72-9095①

ORNL-6603

OAK RIDGE NATIONAL LABORATORY

MARTIN MARIETTA

Updated Evaluation of Polymer Films for Electrical Insulation

H. E. McCoy, Jr.

MASTER

OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831; prices available from (615) 576-8401, FTS 626-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161.
NTIS price codes—Printed Copy: A05 Microfiche A01

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ORNL--6603

DE91 000544

Metals and Ceramics Division

UPDATED EVALUATION OF POLYMER FILMS FOR ELECTRICAL INSULATION

H. E. McCoy, Jr.

Publication Date: August 1990

Prepared for the
Assistant Secretary for Conservation and Renewable Energy
Office of Energy Storage and Distribution
Electric Energy Systems Program
AK 06 02 02 0

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831-6285
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under Contract DE-AC05-84OR21400

CONTENTS

LIST OF FIGURES

Figure Page

LIST OF TABLES

UPDATED EVALUATION OF POLYMER FILMS FOR ELECTRICAL INSULATION*

H. E. McCoy, Jr.

ABSTRACT

Several types of tests have been run on polymer film materials that could be useful for electrical insulation. The polymers studied were polyethylene terephthalate, polycarbonate, polysulfone, polyethersulfsone, polyetherimide, ultrahigh-molecular-weight polyethylene, polyimide, polybutylene terephthalate, and a laminate of Kraft paper and polypropylene. Thermal aging tests were run to 60,000 h on several of the polymers, and the samples were evaluated by tensile tests, electrical breakdown tests, and immersion density measurements.

Because of the wide range of potential service conditions, tensile tests were run on as-received materials over the temperature range of −196 to 200°C. Polyimide is probably the only material suitable for the extremes of this temperature range, but many of the other polymers would be suitable for intermediate temperatures. Creep tests were run in nitrogen and transformer oil at 90°C. It was found that some polymers are weaker and less ductile in oil than in nitrogen and that other polymers have equivalent properties in the two environments.

A means of applying mechanical, thermal, and electrical stresses simultaneously to polymer samples was developed. Tests were run at 90°C in transformer oil on polyethylene terephthalate, polyimide, and polyethersulfone. Tests thus far do not indicate that the creep rate is affected by the application of a 5 kV dc potential.

INTRODUCTION

The Division of Electric Energy Systems of the U.S. Department of Energy is developing equipment to improve power transmission capabilities and reliability, to increase efficiency, and to reduce the environmental

---

*Research sponsored by the Office of Energy Storage and Distribution, Electric Energy Systems Program, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

impact of electric utility transmission systems.[1] The program described in this report was initiated to directly support the efforts to develop a transformer utilizing aluminum conductors polymer insulation, and SF_6 dielectric.[2] Thus, many of the materials and test conditions were influenced by this application. As the more general applicability of polymers for many insulation applications became apparent, other polymer films were involved in the study. The new developments in high-temperature superconductors will make it necessary to have insulators with suitable properties at temperatures of −225 to −175°C. Polymers may be able to fulfill some of these needs, and our program has been modified to determine their properties at applicable temperatures. Several activities were involved in this study, and they are described in this report.

Most polymers were developed for nonelectrical applications, so numerous properties must be measured and evaluated. For example, properties such as dielectric strength, mechanical strength, and maximum use temperature must be evaluated, and chemical compatibility with the particular service environment must be assessed. Properties with such second-order effects as thermal expansion, anisotropy, and creep resistance need to be evaluated. If the application is a long-term one extending over months or years, the stability of these properties must be known as a function of time. This stability question is loosely referred to as aging, although property changes with time are influenced by thermal, electrical, and mechanical stressing. In some cases the chemical environment also plays a significant role in aging, so the conditions during aging must be specified and carefully controlled for the results to be meaningful.

Test retorts were constructed to expose polymer films under conditions involving temperature, mechanical stress, and environment as variables. The exposed polymer samples were tested to evaluate changes in tensile properties, dielectric strength, and density. Samples were exposed up to 60,000 h at temperatures ranging from 70 to 160°C. Short-term tensile properties were determined over the temperature range of −196 to 200°C. Tensile tests were also run at 90°C in air and in transformer oil to determine whether oil altered the properties. A laminated Kraft paper-polypropylene product was also subjected to tensile tests. For long-term applications, creep, or deformation with time, is of concern; and measurements were made in nitrogen and oil environments. A question of concern for polymers is whether they age differently when exposed to single or multiple stresses. The stresses of particular concern are thermal, mechanical, and electrical, and a test has been set up in which all three stresses can be applied simultaneously to a polymer sample. Tests were performed on three polymers, and these results are presented.

EXPERIMENTAL DETAILS

TEST MATERIALS

Several commercial test materials were selected for evaluation; the available information is summarized in Table 1. The program initially concentrated on the gas transformer concept, with polyethylene terephthalate as the reference material. Four other materials were chosen for evaluation in light of the possibility that polyethylene terephthalate might not be adequate for the application or that one of the alternate materials might have some distinct operation advantage. These materials were polycarbonate, polyethersulfone, polyparabanic acid, and polyimide. Two prototypic transformer coils were wound with type-EL polyethylene terephthalate to perform tests on that material. Polysulfone, polyetherimide, ultrahigh-molecular-weight polyethylene, and polybutylene terephthalate were added to the program because their physical and mechanical properties seemed attractive. Multiple lots of some of the materials were obtained to give some indication of lot-to-lot variation.

Although all these materials were commercially available at the time of procurement, the status of Tradlon, or polyparabanic acid, has since changed significantly. The material was produced by Exxon, which has discontinued its manufacture. The production facility is for sale, however, so it may be reactivated and the product may again become available.

Table 1. Test materials

Material	Trade name	Desig-nation	Thickness mm	Thickness in.	Supplier
Polyethylene terephthalate, type C	Mylar	PET	0.0254	0.001	Du Pont
Polycarbonate	Merlon	PC	0.0254	0.001	Mobay
Polyethersulfone	Victrex	PES	0.0635-0.1016	0.0025-0.0040	ICI Americans
Polyparabanic acid	Tradlon	PPBA	0.0254	0.001	Exxon
Polyimide, type H	Kapton	PI	0.0254	0.001	Du Pont
Polyethylene terephthalate, type EL	Mylar	PET	0.0254	0.001	Du Pont
Polysulfone	Thermalux	WPS APS	0.0762 0.0305	0.0030 0.0012	Westlake Ain
Polyetherimide	Ultem	WPEI APEI	0.0762 0.0254	0.0030 0.0010	Westlake Ain
Polyethersulfone	Victrex	WPES APES	0.0508 0.0635	0.0020 0.0025	Westlake Ain
Polyimide	Kapton	API	0.0533	0.0021	Ain
Ultrahigh-molecular-weight polyethylene		UHMWPE	0.1270	0.005	Polypenco. Inc.
Polybutylene terephthalate	Valox	PBT	0.0254 0.0381 0.508	0.010 0.015 0.020	Du Pont

EQUIPMENT

Much of the equipment used in this work was described in detail previously.[3,4] The equipment will be described in this report only as required to interpret the test results.

THERMAL AGING STUDIES

The polymer sheets were alternated between aluminum in the configuration shown in Fig. 1. The polymer sheets were larger than the aluminum, so that the center of the polymer sheet was compressed and the outer portion unstressed. The compressive force was supplied by a hydraulic piston that pushed on top of the stack of aluminum and polymer sheets. The stacks were separated so that they ran at different temperatures of 70, 110, 135, and 160°C. This entire assembly was contained within a tubular section to provide atmospheric control.

Three such chambers were used to expose samples in three environments: SF_6 with a partial pressure of water of 40 Pa (400 μatm) (Chamber A), SF_6 with a partial pressure of water of 4 Pa (40 μatm) (Chamber B), and SF_6 with a partial pressure of water of 4 Pa (40 μatm) and 1 vol % of Freon-113 (Chamber C). The total pressure in each chamber was 0.4 MPa (4 atm). Freon-113 was added to simulate coolant inleakage in systems where Freon-113 is used as a coolant. In the first year of these studies it was concluded that the polymers being evaluated were inert toward SF_6 and Freon-113. As the program turned from the gas transformer, the test gas was changed to nitrogen, and the Freon-113 additive was dropped. Water content was found to be important with some polymers, so the initial water levels were maintained.

Exposed samples were removed periodically from the chambers, and new s inserted over the approximately 60,000 h that the exposures were in gress. The exposed polymer samples were then subjected to several sts. Specimens with the configuration shown in Fig. 2 were prepared with a steel-rule die so that the gage portions consisted of either stressed or unstressed material. Electrical breakdown measurements were made on the film materials. Immersion density measurements were made on ll pieces of sample as the easiest way to detect changes in the degree

ORNL–DWG 81-11497A

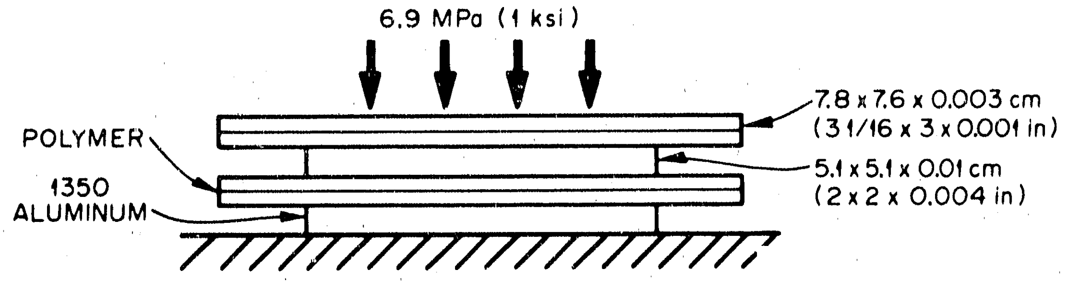

Fig. 1. Schematic diagram of test setup for aging sheets of polymers.

ORNL-DWG 81-11498

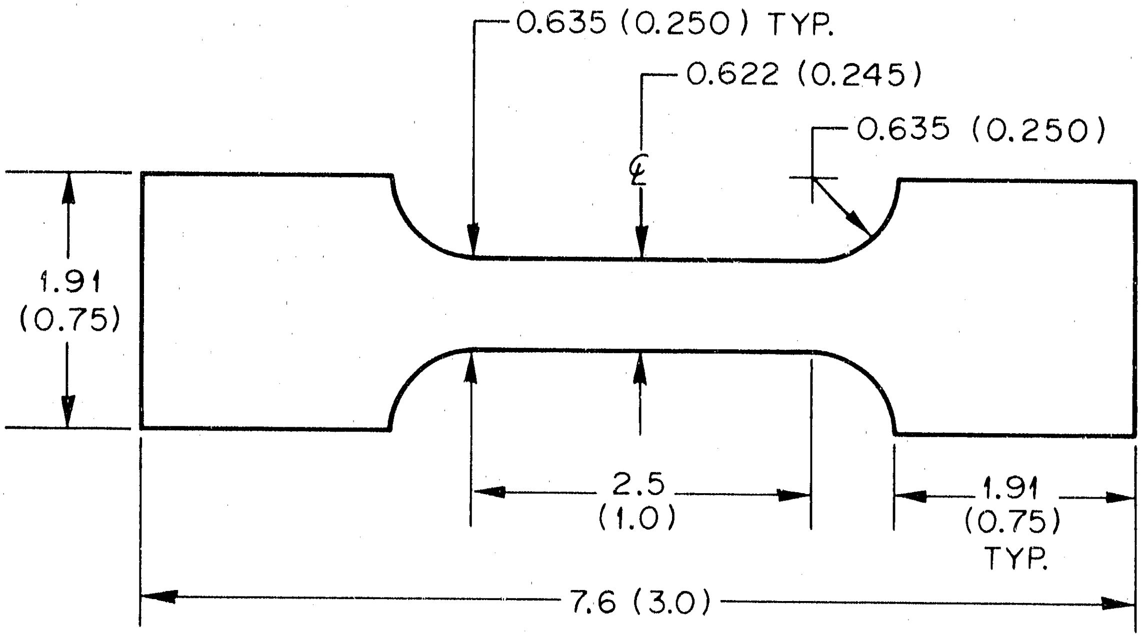

Fig. 2. Diagram of polymer tensile sample. Dimensions are in centimeters (inches).

of crystallinity with aging. Scanning electron microscopy was performed on the fractures of selected tensile specimens or on the as-exposed surfaces to detect changes in surface morphology.

PROCEDURES FOR TENSILE TESTS

The tensile properties of polymer films are generally determined by the test method described in ASTM D 882. In this procedure, strips of uniform width and thickness were cut and tested. The shortest test specimen allowed was 100 mm (4 in.), and the length to be used for high-precision work was 250 mm (10 in.). The procedure for testing plastics in more conventional thicknesses is given in ASTM D 638, and the test specimens have gage sections. Our test specimen was very similar to type IV in that specification, but the overall length of our specimen was less. Several testing speeds are recommended in the two specifications for various types of materials.

The procedure we adopted for our testing was a hybrid of the two specifications just described because the size of the material being exposed (76- × 76-mm exposure coupon) controlled the overall length of the test specimen. The dimensions of our test specimen are shown in Fig. 2. The test section was 25 × 6.22 mm (1.0 × 0.245 in.). This geometry permitted specimens to be cut from the stressed or unstressed regions of the aged sheets and oriented either in the machine direction (MD) or transverse direction (TD) within the plane of the sheet. The specimens were cut with a steel-rule die having the shape of the specimen. The test material was backed by Plexiglas and the die pressed into the material with a force of about 22 kN (5000 lb). The cutting edges of the die were honed periodically to keep them sharp enough to produce test specimens with smooth edges.

