NEW CAPACITOR IMPREGNANTS

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ABSTRACT

Three major classes of fluids useful as capacitor dielectric impregnants are discussed. These include chlorinated biphenyl fluids and the recently introduced ester fluids and ester base and hydrocarbon base fluids with additives to give dielectric constant equal to trichlorobiphenyl. Some of the relative advantages and disadvantages of these classes of fluids are discussed. More commercial experience with the new fluids is needed to completely define their place in capacitor technology.

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NEW CAPACITOR IMPREGNANTS

BACKGROUND

Capacitors are indispensable in present day distribution and use of electrical energy. Their applications range from large banks of power factor correction capacitors through motor start-run capacitors and capacitors for fluorescent lamp ballasts to the many types used in electronic devices. Many types of construction are required depending on the size and use for which they are intended. This paper is concerned with those capacitors constructed of a sheet dielectric, either paper or film or a combination of these, using either foil electrodes or electrodes formed by evaporating a metallic film onto the surface of the dielectric.

A number of advantages can be achieved by impregnating capacitors of the types described above with a suitably chosen liquid. These are:

1) The dielectric strength of a liquid is greater than that of a gas permitting higher voltage rating for a given interelectrode spacing.
2) Corona discharge inception and extinction voltages for liquid impregnated systems are much higher than for unimpregnated ones.
3) The capacitance for a given electrode area and spacing can be greatly increased because liquids have much higher dielectric constants than gases.
4) Better distribution of electrical stress within the composite dielectric system can be achieved

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by proper choice of dielectric constant of the liquid.

5) Heat transfer through the capacitor roll is improved.

6) As a result of the preceding factors, capacitors which give better performance at lower cost can be produced. They use less material, occupy less space, weigh less, and last longer.

In summary, impregnation is essential for the production of capacitors with working voltages required in a wide variety of present day power transmission and use technology.

The first impregnants used were hydrocarbon oils, natural asters, such as castor oil, and waxes. All of these had relatively low dielectric constants. They were displaced to a large extent by pentachlorobiphenyl type material such as Aroclor* 1254 which had a higher dielectric constant, was more stable, easily purified, fire resistant and low in cost. Later trichlorobiphenyl fluids such as Aroclor 1242 were substituted to obtain the advantage of still higher dielectric constant in making still better capacitors at a still lower cost.

Used trichloro biphenyl type fluids for capacitor impregnation has been almost universal in the capacitor industry for many years.

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The properties of the trichlorobiphenyl type fluids (were) are ideal with one major exception. Jensen (1) in 1966 discovered that chlorinated biphenyls could be detected in natural waters and in fish and marine birds. Because of the similarity in chemical nature and properties of these materials to the insecticide DDT, they were assumed to be equally hazardous in the environment.

In this country Monsanto stopped sale of chlorinated biphenyls for all purposes except for use in capacitors and transformers where the use was considered to be non dispersive and essential to the electrical industry. The U.S. Government Interdepartmental Task Force on PCB's carried out a broad investigation of the problem (2) and stated that "Their continued use for transformers and capacitors in the near future is considered necessary because of the significantly increased risk of fire and explosion and the disruption of electrical service which would result from a ban on PCB use".

At this point it should be stated that the chlorinated biphenyl type fluids are mixtures of individual chlorinated biphenyls which vary in the number of chlorine atoms contained and in the way that the chlorine atoms are positioned on the molecule. A typical chlorinated biphenyl fluid such as Aroclor 1242 contains about 70 components. Its average chlorine content is that of trichloro biphenyl but it contains components with fewer as well as more chlorines per molecule. The term PCB is often used to describe

any and all chlorinated biphenyl products. It should be noted however that there is a large difference in properties depending on degree of chlorination.

In a further effort to minimize any possible harmful effects in the environment, Monsanto produced a new fluid, Aroclor 1016, with an average chlorine content equivalent to trichlorobiphenyl. This fluid has a concentration of more highly chlorinated materials (5 or more chlorine atoms per molecule of chlorinated biphenyl) only one-eighth that of Aroclor 1242. The rationale for this is that the more highly chlorinated material biodegrades at a lower rate than material containing less chlorine. Another fluid, MCS 1043, essentially a dichlorobiphenyl, was also developed as a possible substitute for Aroclor 1016 should government regulations require it but this was not offered commercially because of its lower fire resistance and greater cost. Measures to insure that capacitor and transformer users understood the need to minimize discharge of PCB products into the aquatic environment were also taken by American National Standards Institute Committee C-107.

