C_o = 50.3$
2	559.7	549.7	531.6	512.0	486.4	436.6	404.6	367.1	321.2	290.4	264.0	235.3	pF
-9	504.7	486.7	454.6	425.0	394.4	349.6	321.6	294.1	264.3	245.8	229.2	211.0	
-17	421.7	398.7	363.6	338.0	313.4	283.6	264.7	248.1	229.4	218.1	207.0	195.4	
-28	317.7	300.7	279.6	265.0	251.4	235.6	224.7	215.1	204.4	197.3	190.5	182.3	
31	516.7	514.7	511.6	506.0	496.4	488.6	478.6	463.0	435.7	401.9	367.8	316.6	
22	505.7	502.7	497.6	491.0	478.4	466.6	449.6	428.0	391.9	355.6	322.3	279.6	virgin
11	481.7	477.7	470.6	463.0	444.4	424.6	402.6	375.1	338.1	305.2	280.2	248.4	voided
2	460.7	453.7	440.6	426.0	402.4	374.6	349.6	323.1	291.2	265.6	247.5	224.4	element
-8	418.7	404.7	381.6	361.0	336.4	309.6	289.6	270.1	248.3	230.0	219.5	204.2	$C_o = 54.9$
-17	359.7	342.7	319.6	301.0	282.4	261.6	248.7	236.1	221.4	207.2	202.1	191.2	pF
-27	286.7	274.7	259.6	248.0	238.4	224.6	216.7	209.2	200.5	193.3	187.6	179.7	
51	581.7	578.7	574.6	570.0	558.4	552.6	545.6	535.9	523.3	499.1	473.6	422.5	
41	553.7	550.7	546.6	541.0	531.4	525.6	518.6	508.0	489.4	461.8	430.8	374.2	aged
31	542.7	539.7	534.6	531.0	519.4	510.6	499.6	484.0	456.6	421.6	385.8	330.9	voided
22	524.7	521.7	516.6	509.0	496.4	483.6	468.6	446.0	409.8	371.4	335.6	290.6	element
11	498.7	493.7	485.6	476.0	459.4	437.6	413.6	386.1	345.1	311.2	284.1	250.9	$C_o = 54.9$
2	474.7	466.7	452.6	437.0	414.4	383.6	357.6	329.1	295.2	268.6	249.5	226.1	pF
-9	429.7	415.7	391.6	370.0	343.4	314.6	292.6	272.1	249.3	230.0	219.5	204.3	
-17	366.7	348.7	323.6	305.0	284.4	263.6	249.7	236.1	221.4	207.2	201.2	190.4	
-28	288.7	276.7	260.6	248.0	237.4	224.6	216.7	208.2	199.5	192.3	186.6	178.9	

Table I. Capacitance, C, of PVDF elements (picofarads).

T (°C) \ f (kHz)	1	2	5	10	20	50	100	200	500	1000	2000	5000	
31	12.3	12.2	12.1	12.0	11.8	11.4	11.3	10.9	10.1	9.3	8.4	7.0	
22	12.0	11.9	11.8	11.6	11.5	10.9	10.6	10.0	9.0	8.1	7.2	6.1	virgin
11	11.5	11.4	11.2	11.0	10.7	9.9	9.4	8.6	7.6	6.8	6.1	5.3	nonvoided
2	11.0	10.8	10.5	10.1	9.6	8.6	8.0	7.3	6.4	5.8	5.3	4.7	element
-8	10.0	9.6	9.0	8.4	7.8	6.9	6.4	5.8	5.3	4.9	4.6	4.2	
-17	8.3	7.9	7.2	6.7	6.2	5.6	5.3	5.0	4.6	4.4	4.2	3.9	
-27	6.3	6.0	5.6	5.3	5.0	4.7	4.5	4.3	4.1	4.0	3.8	3.7	
51	13.6	13.5	13.3	13.2	13.2	12.6	12.5	12.3	11.9	11.5	10.8	9.4	
41	13.1	13.0	12.9	12.8	12.8	12.3	12.0	11.8	11.3	10.7	9.8	8.2	aged
31	12.7	12.6	12.5	12.4	12.3	11.8	11.5	11.2	10.4	9.7	8.6	7.2	nonvoided
22	12.3	12.2	12.1	11.9	11.8	11.2	10.8	10.3	9.3	8.4	7.4	6.2	element
11	11.7	11.6	11.4	11.1	10.8	10.0	9.5	8.7	7.7	6.8	6.1	5.2	
2	11.1	10.9	10.6	10.2	9.7	8.7	8.0	7.3	6.4	5.8	5.2	4.7	
-9	10.0	9.7	9.0	8.4	7.8	6.9	6.4	5.8	5.3	4.9	4.6	4.2	
-17	8.4	7.9	7.2	6.7	6.2	5.6	5.3	4.9	4.6	4.3	4.1	3.9	
-28	6.3	6.0	5.6	5.3	5.0	4.7	4.5	4.3	4.1	3.9	3.8	3.6	
31	9.4	9.4	9.3	9.2	9.0	8.9	8.7	8.4	7.9	7.3	6.7	5.8	
22	9.2	9.2	9.1	8.9	8.7	8.5	8.2	7.8	7.1	6.5	5.9	5.1	virgin
11	8.8	8.7	8.6	8.4	8.1	7.7	7.3	6.8	6.2	5.6	5.1	4.5	voided
2	8.4	8.3	8.0	7.8	7.3	6.8	6.4	5.9	5.3	4.8	4.5	4.1	element
-8	7.6	7.4	6.9	6.6	6.1	5.6	5.3	4.9	4.5	4.2	4.0	3.7	
-17	6.6	6.2	5.8	5.5	5.1	4.8	4.5	4.3	4.0	3.8	3.7	3.5	
-27	5.2	5.0	4.7	4.5	4.3	4.1	3.9	3.8	3.7	3.5	3.4	3.3	
51	10.6	10.5	10.5	10.4	10.2	10.1	9.9	9.8	9.5	9.1	8.6	7.7	
41	10.1	10.0	10.0	9.9	9.7	9.6	9.4	9.3	8.9	8.4	7.8	6.8	aged
31	9.9	9.8	9.7	9.7	9.5	9.3	9.1	8.8	8.3	7.7	7.0	6.0	voided
22	9.6	9.5	9.4	9.3	9.0	8.8	8.5	8.1	7.5	6.8	6.1	5.3	element
11	9.1	9.0	8.8	8.7	8.4	8.0	7.5	7.0	6.3	5.7	5.2	4.6	
2	8.6	8.5	8.2	8.0	7.5	7.0	6.5	6.0	5.4	4.9	4.5	4.1	
-9	7.8	7.6	7.1	6.7	6.3	5.7	5.3	5.0	4.5	4.2	4.0	3.7	
-17	6.7	6.4	5.9	5.6	5.2	4.8	4.5	4.3	4.0	3.8	3.7	3.5	
-28	5.3	5.0	4.7	4.5	4.3	4.1	3.9	3.8	3.6	3.5	3.4	3.3	