The tensile specimens were tested in an Instron testing machine with a special set of grips in which the ends of the sample were wrapped around rubber rollers. As the sample was pulled, the gripping force increased and prevented slippage. A 222-N (50.0-lb) load cell was used, and the breaking force rarely exceeded 44 N (10.0 lb). The strain rate used was 0.0033 s^{-1} (0.20 min^{-1}).

Some tensile tests on as-received materials were performed at temperatures from −196 to about 200°C. The test specimen was of the same geometry as the one shown in Fig. 2. Holes were punched in each end, and the same aluminum grips used in creep testing were used to grip the sample. The sample and grips were enclosed in a tube furnace for heating or in liquid nitrogen or alcohol-dry ice for cooling. The test strain rate and the load cell were the same as utilized in the tests at 25°C.

Data analysis was done entirely from the charts of load versus crosshead travel and consisted of determining the 0.2% offset yield stress, the ultimate tensile stress, and the elongation to fracture. The results exhibited considerable scatter, which must be considered when drawing conclusions. It is likely that the main contributor to the scatter was the variation in smoothness of the gage-section edges, with the smoother samples straining the most before fracture. This variation had little effect on the yield strength, so the scatter in yield strength is small. For a material that exhibits little strain hardening, large scatter in the fracture strain has little effect on the ultimate tensile strength. For a material that exhibits high strain hardening, a reduction in fracture strain results in a lower ultimate tensile strength. Thus, the edge quality of tensile specimens results in large variations in fracture strain and ultimate tensile strength for materials with high strain hardening. Thickness variation is another parameter that could contribute to the scatter, but only polyethersulfone exhibited measurable thickness variation. The minimum thickness within the gage section was used in computing stresses. Generally, we tested six samples to obtain properties of as-received materials and two samples for each test condition. The average properties and the standard deviation are reported for each test condition.

ELECTRICAL BREAKDOWN EQUIPMENT

Most of the electrical breakdown measurements in this report were made with a Biddle AC/DC Breakdown, Leakage, and Ionization Tester RM Z-15-L/2, which is capable of 12 kV dc and 6 kV ac (rms). We designed and fabricated the measuring probes, which consisted of two opposing cylindrical brass rods 6.4 mm (0.25 in.) in diameter with edges rounded to

0.8-mm (0.031-in.) radius (electrode type 3 in ASTM D149). The bottom electrode was stationary, and the top electrode was movable to permit inserting samples. The top electrode was counterbalanced to result in a net force on the specimen of 50 ± 2 g. The power supply was designed so that the potential could be increased manually until breakdown occurred. This procedure was somewhat operator-dependent, but the potential was normally increased from zero to the breakdown voltage in about 30 s. The voltage rise rate was 100 to 200 V/s, well within the specified rate of rise for the short-time test conditions in ASTM D149. The dc breakdown voltage was usually well defined and not very dependent on operator technique. The ac breakdown voltage was more dependent on operator technique, and surface ionization occurred to such an extent that the sample could be damaged locally. Thus, ionization could lead to formation of a weak area where breakdown would occur. For example, breakdown occurred at about 75% of the normal breakdown voltage if ionization was allowed to occur for a few minutes at the 75% voltage.

Testing experience in this area revealed the need for better control of the voltage rise rate and the ability to go to higher potentials to cause electrical breakdown in thicker samples. Two Hipotronics power supplies were procured to meet those needs. One supply is capable of 50 kV dc and the other of 50 kV ac, and the rate of voltage rise of each supply is adjustable but controlled automatically. This equipment makes use of the electrodes described on the Biddle power supply and the voltage rise rate of 1 kV/s selected for both ac and dc tests. Most of the future testing will be performed with the Hipotronics equipment.

The electrode assembly is in a clear plastic enclosure, which was purged with dry nitrogen, with test samples introduced through a small door. The samples could be placed with a manipulator between the electrodes for measurements without breaking the seal between the nitrogen enclosure and air.

CREEP CHAMBER WITH NITROGEN ENVIRONMENT

A schematic diagram of the chamber used for creep testing is shown in Fig. 3. The specimen shown in Fig. 2 was also used for these tests, with the ends punched and aluminum pads bolted over each end for support while

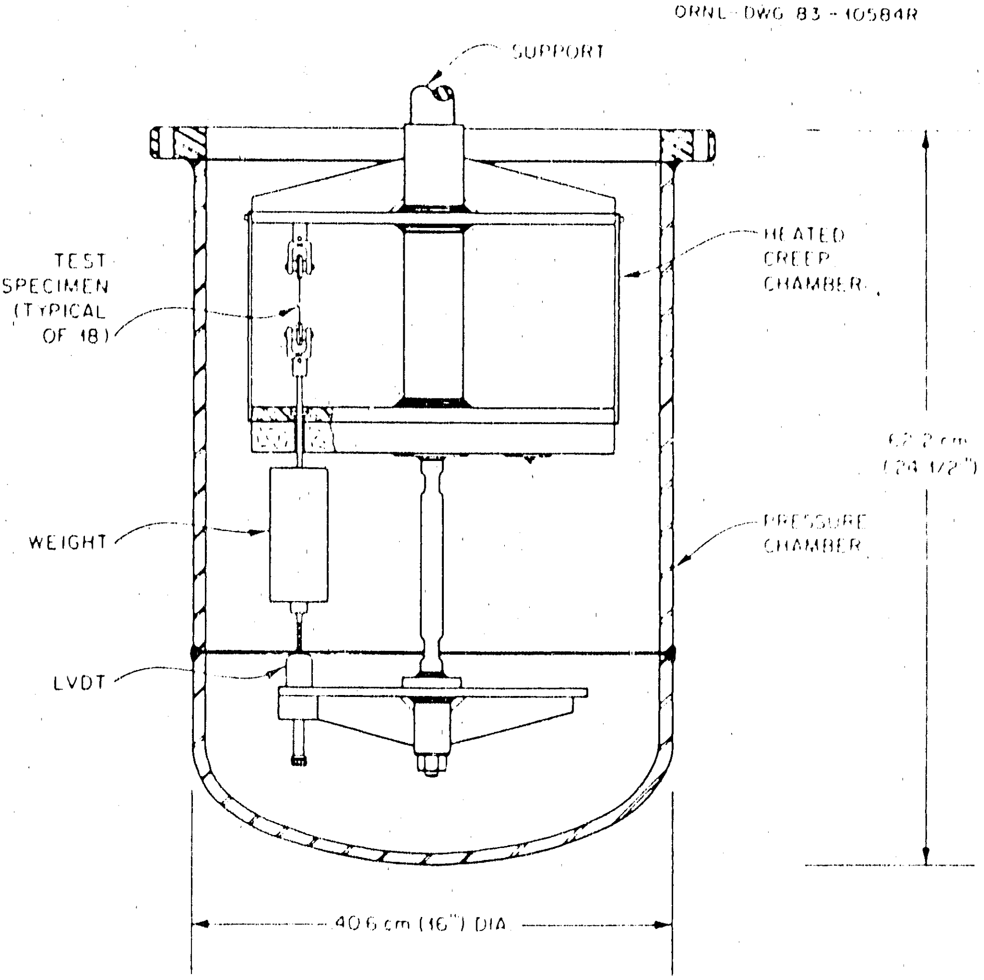

Fig. 3. Schematic of test chamber for creep testing of polymer samples. One of 18 samples is shown in place. A linear variable differential transformer (LVDT) is used to measure creep strain.

stressing. The stress was applied by weights attached to the bottom of the specimen. The core of a linear variable differential transformer (LVDT) was attached to an extension member from the bottom of the weight, and the coil of the transducer was attached to the bottom support plate. The electronics associated with the LVDT were located outside the test chamber, and the strain readings were recorded at controlled intervals.

CREEP IN OIL

Equipment was developed for creep testing samples of polymer film (Fig. 2) in oil. Figure 4 shows five test units, each of which will test ten samples. A closer view of one of the units is shown in Fig. 5. Each sample is independently stressed and free to creep independently. The test environment was either N60 or N61 grade Exxon Univolt transformer oil. The tests were run at 90°C, and the heat source was a hot-plate for each unit of ten tests. The temperature was controlled with a type K thermocouple sensing element and a proportioning controller.

The oil oxidized slowly because it was exposed to air. Each test was interrupted monthly, the old oil pumped out, and new oil pumped in. Plexiglas boxes have been fabricated to contain each test unit in a dry nitrogen environment. This change cleaned up the test environment markedly.

MULTISTRESS AGING

The test sample, shown in Fig. 6, was cut from thin film by a steel-rule die. The silver strips were formed by vapor-depositing silver on the sample while it was covered by a mask with an opening to define the strip. The strip was about 1×10^{-5} cm thick, 0.51 cm wide, and 7.62 cm long. As shown in Fig. 6, the strips were located on each side so that the potential was applied across the longitudinal center 5.1 mm of the sample. The strip was centered (transverse) from side to side of the specimen and covered only about 20% of the sample width. Attempts to make the strip wider resulted in arcing along the sample surface to the strip on the other side. A view of a test sample held above the test equipment is shown in Fig. 7.

ORNL-PHOTO 5896-85

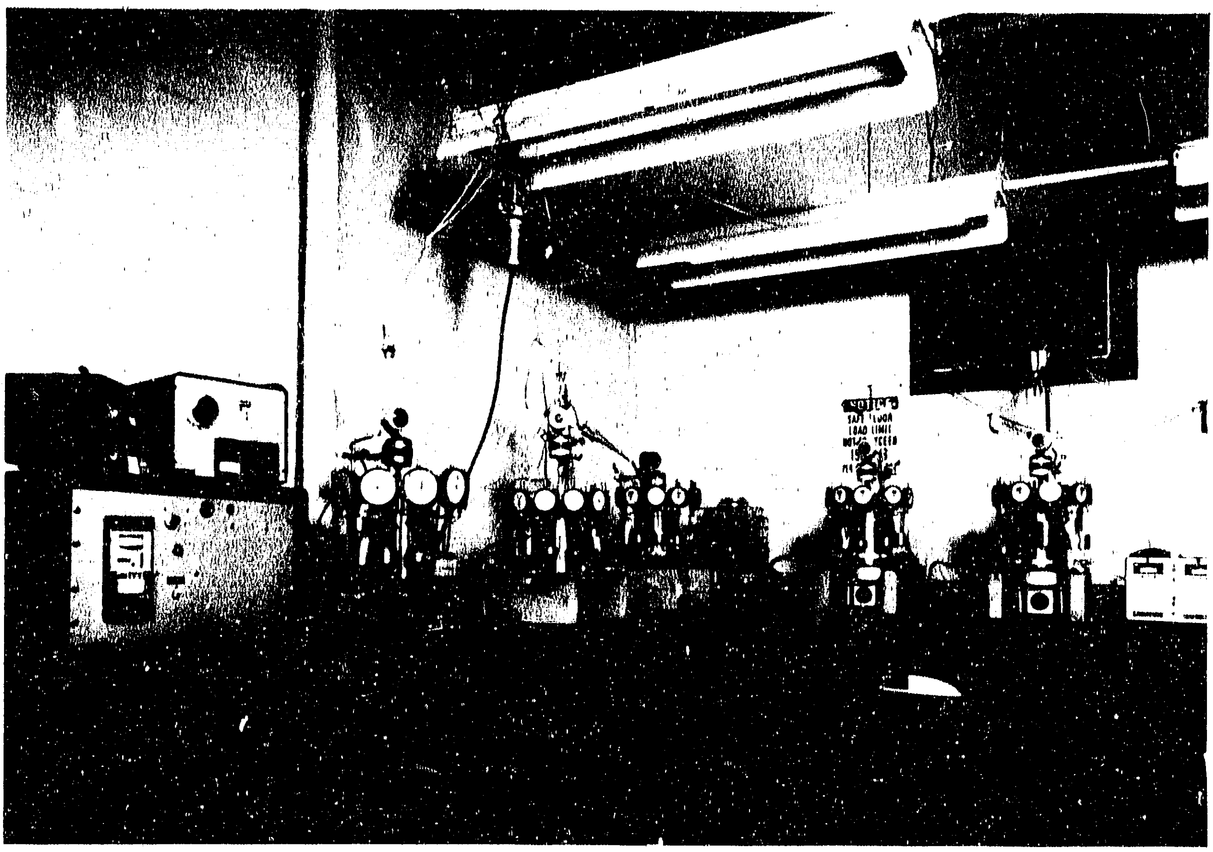

Fig. 4. View of five creep units used to test polymer film specimens in oil.

ORNL-PHOTO 5897-85

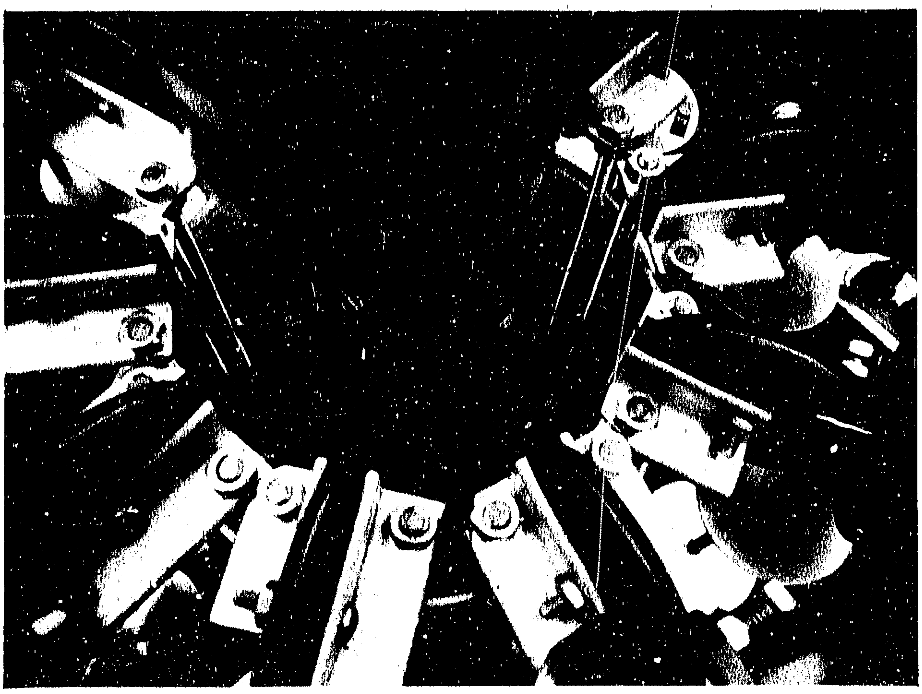

Fig. 5. Close-up of oil creep apparatus. Ten test specimens are in the oil. The load is applied through a wire rope that extends over the pulley. The strain is read on each dial gage.