It is interesting to note that the various measures taken by industry have resulted in a forty-fold decrease in the concentration of PCB type products found in North Atlantic Ocean surface waters since 1972 (3).

The efforts by industry to minimize possible environmental con-

tamination by PCB's and the Government Task Force statement that their continued use should be permitted, allowed their continued use in the United States. However, there was a need for non-PCB fluids to fill the export market to Japan where their use was banned. There was also need to have non-PCB fluids to insure the continued ability to produce efficient low-cost capacitors without PCB should additional facts be discovered which would cause the United States Government to change its attitude and ban PCB use.

For these reasons, many organizations, Monsanto among them, undertook programs to develop non-PCB capacitor impregnants. To aid such development, we must understand the requirements. Properties important to the satisfactory use of a fluid as a capacitor impregnant are listed in Table I.

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TABLE I
IMPORTANT PROPERTIES OF CAPACITOR IMPREGNATION FLUIDS

1. Dielectric Constant - 6 or above at 25°C
2. Low Dissipation Factor < 0.05 @ 100°C 60 Hz
3. High Resistivity > 500 x 10^9 Ω cm @ 100°C
4. High Dielectric Strength ∿ 40 KV per ASTM D877
5. High Corona Inception and Extinction Voltage
6. Ease of Purification
7. Ease of Impregnation
8. Compatibility with paper, film, elastomers, etc.
9. No Acid Gas Formed on Arcing
10. Wide Liquid Range
11. Fire Resistance
12. Stability in Use and Storage
13. Biodegradability
14. Low Toxicity
15. Low Absorption by Organisms
16. Available
17. Price commensurate with performance.

This list could easily be expanded by subdividing individual items. With this many constraints on the choice of fluid, it is obvious that the probability of finding promising candidates is not high.

The basic molecular structure requirements to produce a material of the desired electrical properties are: 1) The chemical bonds involved in its structure must have a minimum of ionic character so that the material can have low power factor and high resistivity; 2) To achieve high dielectric constant there must be a highly unsymmetrical distribution of charge within the molecule; 3) The bonds involved must be highly stable but not so stable that the product will not biodegrade; 4) The molecular weight and functional groups present must be such as to give the desired liquid range with minimum toxicity.

Using these concepts and the list of detailed requirements in Table I as guides, representatives of each of the likely classes of compounds were chosen and evaluated. Our work lead to the result was a decision not to try to combine all the desired properties in one molecule but to choose a high dielectric constant component which could be added to lower dielectric constant materials to give whatever dielectric constant was required.

Diaryl sulfones emerged as a tantalizing class of materials. They possess high dielectric constants and can be readily synthesized from available industrial raw materials. Unfortunately, many of

them are high-melting solids with very low solubility in suitable base fluids. Worse still, they imparted unacceptably high conductivity to fluids. Their use had been suggested long ago (4) but had never been practiced, probably because of this problem, which seemed to be inherent.

To determine if the conductivity was inherent to sulfones, a sample of diphenyl sulfone was exhaustively purified by crystallization from solvent and by zone refining. This experiment proved that sufficiently purified sulfones were satisfactory high dielectric constant components for capacitor impregnating fluids.

This fact made possible design of a series of capacitor impregnation fluids which satisfy the criteria for capacitor impregnants perhaps better than any others with the exception that only those made with polychlorinated biphenyl as the base fluid are as fire resistant as the Aroclor fluids.

NEW FLUIDS

Table II gives data on chlorinated biphenyl type fluids. The top portion of Table II gives the pertinent properties of three chlorinated diphenyl base fluids; the bottom portion gives data on standard 2 μF ballast type capacitors impregnated with these fluids. Data on Aroclor 1016 fluid are given for comparison because it is the most widely used capacitor impregnant. Figure 1 shows the capacitance and dissipation factor of an Aroclor 1016

impregnated capacitor as a function of temperature.) With the exception of possible objections on environmental grounds, Aroclor 1016 is a nearly ideal impregnant.