Table II. Dielectric constant, $\varepsilon_{33}^{T}/\varepsilon_0$ of PVDF elements.

T(°C) \ f(kHz)	1	2	5	10	20	50	100	200	500	1000	2000	5000	
31	.011	.015	.019	.025	.039	.055	.075	.101	.151	.199	.248	.305	
22	.012	.019	.026	.035	.055	.079	.109	.139	.192	.229	.258	.282	virgin
11	.020	.028	.043	.059	.080	.126	.152	.185	.220	.236	.243	.241	nonvoided
2	.035	.050	.074	.098	.127	.171	.188	.206	.214	.212	.204	.193	element
−8	.074	.096	.126	.148	.168	.195	.187	.184	.174	.162	.152	.143	
−17	.117	.134	.150	.156	.160	.168	.148	.140	.128	.117	.111	.107	
−27	.118	.123	.123	.119	.116	.112	.099	.095	.086	.082	.080	.085	
51	.016	.018	.019	.023	.030	.043	.045	.058	.086	.122	.174	.274	
41	.014	.016	.019	.023	.040	.042	.055	.073	.113	.161	.225	.333	aged
31	.014	.018	.023	.028	.040	.055	.073	.099	.149	.197	.249	.312	nonvoided
22	.016	.021	.029	.039	.054	.080	.106	.138	.192	.234	.269	.301	element
11	.022	.031	.046	.062	.086	.125	.155	.190	.226	.244	.253	.261	
2	.037	.052	.076	.100	.130	.175	.192	.210	.219	.218	.211	.204	
−9	.075	.098	.128	.151	.171	.199	.191	.188	.178	.167	.158	.154	
−17	.118	.135	.157	.159	.162	.175	.151	.143	.131	.120	.114	.113	
−28	.118	.123	.123	.120	.116	.116	.099	.095	.087	.082	.080	.085	
31	.010	.015	.020	.026	.033	.049	.066	.092	.133	.168	.200	.231	
22	.012	.018	.025	.034	.046	.068	.092	.123	.164	.191	.206	.218	virgin
11	.019	.028	.042	.058	.077	.105	.130	.157	.131	.194	.194	.188	voided
2	.033	.047	.069	.090	.111	.139	.154	.168	.174	.175	.163	.151	element
−8	.066	.085	.110	.126	.142	.148	.150	.149	.141	.140	.123	.112	
−17	.097	.110	.121	.127	.136	.126	.121	.115	.107	.119	.092	.084	
−27	.092	.097	.099	.098	.097	.090	.083	.078	.074	.111	.063	.061	
51	.015	.017	.020	.022	.024	.030	.039	.051	.079	.104	.142	.206	
41	.013	.015	.018	.023	.026	.034	.046	.064	.098	.131	.173	.231	aged
31	.014	.018	.023	.028	.034	.049	.065	.090	.132	.169	.205	.245	voided
22	.015	.021	.029	.037	.048	.070	.093	.123	.166	.197	.217	.235	element
11	.022	.030	.044	.059	.077	.107	.133	.161	.190	.203	.202	.200	
2	.034	.048	.071	.092	.113	.143	.160	.176	.183	.183	.171	.163	
−9	.069	.087	.113	.130	.146	.156	.156	.156	.149	.146	.129	.120	
−17	.101	.114	.127	.133	.140	.134	.125	.121	.110	.120	.095	.091	
−28	.096	.101	.104	.101	.102	.093	.086	.082	.075	.110	.065	.065	

Table III. Dissipation factor, D, of PVDF elements.