ORNL-DWG 88M-7895

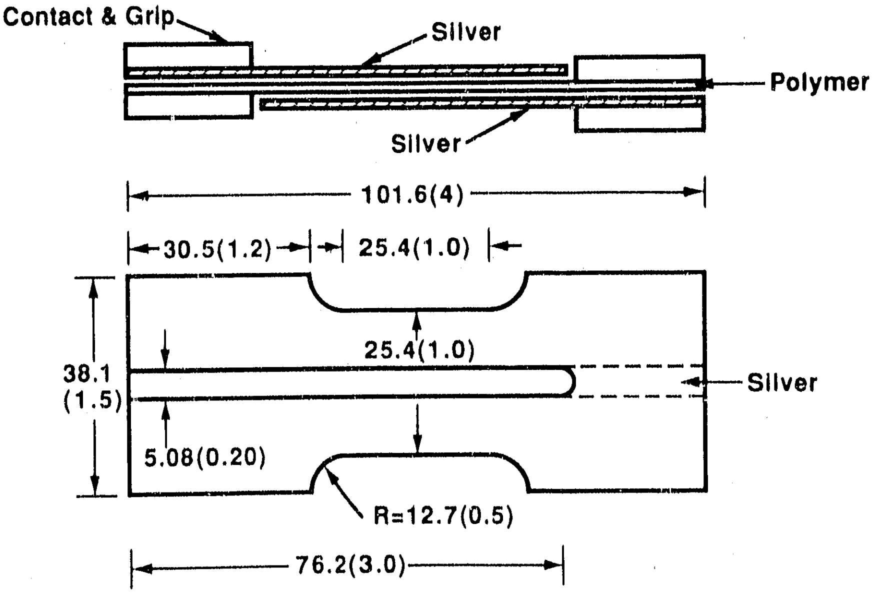

Fig. 6. Test specimen. Thicknesses not to scale. Polymer samples about 2 to 5 × 10^{-2} mm thick, and the silver strip is about 1 × 10^{-4} mm thick. Dimensions are in millimeters (inches). Contact and grip are not shown in plan view.

ORNL-PHOTO 4678-86

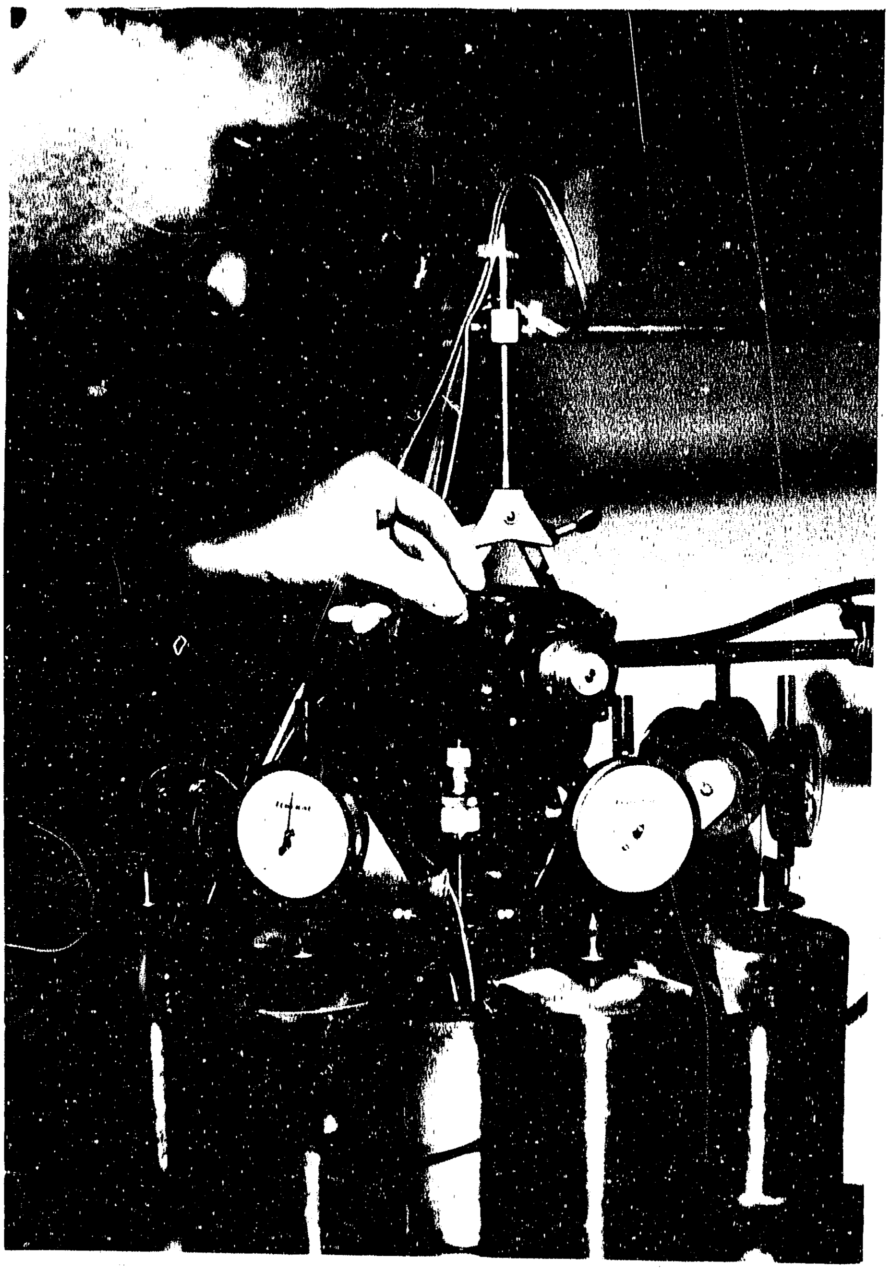

Fig. 7. Apparatus for simultaneously exposing polymer film samples to thermal, mechanical, and electrical stressing. A typical test sample (noted with a large arrow) is held in the operator's hand. Test samples are supported by the bracket noted by the smaller arrow. Samples are immersed in a center vessel of heated oil. The load is applied through the cable linkage by placing shot in the cans. The dial indicators are used to read strain.

General views of the test equipment are shown in Figs. 7, 8, and 9. The center of the apparatus was a vessel partially filled with transformer oil that was stirred and heated. The sample was clamped so that the ends were across the dc power supply. The specimen was immersed in the oil and the mechanical load applied to obtain the desired stress. The loading mechanism involved a steel cable that ran from the top of the specimen over a pulley and attached to a can that contained the appropriate weights. A dial indicator on the load train was used to measure the deformation of the polymer samples.

A Hipotronics dc power supply capable of 50 kV was used to provide the potential across the specimens. The rate of voltage rise was set at 1 kV/s. A current cut-off turned off the supply in the event of short circuits. The most common source of electrical short was the grips. A thin sheet of polymer insulation was placed under one side (the noncontact side) of each grip before it was tightened. However, sufficient creep occurred often enough for the grip to cut through the sample and contact the silver strip on the far side. This was remedied by using a double layer of insulating polyimide film.

The samples were immersed in Univolt N61 insulating oil. This particular oil contains 0.3% by mass antioxidant (2,6-ditertiary-butyl paracresol). The oil was heated and stirred, and the temperature controlled at 90°C. The top of the vessel was open to air, and the oil darkened with time as it oxidized and was contaminated with moisture in a way that is not typical of transformer oils in service. The apparatus will be modified so that the oil surface is exposed only to dry nitrogen.

TEST RESULTS

GENERAL OBSERVATIONS OF AGED SAMPLES

The polymer films were aged in the configuration shown in Fig. 1. The polymer films were in contact with each other and with the thin simulated aluminum conductor. This contact was under a compressive stress of 6.9 MPa (1 ksi). The main concern about the films sticking was that

ORNL-PHOTO 4677-86

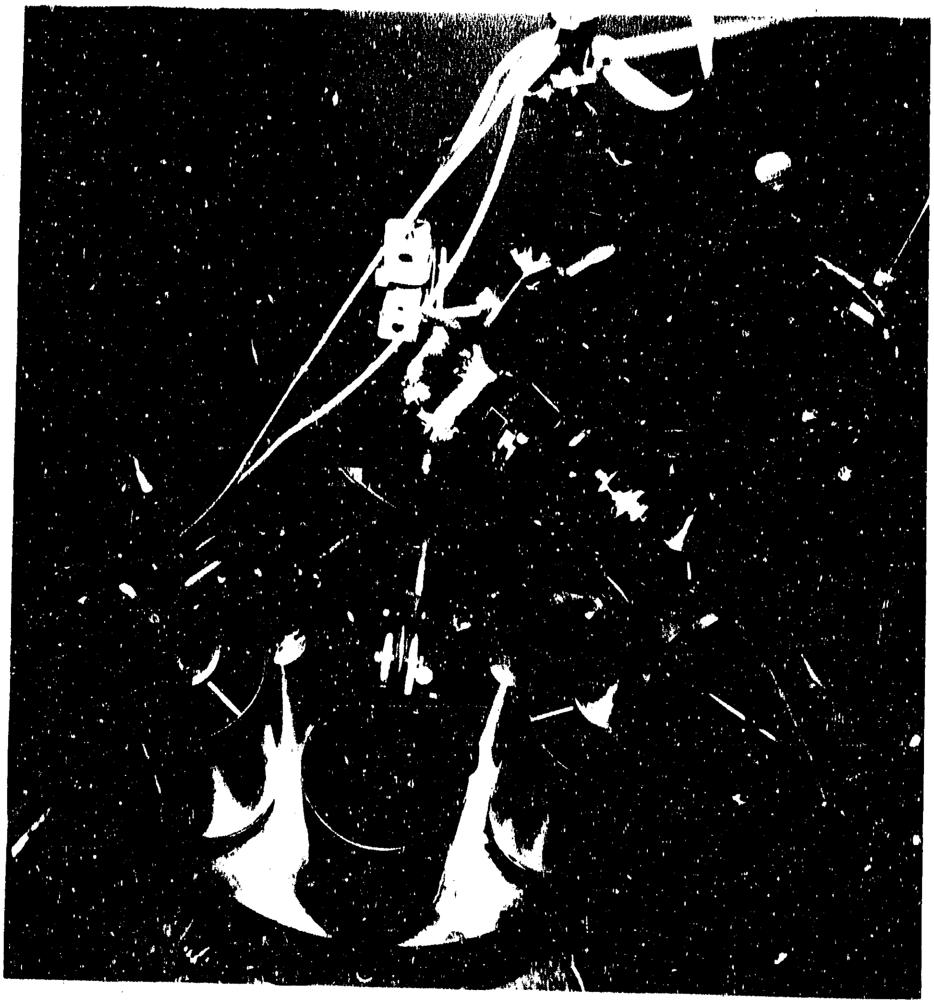

Fig. 8. Top view of apparatus. Note the center vessel where samples are immersed in heated and stirred oil. The support bracket (marked with an arrow) transfers the mechanical load to the test sample.