MCS 1043, ~~was mentioned earlier in this paper as~~ a dichlorobiphenyl type fluid, ~~which~~ could be substituted for trichlorobiphenyl type fluids such as Aroclor 1016 if environmental concern should dictate that Aroclor 1016 could not be used but a lower chlorinated material would be acceptable. It gives equally good performance in capacitors as Aroclor 1016 with one exception. It is not as fire resistant. It would also be more expensive to produce. At present it is not commercially available.

MCS 1489, having a trichlorobiphenyl base, is designed as a higher dielectric constant fluid to permit savings in the amounts of paper and foil required to build a capacitor and thus lower the cost of manufacture. As can be seen from the lower part of Table II, it gives about 12% greater capacitance for the same area and thickness of dielectric as Aroclor 1016 gives. (Figure 2 gives the capacitance and dissipation factor data for a capacitor impregnated with MCS 1489.) It has been extensively tested in our laboratories and by industrial users. Capacitors made using it are currently being field tested.

A second class of fluids of interest are esters and ester base sulfone blends. Table III gives the properties of three ester

type fluids and capacitors made from them. Two of these dioctyl phthalate (DOP) and di iso nonyl phthalate (DINP) are commercial plasticizers which have recently been used to some extent by the capacitor industry (5). The third, MCS 1475, is an ester base fluid designed to have the same dielectric constant as Aroclor 1016. The capacitor use of MCS 1475 is patented by Monsanto (6). (Figure 3 shows capacitance and dissipation factor of an MCS 1475 impregnated capacitor as a function of temperature.) Performance of these three fluids as capacitor impregnants is equivalent with the exception that the higher dielectric constant of MCS 1475 requires a smaller area/thickness ratio and permits manufacture of capacitors of the same capacitance in the same size cans as would be used with Aroclor 1016. Capacitors made with all three of these fluids have been sold in development quantities by various capacitor manufacturers

A third class of fluids is hydrocarbon base-sulfone blends. The capacitor use of these too is covered by Monsanto patent (7). Data for two such fluids are given in Table IV. (Capacitance and dissipation factor data as a function of temperature are given in Figure 4 for MCS 1588 which is typical of the group) Again these fluids have been designed to have the same dielectric constant as Aroclor 1016. Capacitors made from them give excellent results in accelerated life tests. Of the two only MCS 1588 is offered for sale at present.

DISCUSSION

Many factors affect the performance of a capacitor impregnant. In all cases, they must be properly purified. This is usually done by treatment with adsorbents such as fuller's earth, various grades of alumina, silica gel or molecular sieves. It is essential that the capacitor roll be thoroughly dried -- particularly if ester base fluids which are subject to hydrolysis are to be used. Use of proper stabilizers is necessary too. All of the fluids described above should be used with a suitable epoxide type stabilizer for A.C. capacitors. Capacitors for D.C. use require stabilizers such as anthraquinone. Quality of the solid dielectric is all important. Film or paper may have soluble, ionizable impurities which dissolve in the fluid and increase dissipation factor. Or, there may be mechanical imperfections. Uniform fabrication of test capacitors is also essential to avoid mechanical defects. The degree of electrical and thermal stress which the design and use conditions impose are also critical.

The suitability of a fluid as a capacitor impregnant must be judged on the basis of performance in capacitors. (Accelerated life tests and field performance data may indicate that a fluid which might not have been thought suitable on the basis of measurements on the fluid itself gives satisfactory performance, or vice versa.)

We therefore evaluate fluids by impregnating, under carefully controlled conditions, commercial 2 μF capacitors bought in large quantities. These are life tested under conditions specified by Electronic Industries Association and under even more rigorous conditions to assess fluid performance on a comparative basis.

This type of testing plus field results reported to us to date indicate that any of these fluids can be used to produce acceptable commercial capacitors. However, it is important to recognize that there are significant differences among them which a capacitor maker must evaluate for himself in the light of his production methods and facilities, the design he proposes to use, the use for which the capacitor is intended and his desire for ultimate performance. He must also evaluate the relative importance of environmental factors and fire resistance. Last but not least he must examine the economics of the complete operation.