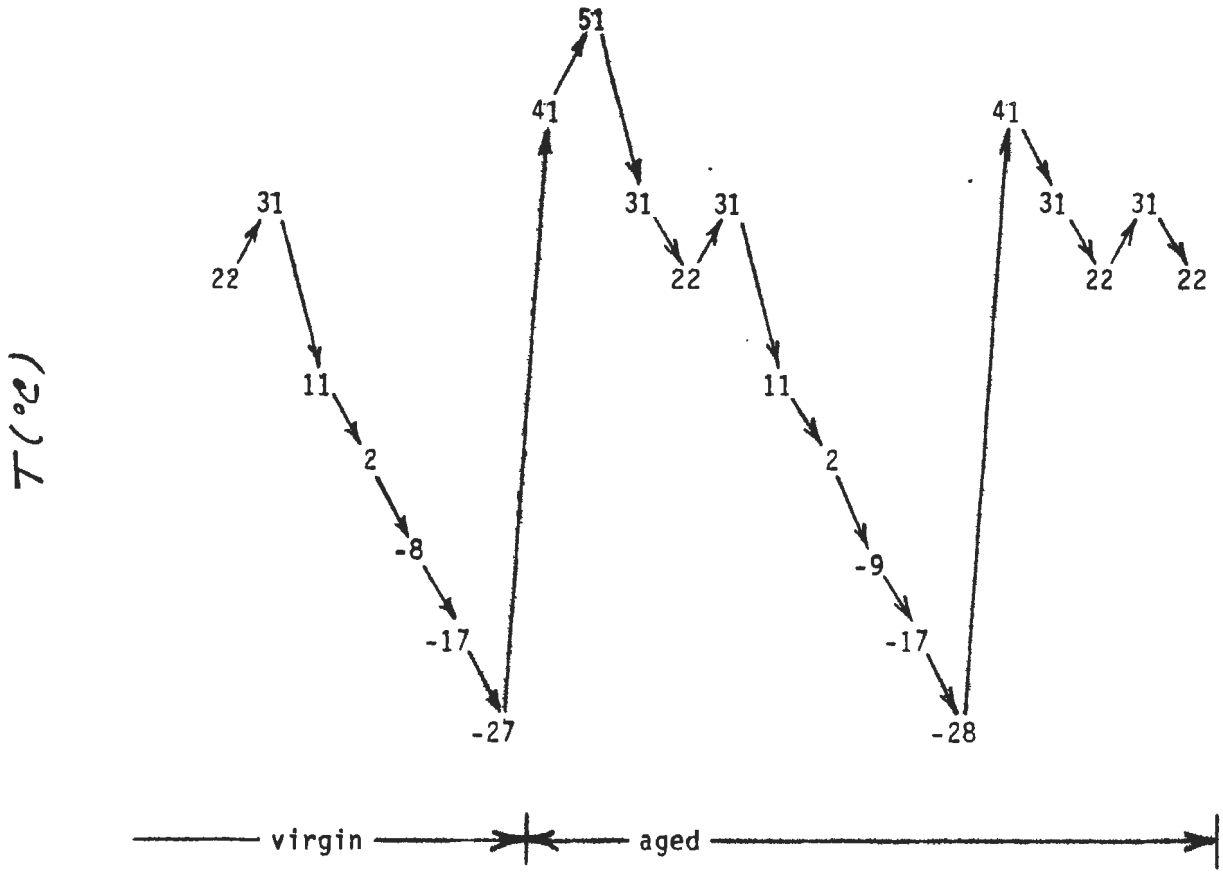

Figure 1. Temperature history of PVDF samples.