ORNL-PHOTO 4676-86

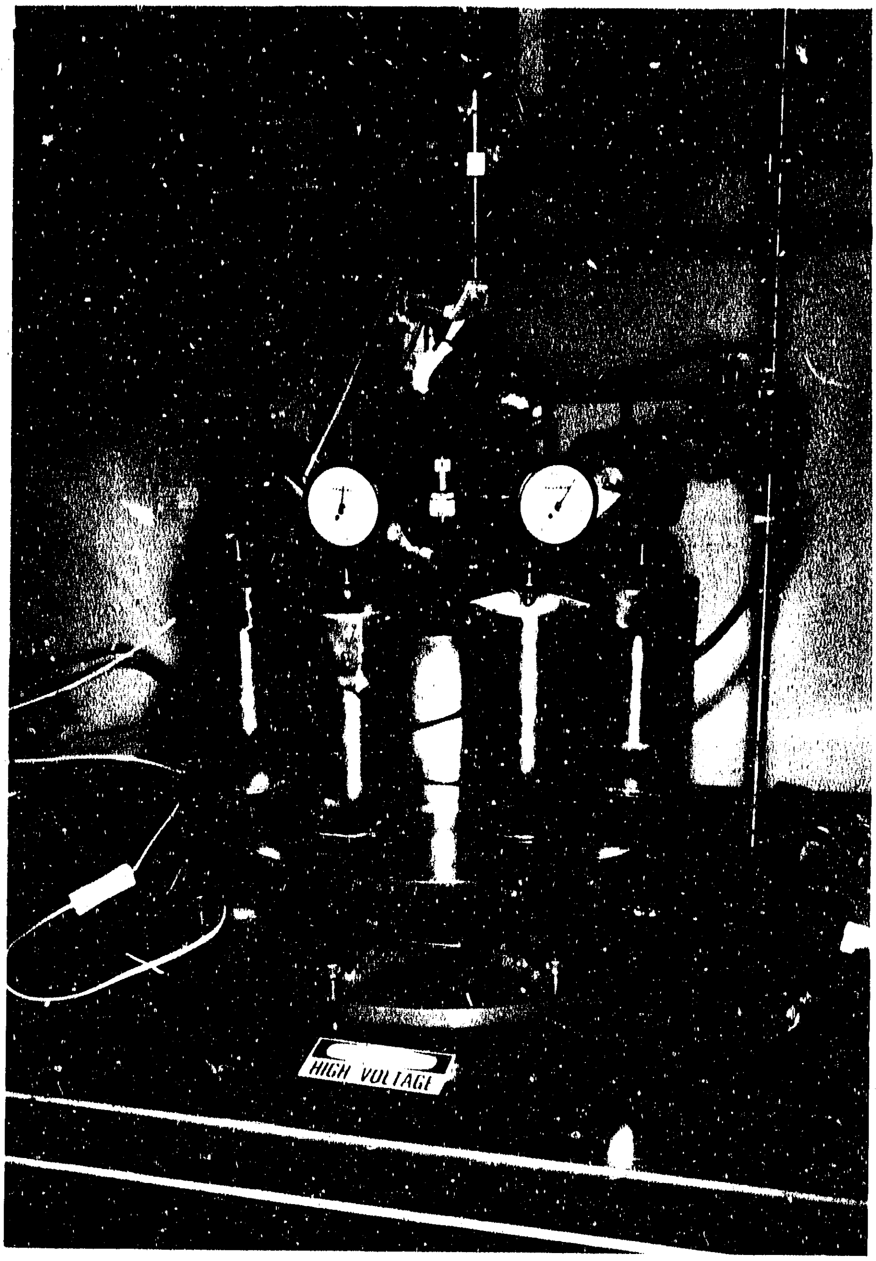

Fig. 9. View of apparatus showing electrical connections.

subsequent movement (due either to thermal expansion or to mechanical forces) might tend to tear the polymer and destroy its usefulness as an insulator.

The observations on the materials that were aged are given in Table 2. Four polymers--polyethersulfone, polyimide, polyarabanic acid, and polyetherimide--did not stick at the highest aging temperature, 160°C. Polysulfone and ultrahigh-molecular-weight polyethylene had intermediate heat resistance, and polyethylene terephthalate and polycarbonate were the least heat-resistant. If the service time was shorter than the experimental times or the degree of contact was not as good, the polymer might have been used at slightly higher temperatures without sticking.

Table 2. Sticking of polymer films

Polymer	Exposure time (h)	Highest temperature at which sticking was not observed[a] (°C)
Polyethylene terephthalate	59,138	70
Polycarbonate	59,138	70
Polyethersulfone	59,138	160
Polyimide	59,138	160
Polyparabanic acid	59,138	160
Polysulfone	37,600	135
Polyetherimide	37,600	160
Ultrahigh-molecular-weight polyethylene	26,400	110[b]

[a] Exposure temperatures were 70, 110, 135, and 160°C.

[b] Not run at higher temperatures.

TENSILE TESTS ON THERMALLY AGED SAMPLES

Tensile tests were performed on duplicate aged specimens. The samples of most polymers were heated almost 60,000 h at temperatures of

70, 110, 135, and 160°C. The basic approach to dealing with this large volume of data is to present a summary of the changes in mechanical properties of each polymer due to aging and show a set of data for one temperature. The notations on the graphs are defined as follows:

MD	machine direction
TD	transverse direction
dry	water partial pressure of 40 μatm
wet	water partial pressure of 400 μatm
stressed	compressive stress of 6.9 MPa (1 ksi)
[no stress notation]	sample aged without applied stress

Some of the detailed observations for shorter aging times were presented previously.[3-6]

Polyethylene Terephthalate

The lot of polyethylene terephthalate was quite anisotropic. Samples with the longitudinal axis parallel to the MD began to deform plastically and work harden so the stress to maintain flow continued to increase. Samples with their longitudinal axis parallel to the TD did not work harden nearly as much, and the ultimate tensile strength was much less in the TD. The fracture strains were about the same in both directions.

Aged at 70°C. Yield strength increased about 10%. The ultimate tensile strength increased about 20% over the first 4000 h of aging and then decreased gradually to about 10% less than original value. The elongation was not altered significantly.

Aged at 110°C. The yield strength was increased about 20%. The ultimate tensile strength in the MD first increased about 30% and then decreased to about 20% less than its initial value. The ultimate tensile strength in the TD remained constant for about 100,000 h and then gradually decreased about 15% to 60,000 h (Fig. 10). The elongation at fracture varied from 25 to 112%, but most of the data fell above the 49% for the as-received material in the MD (Fig. 11).

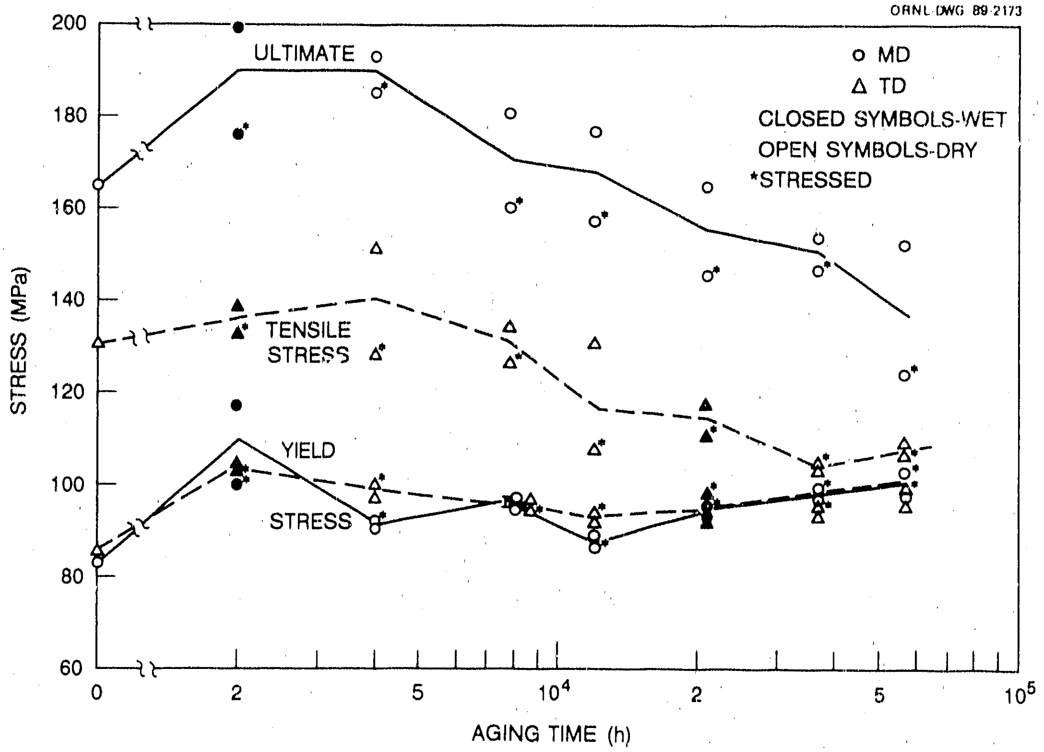

Fig. 10. Influence of aging at 110°C on the tensile properties of polyethylene terephthalate.

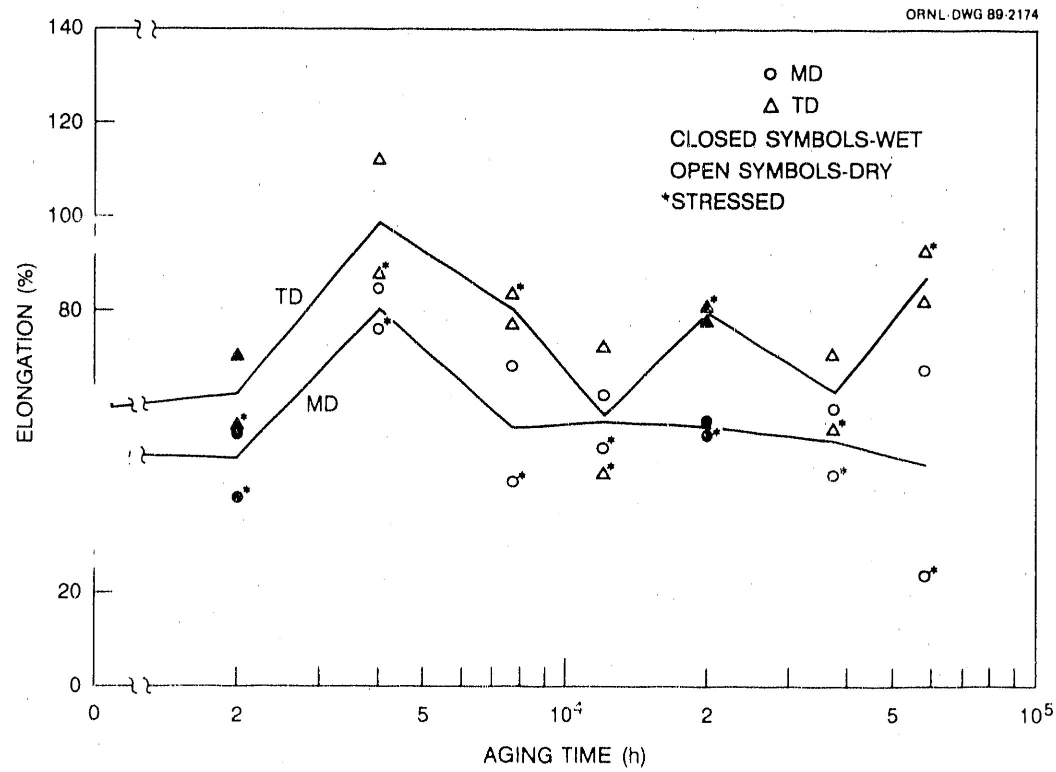

Fig. 11. Fracture strain of polyethylene terephthalate following aging at 110°C.

Aged at 135°C. The yield strength was increased about 10% by aging at 135°C. This value was rather constant except for a set of samples aged 21,000 h in which the yield strength decreased about 10%. The ultimate tensile strength in the MD increased about 40% after a few hours of aging, and then decreased to near the as-received value. The ultimate tensile strength in the TD decreased about 40% over the nearly 60,000 h of aging. The elongation at fracture generally decreased as a result of aging, and the decrease was larger for stressed samples and samples exposed in a moist environment. Seven samples fractured with 10% or less strain, and all of these samples were either stressed or exposed to the higher moisture environment. Thirteen specimens fractured at between 10 and 50% strain (as-received level). Ten of these samples were either stressed or exposed to higher moisture. Ten samples fractured with more than 50% strain; none of these samples was stressed or exposed to the higher moisture.

Aged at 160°C. It was not possible to obtain test samples on much of the stressed material since the degree of sticking was so high that the sheets of polymer could not be separated. The yield strength was increased about 10% except for a group of samples aged 23,000 h in higher moisture. The ultimate tensile strength in the TD was not altered appreciably by aging, but the ultimate tensile strength in the MD was decreased about 25%. Six samples failed at strains of 10% or less. Three of these were either stressed or aged in higher moisture. Five samples fractured at strains between 10 and 50% (as-received level). Two of these samples were aged under stress or high moisture. Six samples failed at higher than 50% strain, and none was stressed or exposed to higher moisture.

Polycarbonate

Polycarbonate is a very anisotropic material. The yield strength is about equivalent in the MD and TD. The ultimate tensile strength in the TD is only about 20% higher than the yield strength, but the ultimate tensile strength in the MD is about 160% higher than the yield strength. The elongation at fracture is about 30% in the MD and 130% in the TD.

Aged at 70°C. Aging caused increases up to about 50% in the yield strength in both orientations and the ultimate tensile strength in the TD. Aging caused the ultimate tensile strength in the MD to gradually decrease about 20%. The elongation at fracture in the MD was altered appreciably by aging. The fracture strain in the TD was decreased from 135% to 10%.

Aged at 110 and 135°C. Aging caused increases up to about 40% in the yield strength in both orientations and in the ultimate tensile strength in the TD. Aging caused the ultimate tensile strength in the MD to gradually decrease about 30%. The elongation at fracture in the TD decreased from 135% to about 10%.

Aged at 160°C. The yield strength in the MD and TD and the ultimate tensile strength in the TD were not greatly affected by aging. There were small variations, but the data were insufficient to determine whether the changes were real or due to scatter. The ultimate tensile strength in the MD showed variations up to 35%, all in the lowered direction (Fig. 12). The elongation at fracture decreased gradually in the MD from 30 to 10%, and in the TD the fracture decreased drastically from 135% to the range of 10 to 20% (Fig. 13).

Polyethersulfone

Polyethersulfone was more heat-resistant than polyethylene terephthalate and polycarbonate and is also more isotropic.

Aged at 70°C. There were no detectable changes in the tensile properties.

Aged at 110 and 135°C. The strength values remained very close to those of the starting material. The fracture strain gradually decreased about 50% to values as low at 9.5%.

Aged at 160°C. The strength values shown in Fig. 14 indicate that the minimum yield and tensile strengths remained very close to that of the starting material. There is some strengthening up to about 25%. The samples aged 23,000 h were in the higher moisture environment. The fracture strain decreased about 50% after aging a few thousand hours and remained nearly constant (Fig. 15).