It is probably safe to say that trichlorobiphenyl fluids give the ultimate in performance in a broad range of impregnated capacitor types with exceptions which represent only a very small part of the field. However, as noted above they have been accused of being environmental pollutants.

The fire resistance of the trichlorobiphenyl fluids is an advantage which should not be given up without considering whether advantages to be gained are worth the loss. Due to the fact that PCB fluids have been universally used, fires from capacitors are so rare that we have come to discount the possibility. This is because of the fact that gases expelled by a failed trichlorobiphenyl capacitor are not ignited even by a spark such as is used to ignite an oil furnace burner. Gases from capacitors impregnated with the other types of fluids discussed above ignite and burn like a gas flame.

Esters are widely available low price materials. Against the low price per unit volume the manufacturer must weigh the fact that they require special purification. Because of their susceptibility to hydrolysis, excess recycled material from capacitor production may be especially difficult to upgrade for reuse. Special care is required during the impregnation operation in order to produce capacitors with good service reliability (5a).

The hydrocarbon base fluids described above are preferred over ester fluids by some manufacturers because they are more easily treated, the recovered material is more easily up-graded for reuse and because they seem to have greater ultimate performance in A.C. capacitors than the ester fluids. This difference was demonstrated in a test under rather greater stresses than normal, but very long term tests at lower stresses show the same trend in less dramatic fashion. Table V shows the accelerated test data.

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TABLE V

A.C. TEST DATA

Impregnant	Percent Failed	Unit Hours
Aroclor 1016	25	7844
MCS 1475 (ester type)	100	988
MCS 1588 (hydrocarbon type)	25	7936

Test Conditions: 8 capacitors per set, paper dielectric, 1000 volts A.C./min, 90°C, Total Possible Unit Hours/set 10,208 hrs.

MCS 1475 and MCS 1588 also seem to have promise in D.C. capacitors. Data in support of this are shown in Table VI.

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TABLE VI

D.C. TEST DATA

Impregnant	Percent Failed	Unit Hours
Aroclor 1016 + 0.3% Unox 4221	87.5	2,498
Aroclor 1016 + 0.5% anthraquinone	25	3,497
MCS 1489 + 0.5% anthraquinone	25	3,857
MCS 1475 + 0.5% anthraquinone	12.5	3,933
MCS 1588 + 0.5% anthraquinone	0.0	4,416

Test Conditions: 8 capacitors per set, paper dielectric, 1000 volts D.C./mil, 85°C, Total Possible Unit Hours/set 4,416 hrs.

It is well known that chlorinated biphenyl fluids must have an additive to make them useful in D.C. capacitors. As can be seen from the data in Table VI, Aroclor 1016 plus epoxide, a good A.C. stabilizer, gives relatively poor D.C. performance. With anthraquinone, the results are much better. MCS 1489 with the same stabilizer gives slightly better performance. MCS 1475 with the same stabilizer still better results, and MCS 1588 shows no failures to date.

(As can be seen by comparison of data in Figures 1, 2, 3 and 4,) both esters and hydrocarbon base fluids can be used to much lower temperatures than trichlorobiphenyl. However, suitable trichlorobiphenyl-trichlorobenzene mixtures can also be used at very low temperatures.

Another important difference among these types of fluids is that trichlorobiphenyl is not ideal for use in metallized paper or film capacitors which are expected to "self heal". This is because the chlorinated compounds generate hydrogen chloride which may lead to permanent breakdown even when a very minor discharge takes place. MCS 1475 is reported to show good performance in metallized paper capacitors. MCS 1588 would probably be even better. This may seem surprising in view of their sulfur content which many would expect would lead to the formation of sulfuric acid under arcing conditions within a capacitor. In fact, however, the conditions during arcing are highly reducing so that

the sulfone decomposition products are polysulfones and reduced sulfur compounds, none of which cause permanent failure.

In summary, it can be said that, if environmental or other considerations should ever become important enough to outweigh "the significantly increased risk of fire and explosion and the disruption of electrical service which would result from a ban on PCB use" as recognized by the Interdepartmental Task Force on PCB's, the capacitor maker now has a number of possible alternate fluid impregnants to choose among. He must evaluate these carefully and make his choice giving suitable weight to the importance of each of the factors involved. More commercial experience with the new fluids will be necessary to evaluate their place in capacitor technology.