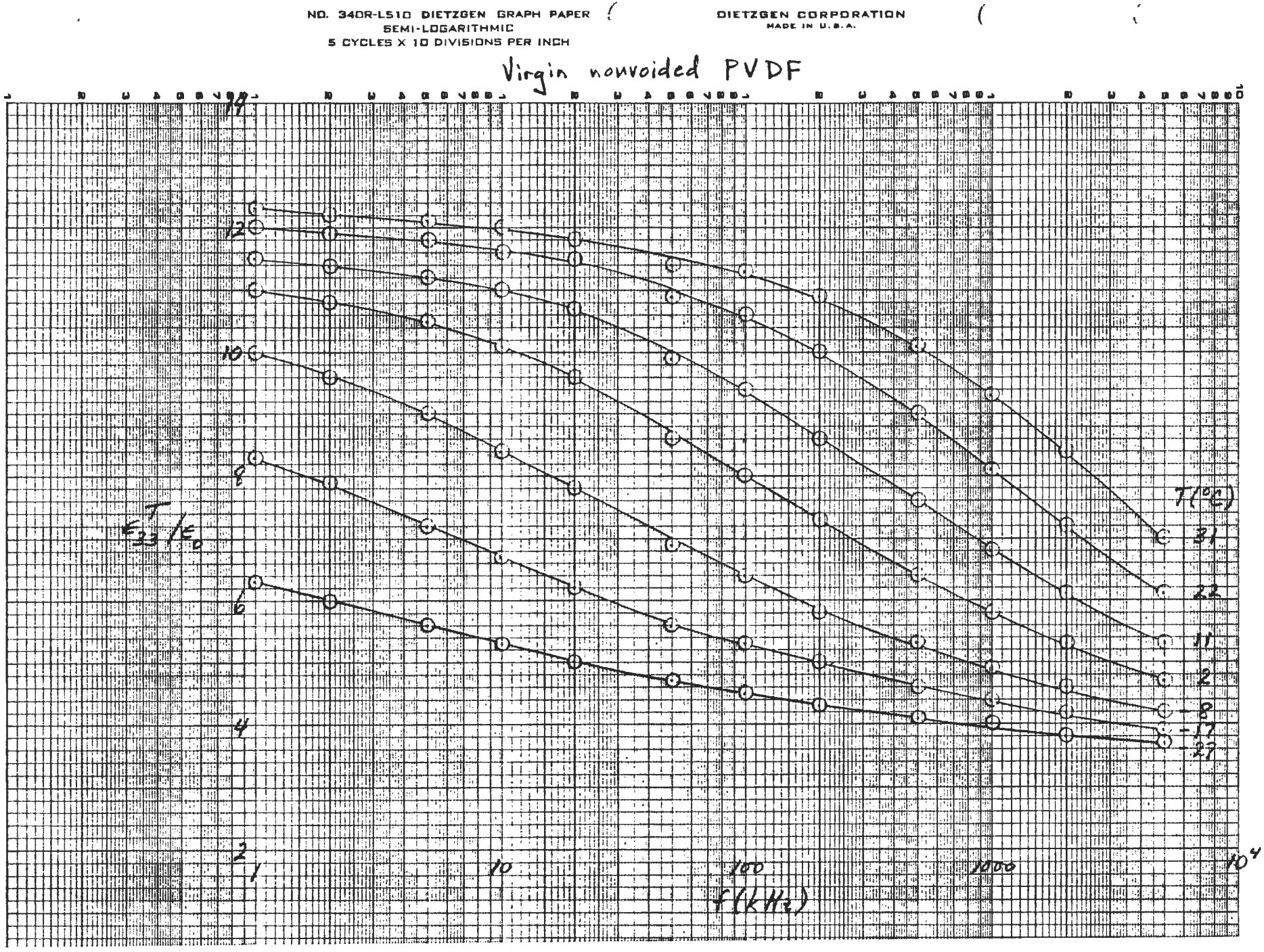

Figure 2. Dielectric constant, virgin nonvoided PVDF.

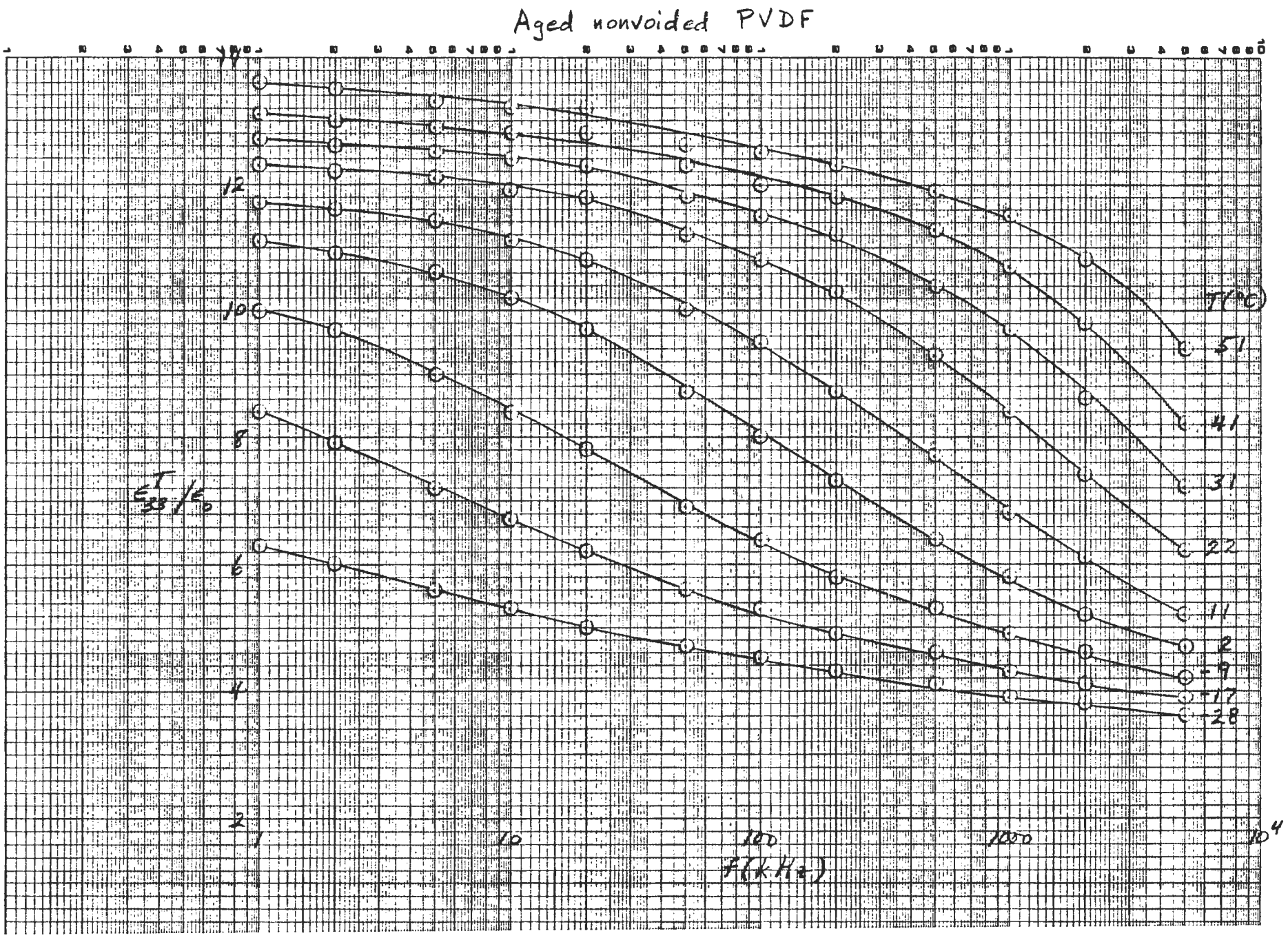

Figure 3. Dielectric constant, nonvoided PVDF after exposure to 51°C.