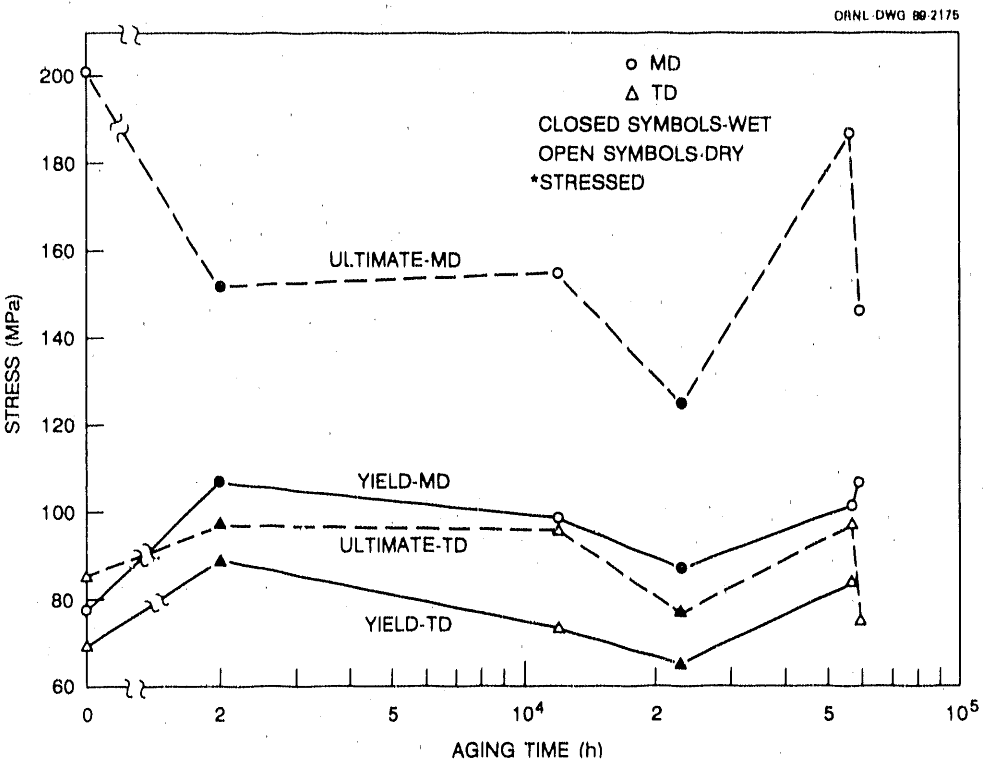

Fig. 12. Influence of aging at 160°C on the tensile properties of polycarbonate.

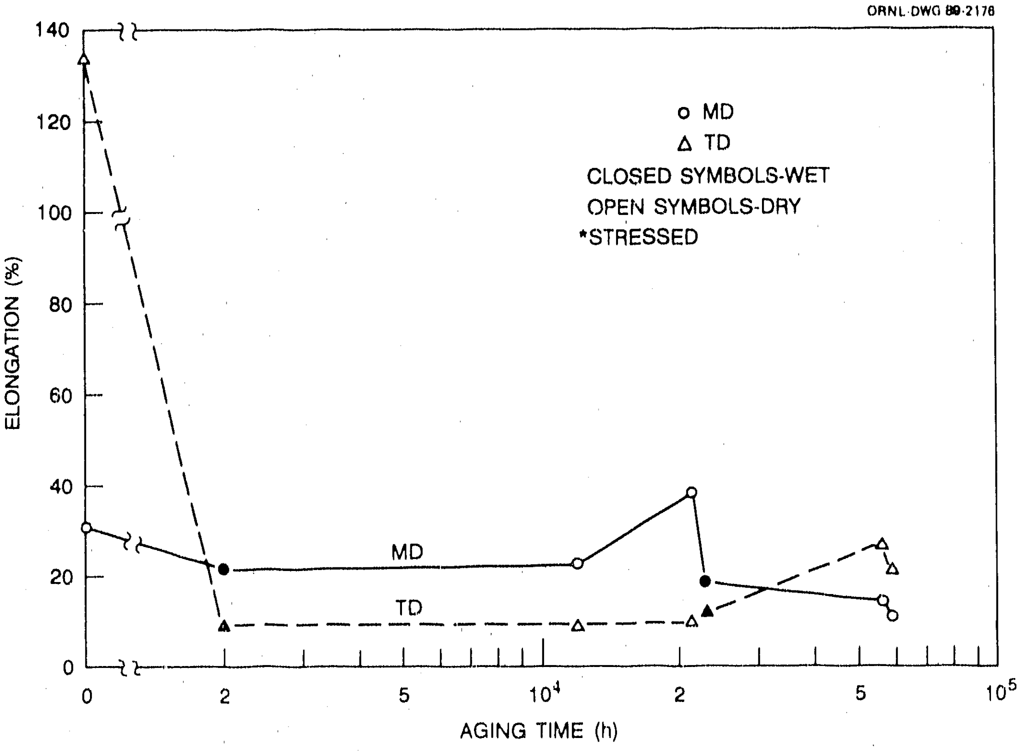

Fig. 13. Fraction elongation of polycarbonate following aging at 160°C.

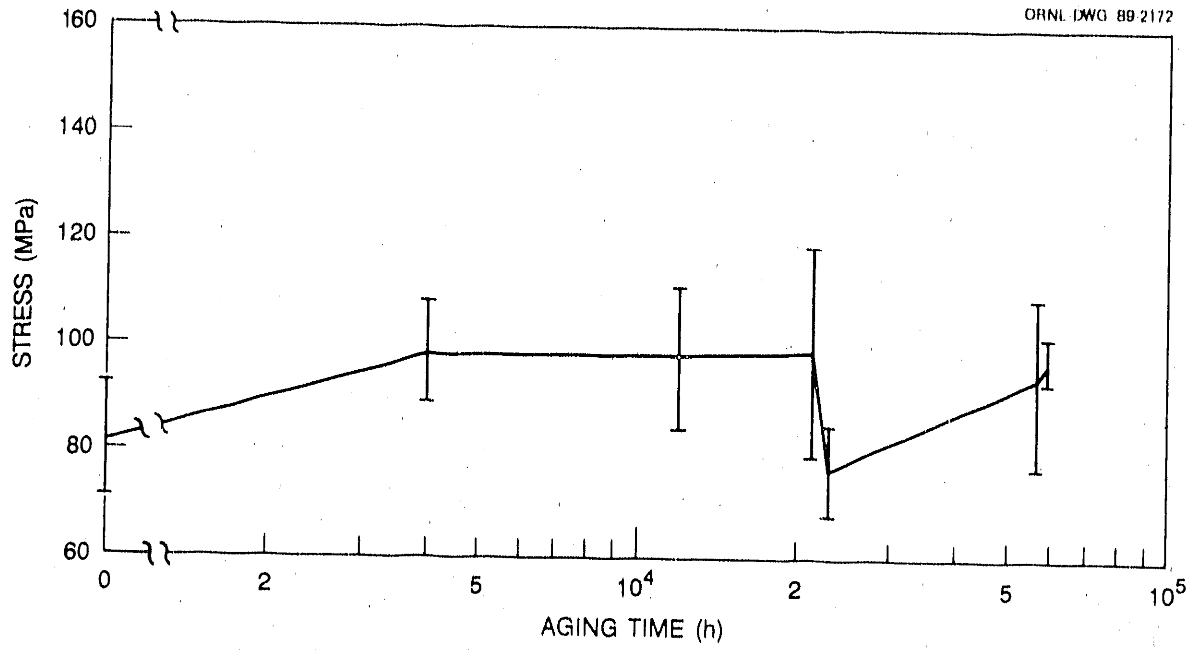

Fig. 14. Influence of aging at 160°C on the tensile properties of polyethersulfone.

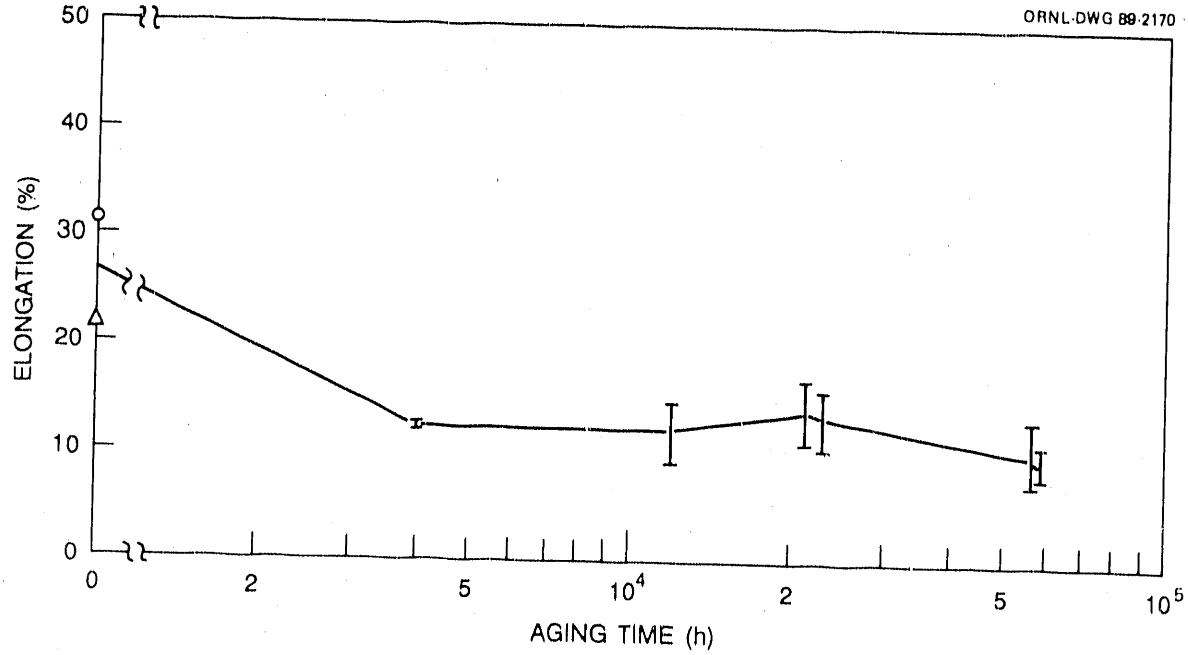

Fig. 15. Fracture elongation of polyethersulfone following aging at 160°C.

Polyimide

The properties of polyimide were not altered appreciably when aged at 135 and 160°C. The results shown in Figs. 16 and 17 for 160°C are typical for 135 and 160°C. The strength of the aged material falls within a range close to that of the as-received material. Similarly, the fracture strain falls close to the values of the as-received material. One group of samples that may be significant are those aged 8500 h in a moist environment. The fracture strains of these samples are slightly lower than those of the other samples.

Polyparabanic Acid

Polyparabanic acid was in limited supply, and samples were aged to 23,000 h at 135 and 160°C. The tensile properties are quite isotropic, but the data scatter makes it difficult to know whether small changes are significant. The fracture elongation of the as-received material is 33%, with a standard deviation of 18%. The general trend seems to be that aging 23,000 h at 135 or 160°C results in a decrease in the yield strength of about 10%, a decrease in the ultimate tensile strength of about 20%, and a decrease in the fracture elongation of about 50%.

Miscellaneous Polymers

Several polymers were subject to a single aging run of 26,417 h, and the results are summarized in Table 3. The ultrahigh-molecular-weight polyethylene (UHMWPE) was aged at 70 and 110°C, and there were no detectable changes in the tensile properties.

A new lot of polyimide (API) was obtained and exposed at 135 and 160°C. The property changes were relatively small. Aging at both temperatures increased the yield strength about 20%, decreased the ultimate tensile strength about 10%, and decreased the fracture strain slightly. The scatter in the test results was quite large (note the large standard deviation).

Two lots of polysulfone (APS and WPS) were aged. The yield and ultimate tensile strengths were increased, but the property change of most concern is the reduction in the fracture strain. Aging at 70°C

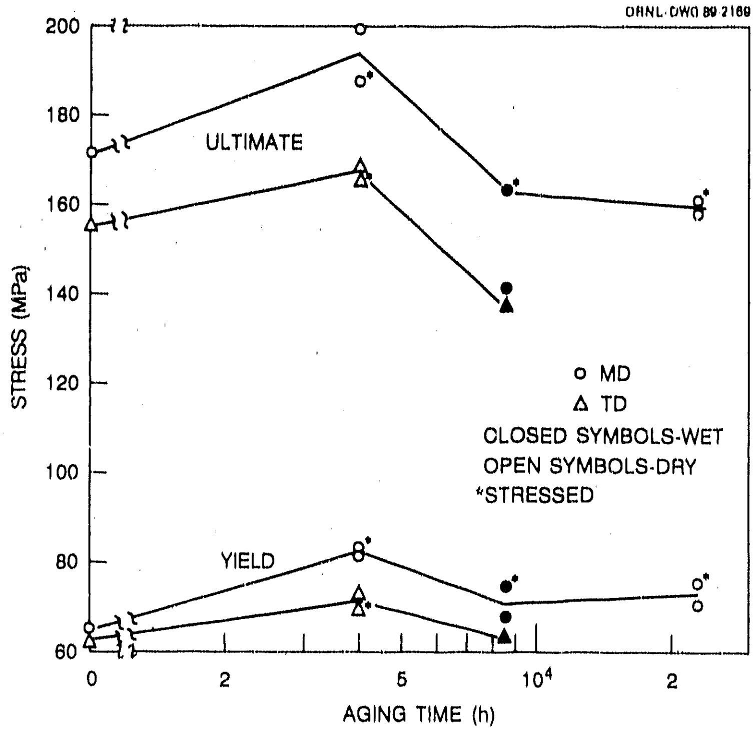

Fig. 16. Effect of aging at 160°C on the tensile properties of polyimide.