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TABLE II

PROPERTIES OF AROCLOR TYPE DIELECTRIC FLUIDS

Properties	Aroclor 1016	MCS 1043	MCS 1489
	Data on Fluids		
Sp. G. 25/25°C	1.368	1.246	1.348
DK 25°C	5.9	6.0	8.1
100°C	4.85	5.0	6.5
Tan δ 60 Hz 100°C	0.0025	0.0006	0.05
R_S 100°C x 10^9 Ω cm	150,000	10,000	2,000
Viscosity, cs.			
@ 100°F.	14.3	5.6	21.5
@ 210°F	2.12	1.47	2.54
Flash Point °F.	358°	318°	358°
Fire Point °F	None to BP (620)	None to BP (515)	610°
AIT °F.	1185°	1155°	1125°
Hydrolysis **	0.00	0.00	0.00
	Data on 2 μF Capacitors* Impregnated with Above Fluids		
Capacitance @ 25°C.	1.95	1.99	2.20
Dissipation Factor			
@ 25°C.	0.0034	0.0037	0.0040
Capacitance @ 90°C.	1.83	1.89	2.07
Dissipation Factor			
@ 90°C.	0.0032	0.0037	0.0033
Terminal to Case Resist.	240 x 10^9		89 x 10^9

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* Standard 2 μF, 585 VAC Capacitors, 2 layers 0.00066, 0.90 density paper, 0.3% Unox† 4221.

** Neutralization number of sample with 0.5% water added heated for 168 hrs. at 210° in a stainless steel bomb along with an aluminum and a mild steel coupon.

† Registered trademark of Union Carbide Corp.

TABLE III

PROPERTIES OF ESTER TYPE DIELECTRIC FLUIDS

Properties	DOP	DINP	MCS 1475
	Data on Fluids		
Sp. G. 25/25°C	0.983		0.988
DK 25°C	5.3	4.6	6.2
100°C	4.3	3.9	5.0
Tan δ 60 Hz 100°C	0.075	0.01	0.022
R_s 100°C x 10^9 Ω cm	211	1900	500
Viscosity, cs.			
@ 100°F.	29.0		26.4
@ 210°F	4.3		4.33
Flash Point °F.	440°	430°	432°
Fire Point °F	-	495°	495°
AIT °F.	-	-	730°
Hydrolysis**	0.11	0.15	0.17
	Data on 2 μF Capacitors* Impregnated with Above Fluids		
Capacitance @ 25°C.	1.85		1.95
Dissipation Factor @ 25°C.	0.0033		0.0034
Capacitance @ 90°C.	-		1.84
Dissipation Factor @ 90°C.	-		0.0032
Terminal to Case Resist.	1 x 10^9		2.6 x 10^9

*Standard 2 μF, 585 VAC Capacitors, 2 layers 0.00066, 0.90 density paper, 0.3% Unox 4221 added.

**Neutralisation number of sample with 0.5% water added heated for 168 hrs. at 210°F in a stainless steel bomb along with an aluminum and a mild steel coupon.

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TABLE IV

PROPERTIES OF HYDROCARBON BASE DIELECTRIC FLUIDS

Properties	MCS 1238	MCS 1588
	Data on Fluids	
Sp. G. 25/25°C	1.018	1.030
DK 25°C	6.0	6.1
100°C	5.1	5.1
Tan δ 60 Hz 100°C	0.046	0.1
R_s 100°C x 10^9 Ω cm	1,000	620
Viscosity, cs.		
@ 100°F.	9.1	7.14
@ 210°F	1.98	1.96
Flash Point °F.	303°	325°
Fire Point °F	328°	350°
AIT °F.	835°	870°
Hydrolysis**	0.00	0.00
	Data on 2 μF Capacitors* Impregnated with Above Fluids	
Capacitance @ 25°C.	1.97	1.98
Dissipation Factor @ 25°C.	0.0037	0.00361
Capacitance @ 90°C.	1.87	1.87
Dissipation Factor @ 90°C.	0.0039	0.0035
Terminal to Case Resist.	-	51 x 10^9

*Standard 2 μF, 585 VAC Capacitors, 2 layers 0.00066, 0.90 density paper, 0.3% Unox added.