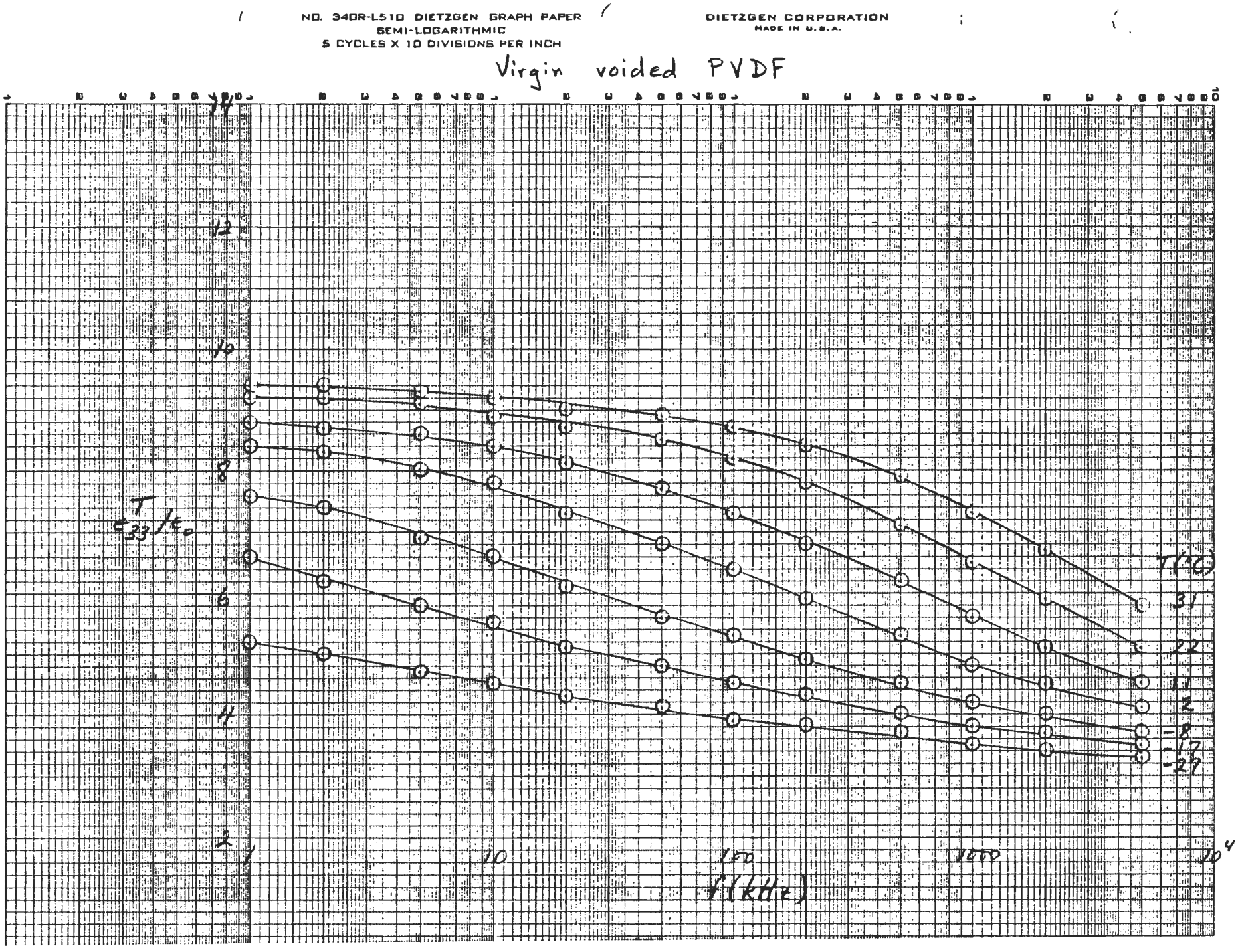

Figure 4. Dielectric constant, virgin voided PVDF.