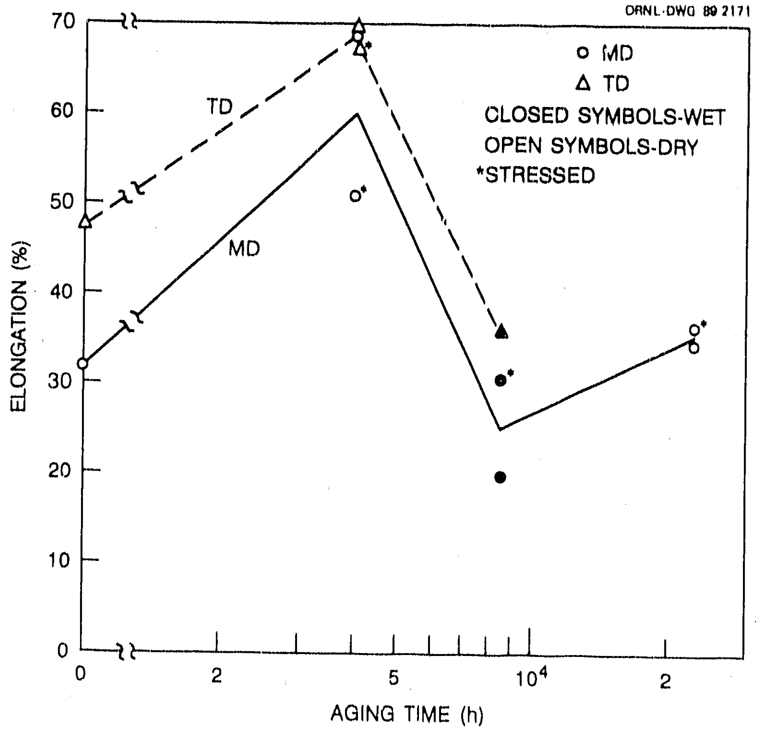

Fig. 17. Fracture elongation of polyimide following aging at 160°C.

Table 3. Influence of aging (26,417 h) on tensile properties of several polymers

Aging temperature (°C)	Orientation	Yield strength (MPa ±1 σ)	Ultimate tensile strength (MPa ±1 σ)	Elongation (% ±1 σ)
		UHMWPE		
NA	MD	14.5 ± 0.7	41.4 ± 5.5	477 ± 65
NA	TD	15.2 ± 0.7	40.0 ± 6.2	486 ± 59
70	MD	15.5 ± 0.9	48.1 ± 7.4	470 ± 63
70	TD	15.9 ± 1.2	51.1 ± 4.3	539 ± 36
110	MD	17.9 ± 0.6	55.2 ± 7.5	443 ± 18
110	TD	18.4 ± 7.0	48.9 ± 6.2	445 ± 51
		API		
NA	MD	60.7 ± 1.4	174.4 ± 22.1	61.6 ± 22.8
NA	TD	62.7 ± 3.4	175.8 ± 15.9	64.4 ± 17.9
135	MD	76.5 ± 2.8	162.7 ± 11.5	43.0 ± 13.3
135	TD	72.4 ± 2.3	151.7 ± 21.4	38.8 ± 25.2
160	MD	77.2 ± 2.2	180.6 ± 4.6	59.6 ± 3.7
160	TD	74.4 ± 3.9	160.6 ± 17.4	43.3 ± 21.2
		APS		
NA	MD	52.4 ± 2.1	60.7 ± 1.4	30.8 ± 22.3
NA	TD	55.2 ± 1.4	60.7 ± 1.4	15.5 ± 5.0
70	MD	62.5 ± 5.1	73.1 ± 1.8	12.3 ± 5.8
70	TD	59.3 ± 2.9	65.5 ± 4.8	8.1 ± 3.0
110	MD	-	74.5 ± 3.5	10.0 ± 3.4
110	TD	-	71.7 ± 2.1	8.9 ± 2.5
135	MD	-	74.5 ± 4.8	7.4 ± 0.9
135	TD	-	73.8 ± 4.0	7.5 ± 1.1

Table 3. (continued)

Aging tempera- ture (°C)	Orientation	Yield strength (MPa ±1 σ)	Ultimate tensile strength (MPa ±1 σ)	Elongation (% ±1 σ)
		WPS		
NA	MD	55.3 ± 1.4	66.9 ± 1.4	29.8 ± 15.2
NA	TD	55.2 ± 2.8	65.5 ± 5.5	97.9 ± 46.6
70	MD	68.3 ± 3.7	77.2 ± 1.3	20.0 ± 11.0
70	TD	68.3 ± 2.8	75.8 ± 0	15.9 ± 5.5
110	MD	77.2 ± 1.5	80.7 ± 2.3	11.8 ± 2.1
110	TD	79.3 ± 1.8	82.0 ± 2.0	9.3 ± 3.2
135	MD	–	84.8 ± 4.2	8.4 ± 0.8
135	TD	–	84.8 ± 3.5	9.3 ± 0.6
		WPEI		
NA	MD	76.6 ± 3.5	96.5 ± 2.1	28.2 ± 11.7
NA	MD	79.3 ± 3.5	96.5 ± 2.1	50.6 ± 28.2
70	MD	56.5 ± 9.2	70.3 ± 14.5	11.5 ± 1.1
70	TD	57.9 ± 3.7	65.5 ± 0.9	16.9 ± 8.2
110	MD	93.8 ± 7.6	106.2 ± 5.2	16.9 ± 9.4
110	TD	93.8 ± 0	106.2 ± 0.9	14.9 ± 5.9
135	MD	95.8 ± 4.1	107.5 ± 6.4	12.3 ± 2.8
135	TD	100.6 ± 1.4	116.5 ± 0.7	16.0 ± 4.3
160	MD	117.2 ± 5.5	131.0 ± 4.1	10.5 ± 0.6
		APEI		
NA	MD	64.1 ± 2.8	73.1 ± 2.1	22.5 ± 11.0
NA	TD	60.7 ± 1.4	69.6 ± 0.7	23.0 ± 9.8

Table 3. (continued)

Aging temperature (°C)	Orientation	Yield strength (MPa ±1 σ)	Ultimate tensile strength (MPa ±1 σ)	Elongation (% ±1 σ)
		APEI (continued)		
70	MD	70.3 ± 0	85.5 ± 1.4	11.8 ± 2.8
70	TD	72.4 ± 4.1	92.4 ± 1.4	10.5 ± 0.6
110	MD	73.1 ± 4.8	82.7 ± 4.9	10.5 ± 1.9
110	TD	77.9 ± 1.4	80.7 ± 5.7	8.6 ± 1.6
135	MD	77.2 ± 1.6	88.9 ± 3.5	9.3 ± 0.5
135	TD	75.1 ± 3.2	79.3 ± 8.3	8.3 ± 1.6
160	MD	92.4 ± 4.1	106.9 ± 2.1	10.0 ± 1.5
160	TD	91.7 ± 2.1	102.0 ± 0.7	10.3 ± 1.8
		WPES		
NA	MD	53.8	71.0	118
NA	TD	55.2	73.8	10.5
70	MD	65.5 ± 1.8	75.8 ± 1.3	38.0 ± 21.0
70	TD	86.9 ± 3.0	100.0 ± 1.3	50.3 ± 33.2
110	MD	61.4 ± 1.7	67.6 ± 0.8	16.8 ± 7.8
110	TD	80.7 ± 6.6	92.4 ± 9.0	25.4 ± 23.6
135	MD	82.0 ± 4.5	91.7 ± 4.0	11.8 ± 3.0
135	TD	83.4 ± 1.3	94.5 ± 1.7	9.0 ± 1.2
160	MD	84.1 ± 3.2	93.8 ± 1.2	12.0 ± 5.6
160	TD	109.6 ± 4.6	122.0 ± 1.0	13.3 ± 5.5

Table 3. (continued)

Aging temperature (°C)	Orientation	Yield strength (MPa ±1 σ)	Ultimate tensile strength (MPa ±1 σ)	Elongation (% ±1 σ)
		APES		
NA	MD	67.9 ± 3.9	77.2 ± 1.4	36.8 ± 9.9
NA	TD	69.6 ± 1.4	75.1 ± 2.1	18.1 ± 8.3
70	MD	54.1 ± 4.1	60.7 ± 4.1	13.6 ± 4.9
70	TD	56.2 ± 0.7	61.8 ± 0.4	48.0 ± 42.7
110	MD	61.2 ± 1.5	68.3 ± 2.9	11.3 ± 2.1
110	TD	63.8 ± 0.7	73.8 ± 0.8	13.5 ± 2.7
135	MD	73.1 ± 1.1	82.0 ± 0.6	19.5 ± 7.4
135	TD	69.6 ± 2.5	76.5 ± 2.6	13.0 ± 8.7
160	MD	78.6 ± 4.8	87.6 ± 2.5	10.0 ± 2.0
160	TD	87.6 ± 2.8	93.8 ± 2.0	10.8 ± 3.9

brought about a significant reduction in fracture strain, and aging at higher temperatures had an even larger effect.

Two lots of polyetherimide (WPEI and APEI) were aged. These two lots had different properties in the as-received condition, and the properties remained different under all aging conditions. The yield and tensile strengths and the fracture elongation of WPEI decreased when aged at 70°C. Aging at 110, 135, and 160°C caused the yield and tensile strengths to increase and the fracture elongation to decrease. The yield and ultimate strengths of APEI increased and the fracture strain decreased with increasing aging temperature.

The two lots of polyethersulfone (WPES and APES) had different properties but responded similarly to aging. Generally, aging increased the yield and ultimate strengths and decreased the fracture elongation. Aging lot WPES at 110°C and lot APES at 70°C caused slight decreases in yield and ultimate tensile strengths.

DENSITY AND ELECTRICAL BREAKDOWN MEASUREMENTS ON AGED SAMPLES

Immersion density measurements were an effective way of determining the transformation from amorphous to crystalline states during aging. Generally there are significant differences in the density of each polymer in the amorphous and crystalline condition. Detailed measurements were made on several polymers following 23,000 h of aging, and these were reported previously.[4] Additional measurements have not been made since that time.

The principal findings from our previous measurements was that all polymers being studied densified slightly when aged at 160°C. Polyethylene terephthalate underwent the largest change. It was 57% crystalline in the as-received condition and 83% crystalline after aging 21,515 h at 160°C.

Electrical breakdown measurements were made of many of the aged samples. Some of the polymers had slight increases in breakdown voltage after aging, but decreases occured only when the polymer was visibly damaged. Under conditions where the polymers stuck together, the polymer sheets were often damaged in trying to pull them apart.

TENSILE PROPERTIES FROM -196 to 200°C

Polymer insulations are needed for a broad range of temperatures. Tensile tests were run on several polymers from -196°C to about 200°C. The properties are tabulated at -196, 24, and 110°C in Tables 4, 5, and 6. At -196°C only one polymer, polyimide (API), had a well-defined yield stress. The other polymers deformed almost linearly with increasing stress and fractured abruptly. API clearly has the most attractive properties at this temperature. Several polymers--including polyethylene terephthalate (PET), polybutylene terephthalate (PBT), and polyethersulfone (PES, APES, and WPES)--have reasonably high fracture stresses but rather low fracture strains. A third group--including polysulfone (WPS, APS), polyetherimide (APEI and WPEI), and UHMWPE--have lower fracture stresses and low fracture strains.

Table 4. Tensile properties of several polymers at -196°C

Polymer	Ultimate tensile strength		Elongation (%)
	MPa	ksi	
PET	261	37.9	7.5
API[a]	272	39.5	18.7
PBT	275	39.9	5.0
WPS	117	16.9	6.0
APS	100	14.5	6.5
APEI	113	16.4	6.5
WPEI	121	17.6	6.5
PES	194	28.2	6.C
APES	134	19.5	5.8
WPES	140	20.3	6.3
UHMWPE	129	18.7	4.3

[a] Yield strength of 106 MPa (15.4 ksi).

Table 5. Tensile properties of several polymers at 24°C

Polymer	Orientation	Yield strength		Ultimate tensile strength		Elongation (%)
		MPa	ksi	MPa	ksi	
PET	MD	82.7	12.0	164.8	23.9	49.0
	TD	84.8	12.3	129.6	18.8	58.1
API	MD	60.7	8.8	174.4	25.3	61.6
	TD	62.7	9.1	175.8	25.5	64.4
PBT	MD	45.5	6.6	51.7	7.5	86.5
	TD	44.8	6.5	49.6	7.2	93.5
WPS	MD	55.8	8.1	66.9	9.7	29.8
	TD	55.1	8.0	65.5	9.5	97.9
APS	MD	53.8	7.8	60.7	8.8	30.8
	TD	55.2	8.0	60.7	8.8	15.5
APEI	MD	64.1	9.3	73.1	10.6	22.5
	TD	60.7	8.8	69.6	10.1	23.0
WPEI	MD	76.5	11.1	96.5	14.0	28.2
	TD	79.3	11.5	96.5	14.0	50.6
PES	MD	80.7	11.7	91.7	13.3	32.8
	TD	71.0	10.3	84.1	12.2	21.9
APES	MD	68.3	9.9	77.2	11.2	36.8
	TD	69.6	10.1	75.1	10.9	18.1
WPES	MD	53.8	7.8	71.0	10.3	118
	TD	55.2	8.0	73.8	10.7	11
UHMWPE	MD	14.5	2.1	41.4	6.0	477
	TD	15.2	2.2	40.0	5.8	486

Table 6. Tensile properties of several polymers at 110°C

Polymer	Orientation	Yield strength		Ultimate tensile strength		Elongation (%)
		MPa	ksi	MPa	ksi	
PET	MD	0	0	95.8	13.9	140
	TD	0	0	102.7	14.9	78
API	MD	52.4	7.6	160.6	23.3	96
	TD	51.0	7.4	155.1	22.5	88
PBT	MD	6.9	1.0	20.7	3.0	250
	TD	9.7	1.4	17.9	2.6	205
WPS	MD	42.1	6.1	47.6	6.9	9.0
	TD	39.3	5.7	46.9	6.8	7.5
APS	MD	31.0	4.5	40.0	5.8	10.0
	TD	25.5	3.7	31.0	4.5	7.0
APEI	MD	25.5	3.7	36.5	5.3	28.5
	TD	28.3	4.1	36.5	5.3	25.0
WPEI	MD	58.6	8.5	64.8	9.4	29.5
	TD	54.5	7.9	60.0	8.7	40.0
PES	MD	57.9	8.4	62.7	9.1	11.0
	TD	46.9	6.8	56.5	8.2	22.5
APES	MD	51.0	7.4	57.9	8.4	36.5
	TD	49.6	7.2	56.5	8.2	44.0
WPES	MD	53.8	7.8	57.2	8.3	16
	TD	48.9	7.1	53.8	7.8	16
UHMWPE	MD	0	0	17.2	2.5	887
	TD	0.7	0.1	19.3	2.8	997

At a test temperature of 24°C all of the polymers tested exhibited sufficient plastic strain to have a yield strength and an ultimate tensile strength (Table 5). The "best" material at 24°C depends on whether yield, ultimate tensile strength, or fracture elongation is the most important characteristic.