**Neutralization number of sample with 0.5% water added heated for 168 hrs. at 210°F in a stainless steel bomb along with an aluminum and a mild steel coupon.

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Fig. 1

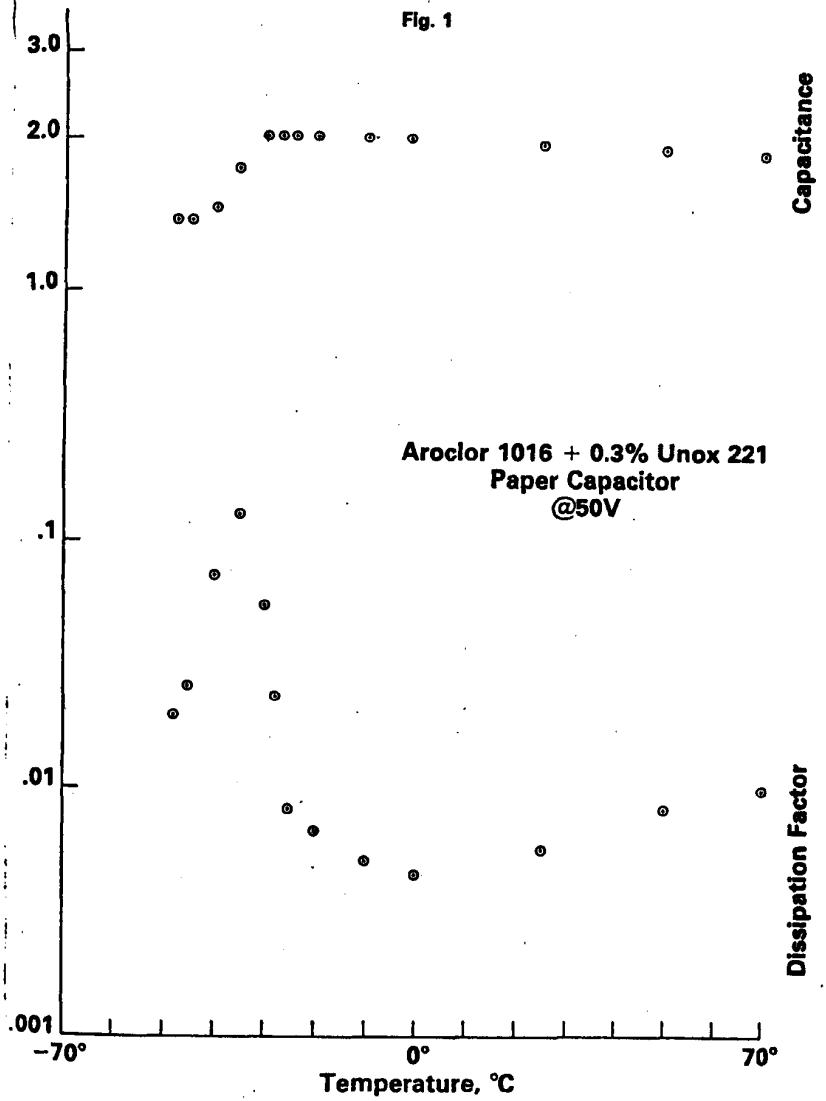