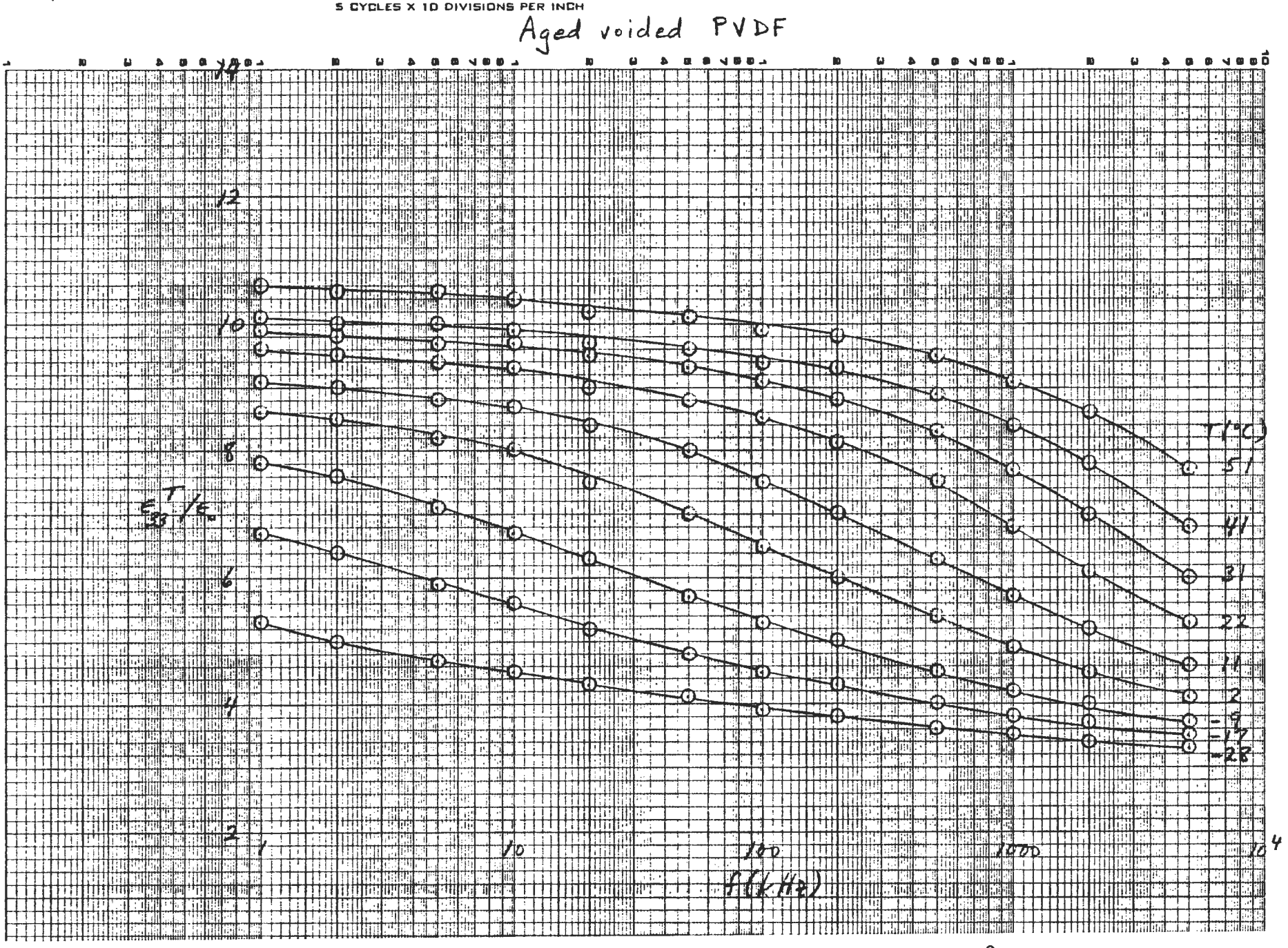

Figure 5. Dielectric constant, voided PVDF after exposure to 51°C.

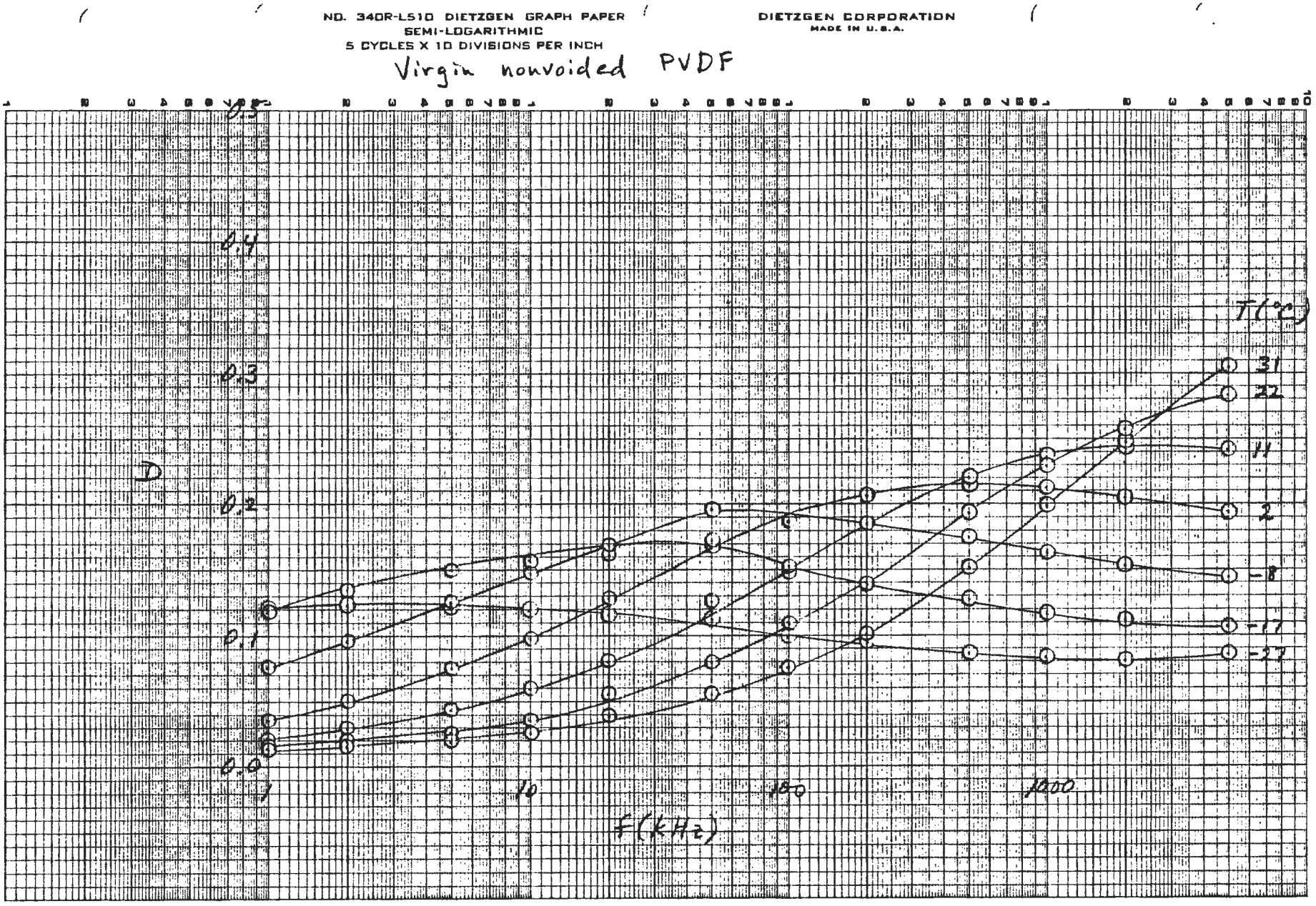

Figure 6. Dissipation factor, virgin nonvoided PVDF.