At a test temperature of 110°C, all polymers tested are weaker than they were at 24°C. Polyimide (API) retains a larger fraction of its strength at 24°C than the other polymers. Several polymers (e.g., PET, PBT, and UHMWPE) begin to deform at a very low stress. The same polymers have extremely high elongation before fracture.

Polyethylene Terephthalate

Polyethylene terephthalate (PET) was tested at numerous temperatures of over the temperature range of –196 to 200°C. The results for tensile tests are given in Table 7. These samples were all oriented with the MD parallel to the long axis of the sample. Previous

Table 7. Tensile properties of polyethylene terephthalate (PET) oriented in the machine direction

Temperature (°C)	Yield strength[a]		Ultimate tensile strength		Elongation[b] (%)
	psi	MPa	psi	MPa	
185	0	~0	12,700	87.6	250
160	~0	~0	10,600	73.1	75
135	~0	~0	14,200	97.9	114
110	~0	~0	14,900	102.7	78
70	8,200	56.5	22,400	154.4	75
24	12,000	82.7	23,900	164.8	49
0	13,100	90.3	18,000	124.1	45
-10	14,700	101.3	16,700	115.1	11
-20	15,100	104.1	19,600	135.1	46
-30	15,500	106.9	19,200	132.4	18
-40	17,100	117.9	20,400	140.6	13
-50	17,100	117.9	21,800	159.3	18
-60	17,100	117.9	23,700	163.4	23
-70	16,300	112.4	24,300	167.5	13
-75	21,600	148.9	25,100	173.0	17
-80	18,800	129.6	26,700	184.1	26
-90	20,000	137.9	28,000	193.0	20
-100	22,600	155.8	27,800	191.7	11
-196	[c]	[c]	37,900	261.3	7.5

[a] Based on 0.2% offset method.

[b] Measured over 25-mm (1-in.) gage length.

[c] Did not exhibit significant yielding before fracture.

tests on this lot of material showed that the only mechanical property that varied significantly between the MD and TD was the ultimate tensile strength, which at 24°C was 20% weaker in the TD. The yield strength and fracture elongation were equivalent in the two directions. The yield and ultimate tensile strengths are shown as a function of temperature in Fig. 18. The yield strength data were fit by a linear model, and the ultimate tensile strength data were fit by a second-order polynomial. This material flowed (yielded) at very low stresses at temperatures above 100°C, but hardened sufficiently to have a fracture strength of about 100 MPa. At 25°C and lower temperatures, the yield and ultimate tensile strengths became closer. The yield and ultimate tensile strengths both increased with decreasing temperature, with no abrupt steps at any particular temperature.

The fracture elongation is shown as a function of temperature in Fig. 19. The line drawn through the data is a third-order polynomial. The elongation is 10% or less below -100°C, 10 to 20% from -100 to 0°C, and 50% and higher at temperatures above 0°C. There is considerable scatter in the results, but the trends seem well defined.

Polyimide

The results of tensile tests on polyimide over the temperature range of -196 to 270°C are summarized in Table 8. This material continued to display yielding and strain hardening up to the ultimate tensile strength over the entire temperature range. The yield and ultimate tensile strengths are shown in Fig. 20 as a function of test temperature. The lines fit through the data are second-order polynomials. Both strength parameters increased smoothly with decreasing temperature. The test results for the fracture elongation are shown in Fig. 21. Although there considerable data scatter, the fracture elongation increases from about at -196°C to about 100% above 200°C.

Fractography of Polyethylene Terephthalate and Polyimide

The fracture surfaces of several specimens were examined in the scanning electron microscope. Typical electron micrographs of PET are shown in Fig. 22. The microstructure of this material and the tensile properties were anisotropic. Individual grains were elongated with the

ORNL-DWG 90-8103

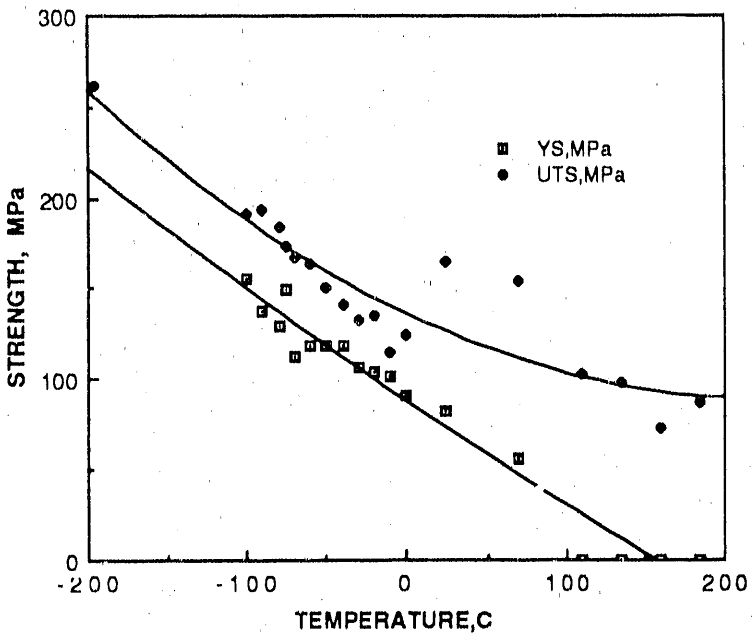

Fig. 18. Yield and tensile strengths of polyethylene terephthalate as a function of temperature.

ORNL-DWG 90-8104

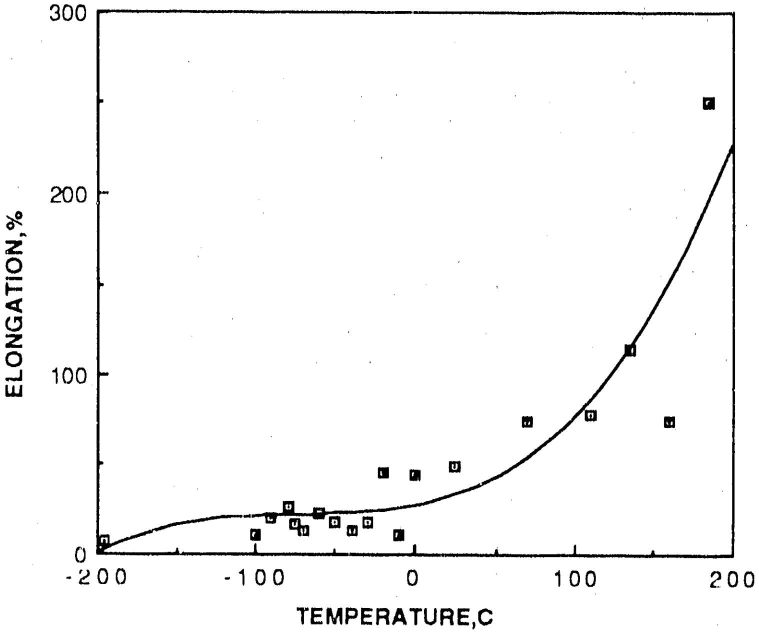

Fig. 19. Fracture elongation of polyethylene terephthalate when subjected to a short-term tensile test at various temperatures.

ORNL-DWG 90-8105

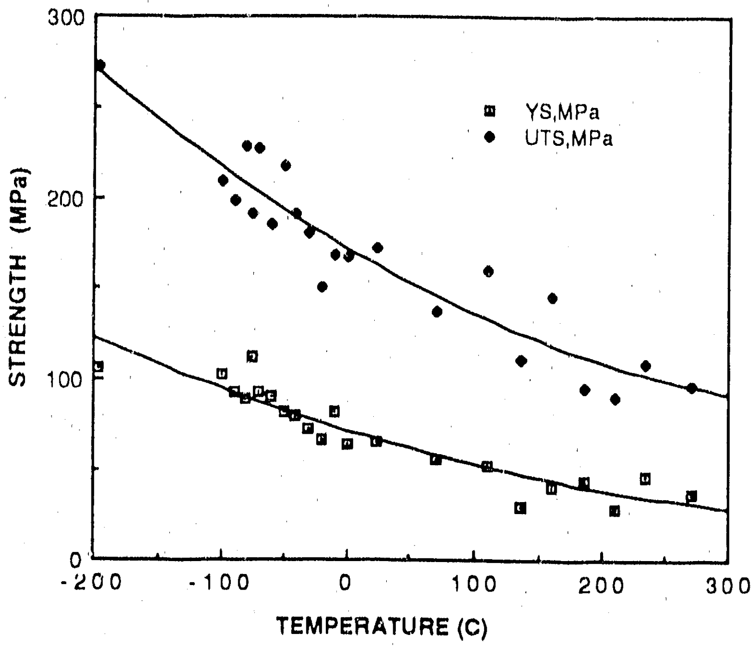

Fig. 20. Yield and ultimate tensile strengths of polyimide at various temperatures.

ORNL-DWG 90-8106

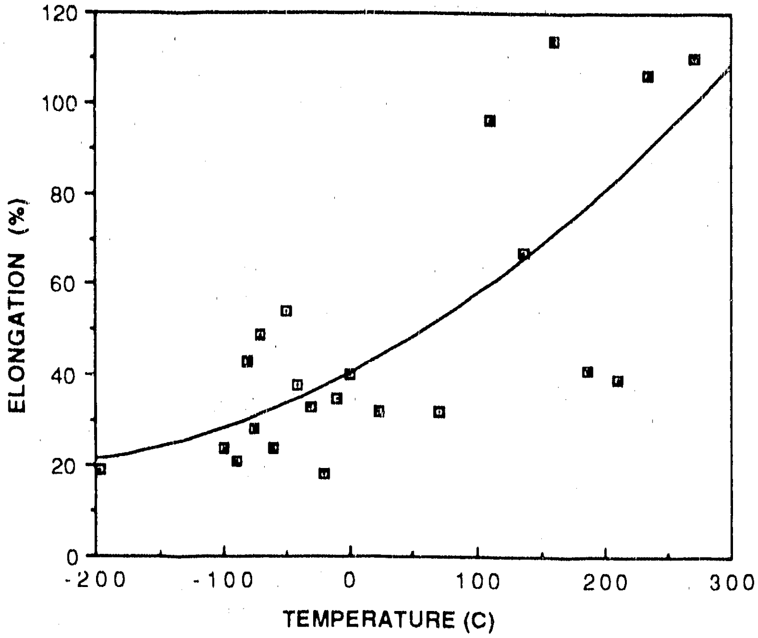

Fig. 21. Fracture elongation of polyimide tensile-tested at various temperatures.