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Fig. 2

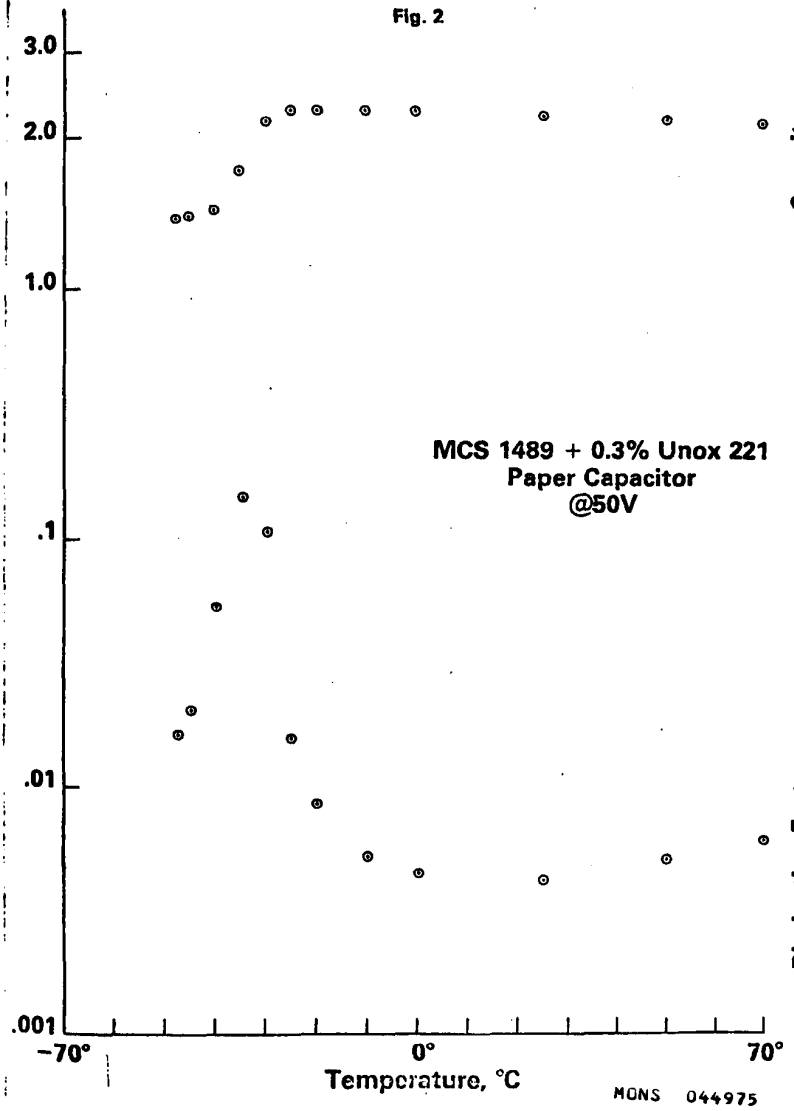
3.0
2.0
1.0
.1
.01
.001
Capacitance
Dissipation Factor
MCS 1489 + 0.3% Unox 221
Paper Capacitor
@50V
−70°
0°
70°
Temperature, °C
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Fig. 3