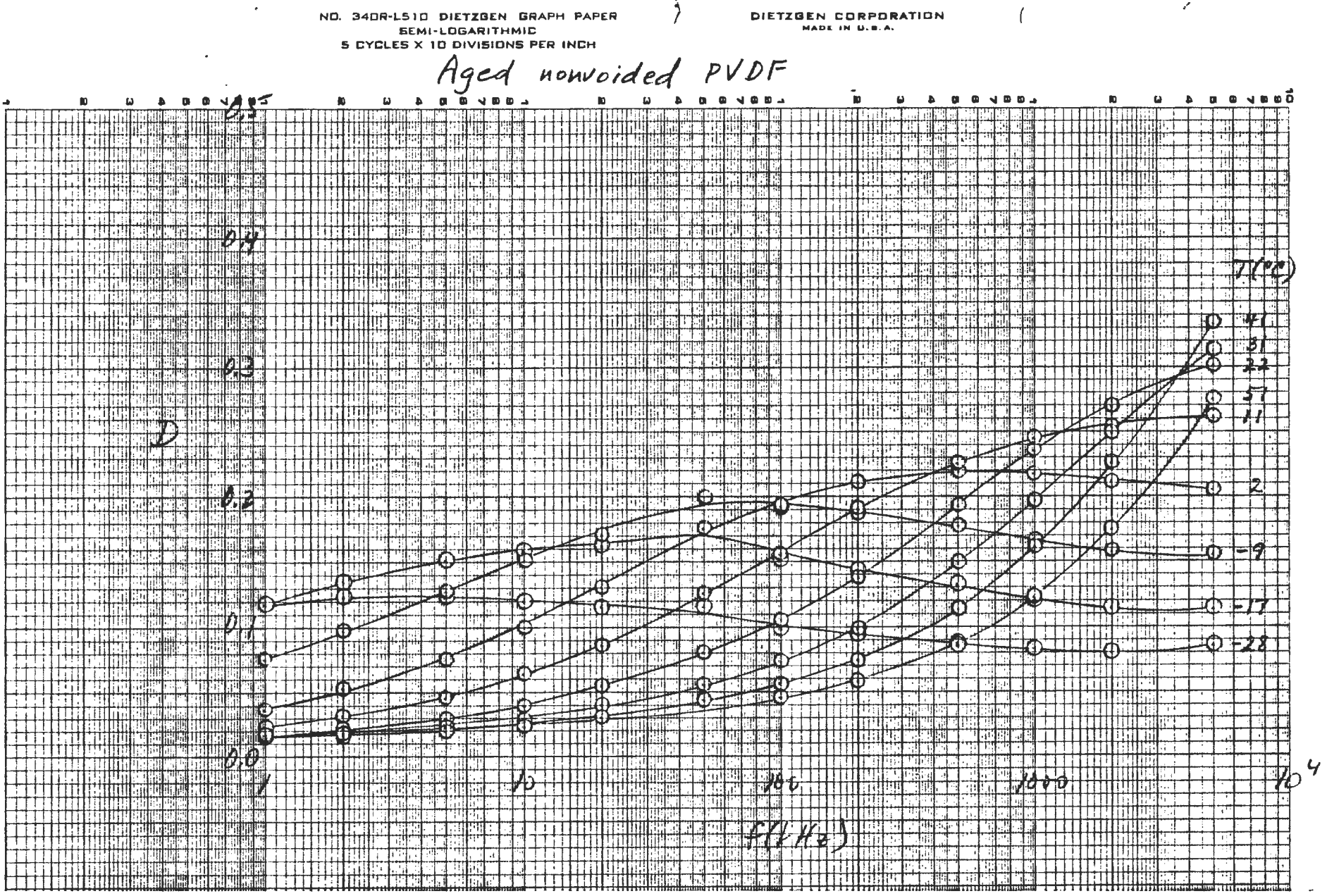

Figure 7. Dissipation factor, nonvoided PVDF after exposure to 51°C.

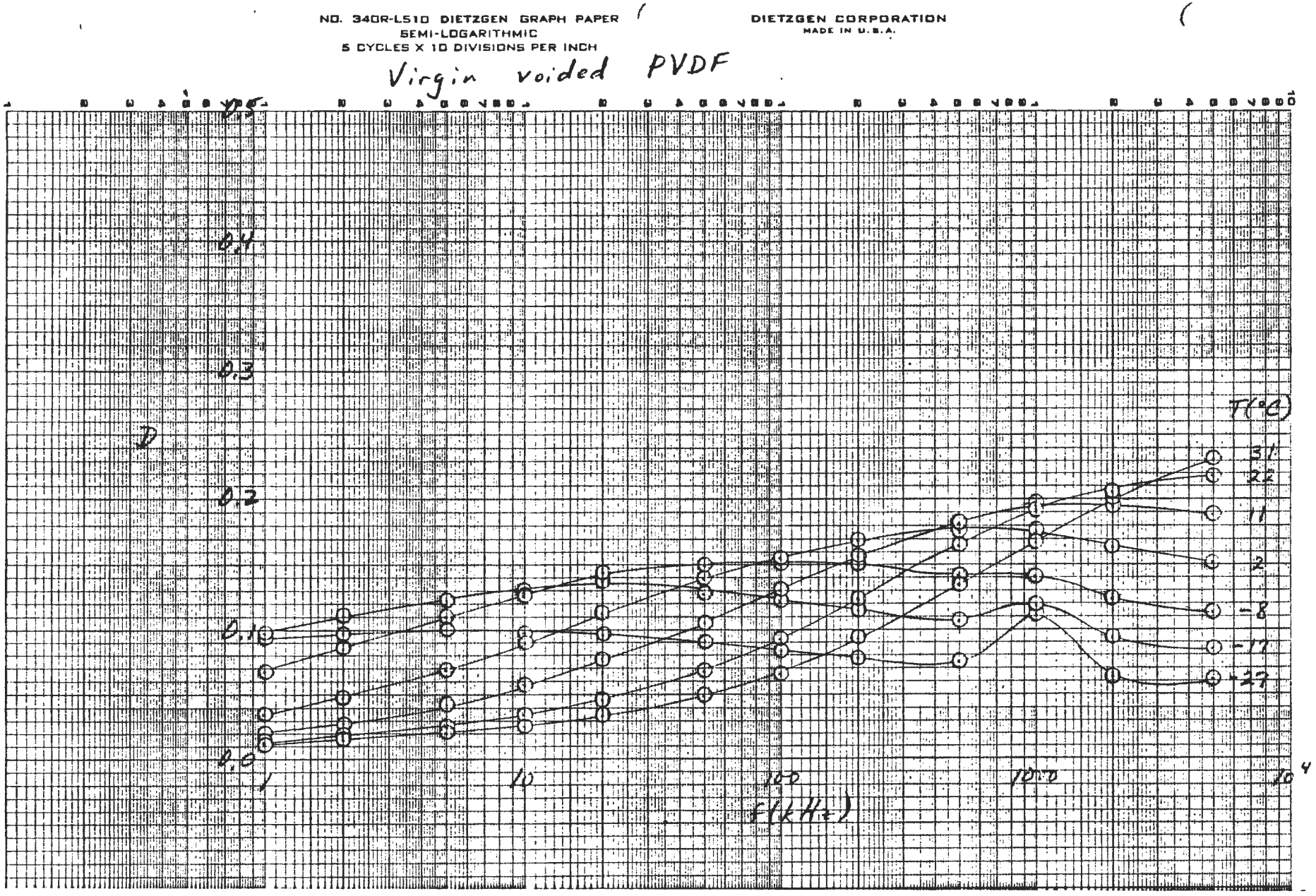

Figure 8. Dissipation factor, virgin voided PVDF.

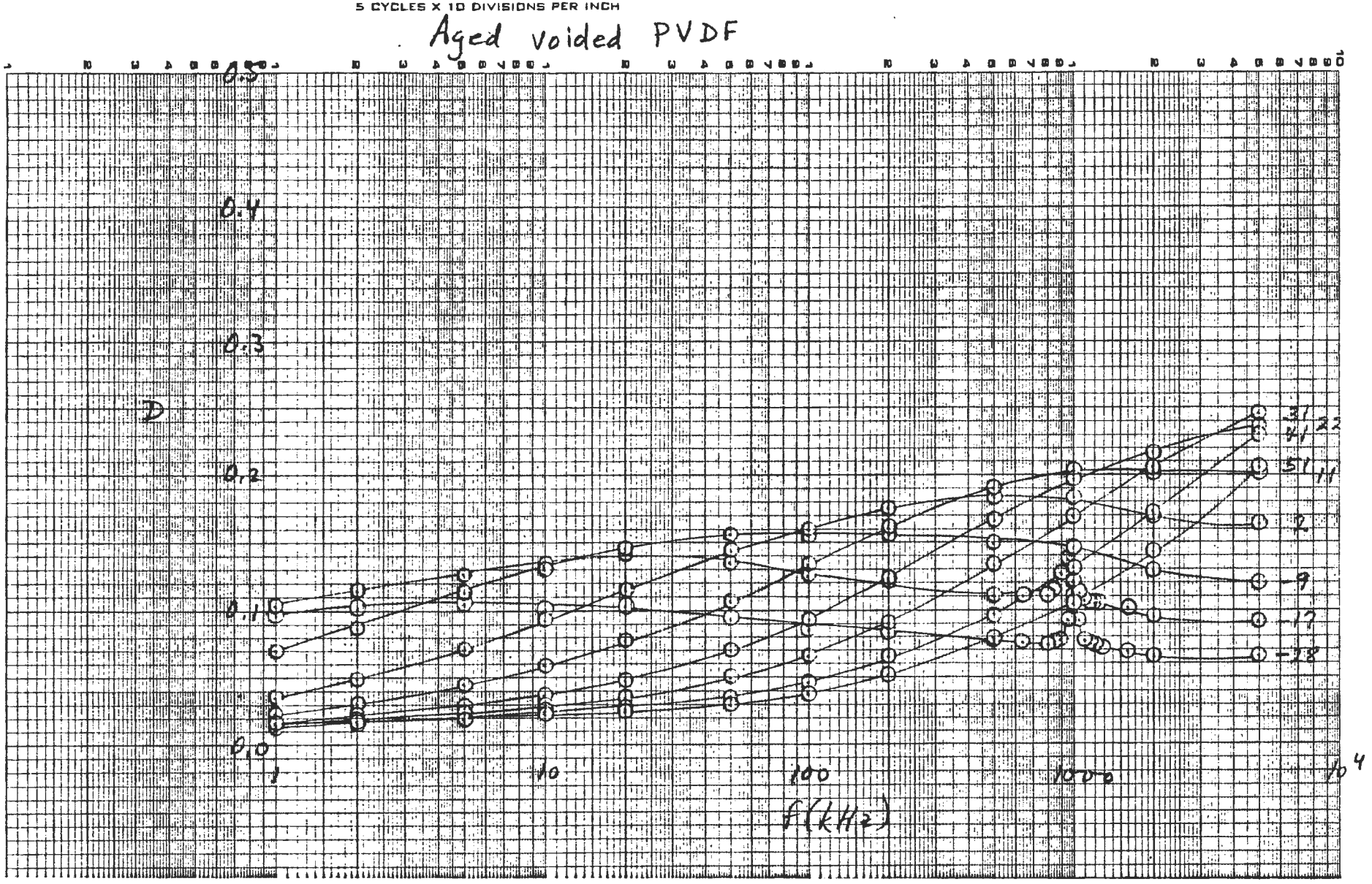

Figure 9. Dissipation factor, voided PVDF after exposure to 51°C.

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Target Physics Sensor Technology Branch
James M. Powers
Electroacoustic Transduction Branch
TM No. 841072
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