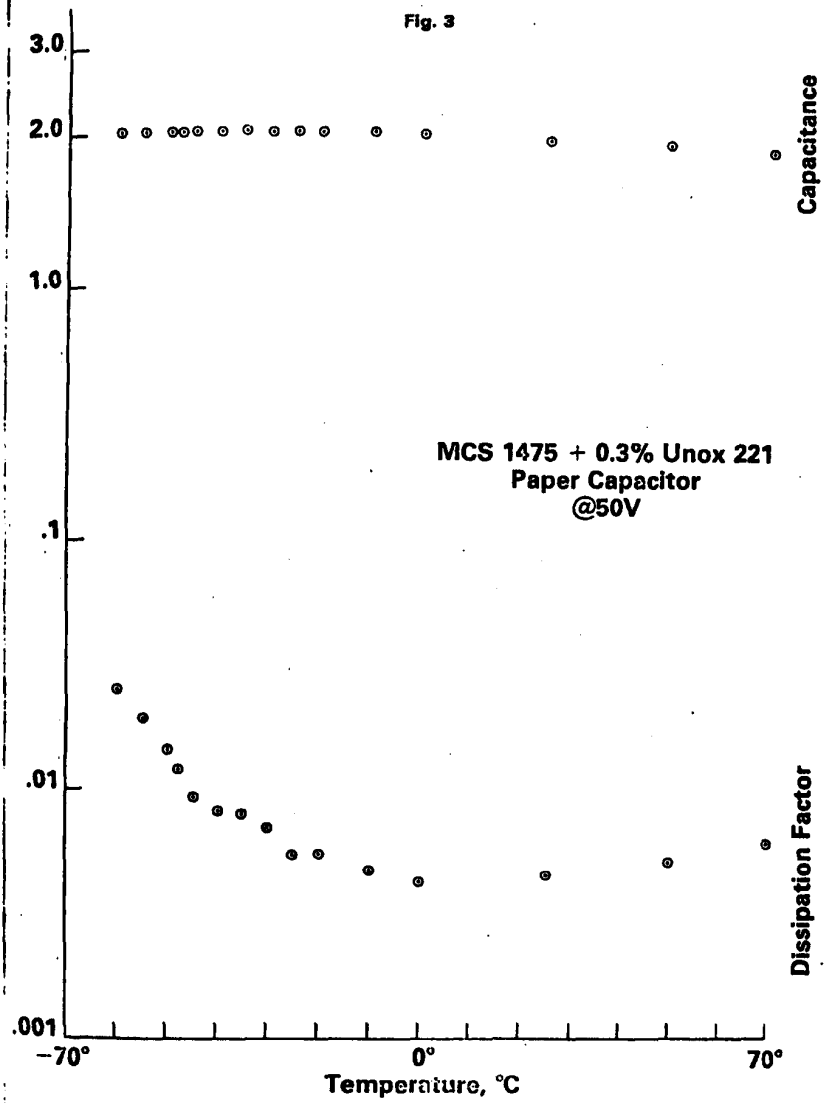

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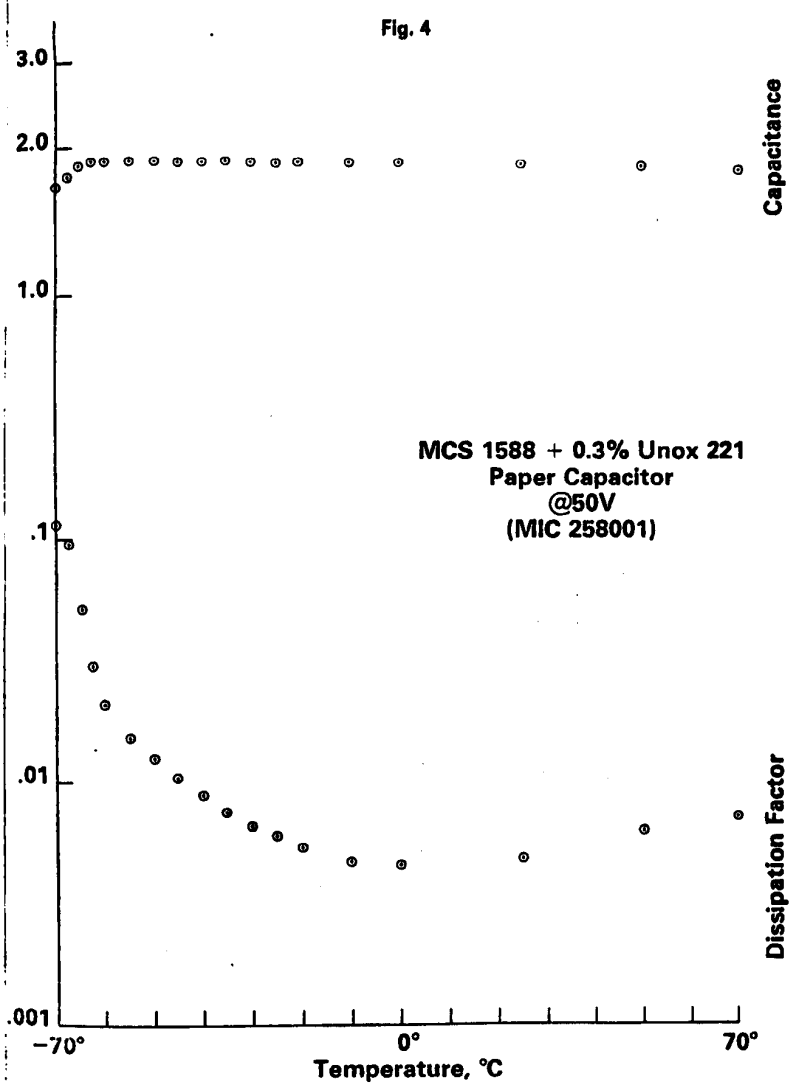
Fig. 4



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