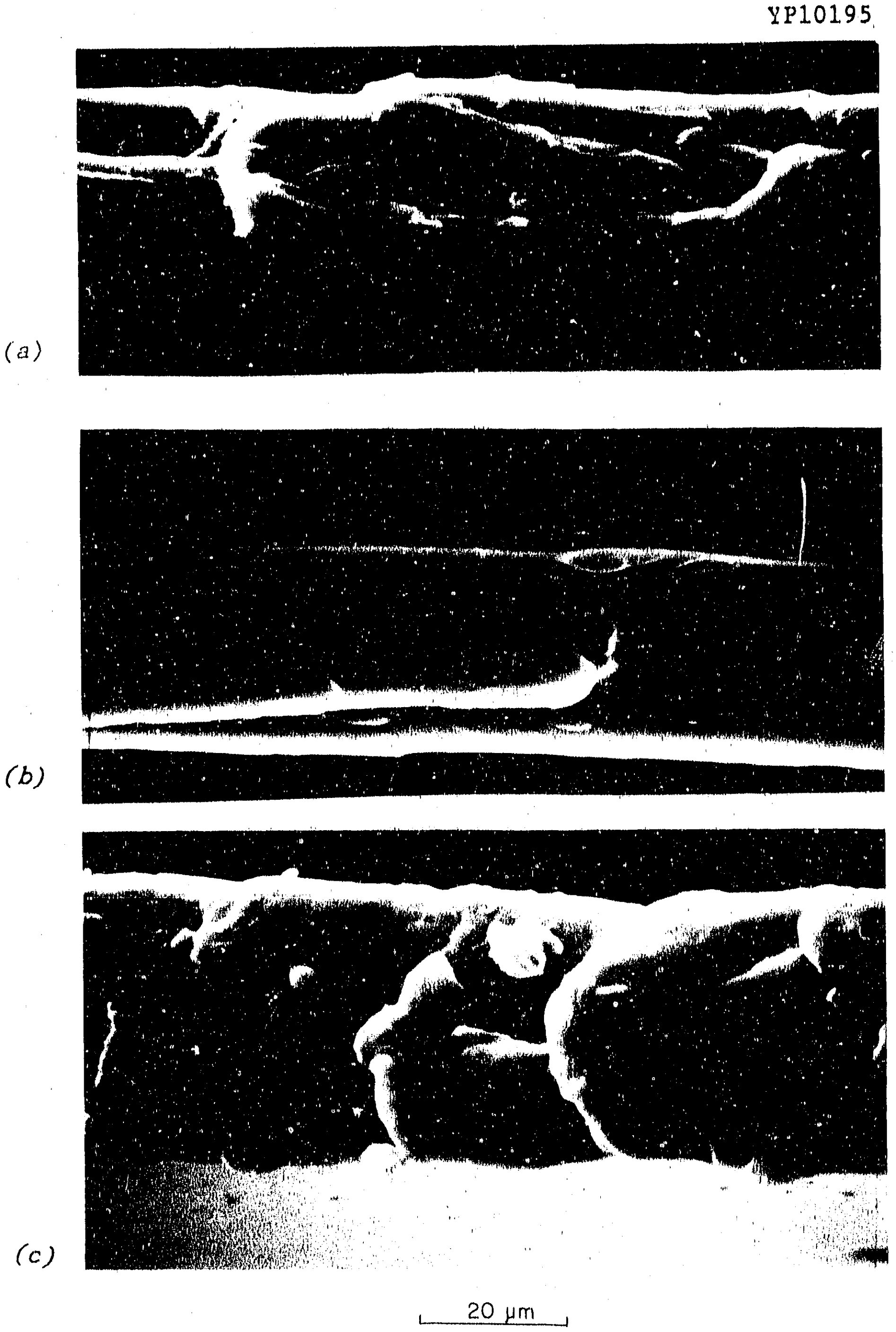

Fig. 22. Fractographs of polyethylene terephthalate tested at (*a*) −196°C, (*b*) 0°C, and (*c*) 110°C. Samples are oriented so that the machine direction comes out of the plane of the page.

Table 8. Tensile properties of polyimide (API) oriented in the machine direction

Temperature (°C)	Yield strength[a]		Ultimate tensile strength		Elongation[b] (%)
	psi	MPa	psi	MPa	
270	5,300	36.5	14,100	97.2	110
235	6,800	46.9	15,800	108.9	106
210	4,100	28.3	13,100	90.3	39
185	6,400	44.1	13,800	95.1	41
160	5,900	40.7	21,200	146.2	114
135	4,300	29.6	16,200	111.7	67
110	7,600	52.4	23,300	160.6	96
70	8,200	56.5	20,000	137.9	32
24	9,500	65.5	24,900	171.7	32
0	9,300	64.1	24,300	167.5	40
-10	12,000	82.7	24,500	168.9	35
-20	9,700	66.9	21,900	151.0	18
-30	10,500	72.4	26,200	180.6	33
-40	11,700	80.7	27,800	191.7	38
-50	12,000	82.7	31,500	217.2	54
-60	13,200	90.0	26,800	184.8	24
-70	13,600	93.8	33,000	227.5	49
-75	16,300	112.4	27,800	191.7	28
-80	13,000	89.6	33,200	228.9	43
-90	13,600	93.8	28,700	197.9	21
-100	14,900	102.7	30,400	209.6	24
-196	15,400	106.2	39,500	272.3	19

[a] Based on 0.2% offset method.

[b] Measured over 25-mm (1 in.) gage length.

longest dimension oriented primarily in the MD. At −196°C, the ductility of the material was low and the grains seemed to pull apart with little plastic deformation [Fig. 22(*a*)]. At 0°C, the material was more ductile and the individual grains deformed sufficiently to make the fracture surface rather smooth [Fig. 22(*b*)]. At 110°C, the material was very ductile and the grains grew under the combined influences of the elevated temperature and strain [Fig. 22(*c*)].

Fractured polyimide samples were also examined. Several electron micrographs of fracture surfaces are shown in Fig. 23. This material was isotropic and appeared to have a very fine grain size. The material had 19% strain at fracture at −196°C. The fracture surface at this

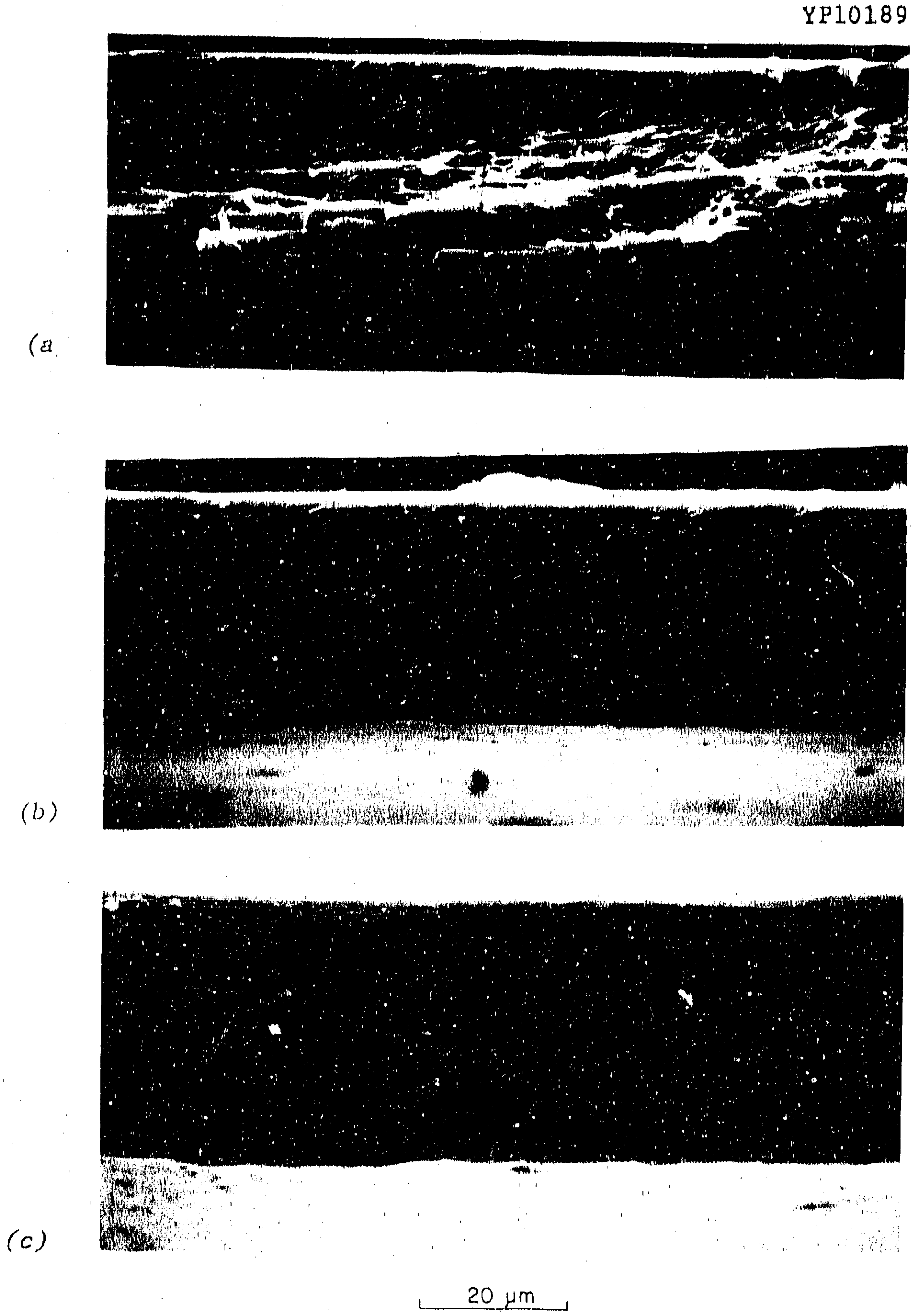

Fig. 23. Scanning electron micrographs of fracture surfaces of polyimide for samples tested at (*a*) −196°C, (*b*) 0°C, and (*c*) 110°C. Samples are oriented so that the machine direction comes out of the page.

temperature [Fig. 23(*a*)] had some areas of cleavage (brittle) fracture, but most of the fracture was ductile tearing. At 0°C [Fig. 23(*b*)], the strain at fracture was higher and the fracture was totally ductile tearing. The fracture strain was even higher at 110°C, but the fracture surface was smoother. There was also a layered appearance to the microstructure [Fig. 23(*c*)].

TENSILE PROPERTIES OF POLYMERS IN OIL

The tensile properties of several polymers were measured in transformer oil (Exxon Univolt N60) at 24 and 90°C. The samples were immersed in the test environment and equilibrated approximately 5 min before being tested. These test results were then compared with those obtained in an air environment. The ratio of the ultimate tensile stress to that in air is compared in Fig. 24 for several polymers tested at 24 and 90°C. The plain horizontal lines on the bars in Fig. 24 represent the 24°C test condition. At a ratio of 1, the tensile strength was the same in air and oil; at a ratio of <1 the polymer was strengthened by the oil; and at a ratio of >1, the polymer was weakened by the oil. At 24°C, all but two of the ratios fell between 0.9 and 1.1, indicating that the oil has very little effect. One lot of polyethersulfone (PES) had a ratio of 1.15 (weakened), but the two other lots of polyethersulfone (APES and WPES) had ratios very near 1. One lot of polyetherimide (APEI) had a ratio of 0.82 (strengthened by the oil), but the other lot (WPEI) had a ratio of 0.98.

The horizontal lines with asterisks in Fig. 24 are for a test temperature of 90°C. None of the materials had a ratio less than 1, so none of the polymers are strengthened by oil at this temperature. Most materials showed weakening effects due to oil of 25% or less. One lot of polyethersulfone (WPES) was weakened 33%, polyethylene terephthalate (PET) was weakened 150%, and one lot of polysulfone (WPS) was weakened 226%.

The ratio of the fracture elongation in air compared with that in oil is given in Fig. 25. The plain horizontal line is for the 24°C tests. A ratio of 1 indicates no effect, a ratio <1 indicates that the elongation is greater in oil than in air, and a ratio >1 indicates that the elongation is less in oil than in air. Three polymers had greater

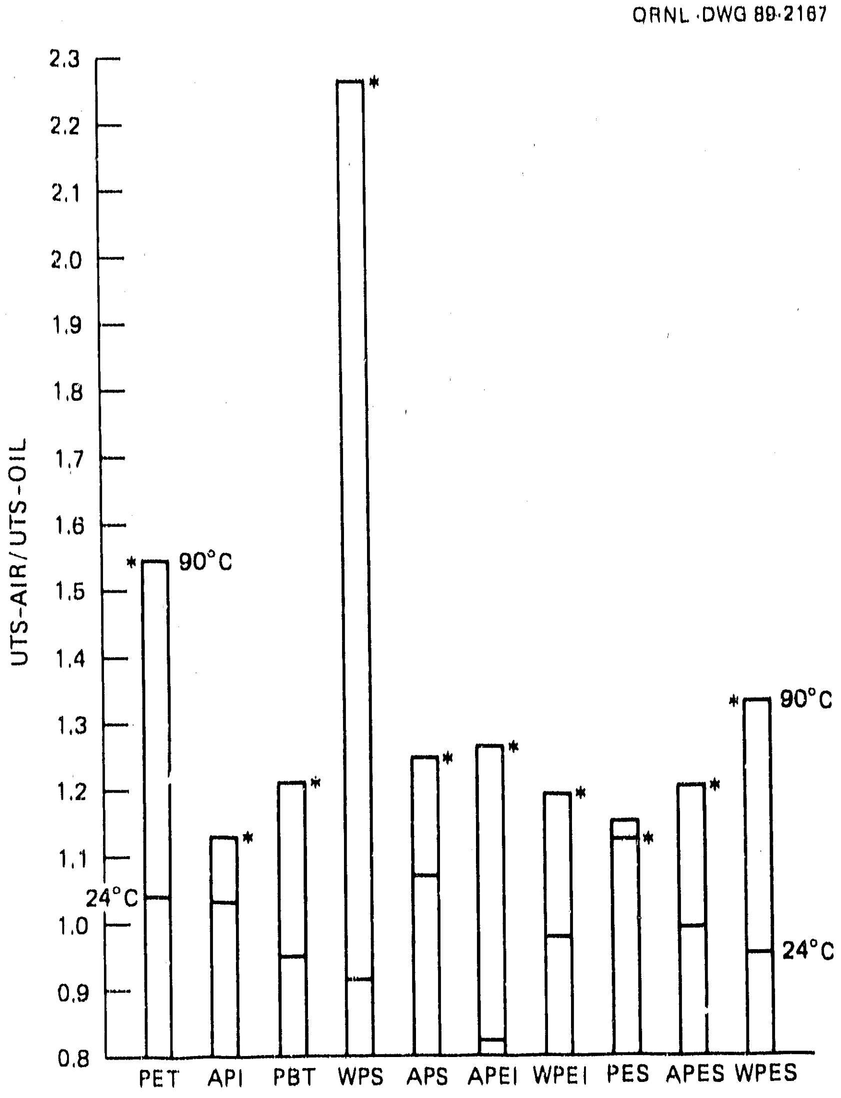

Fig. 24. Comparison of ultimate tensile strength of several polymers in oil and air.

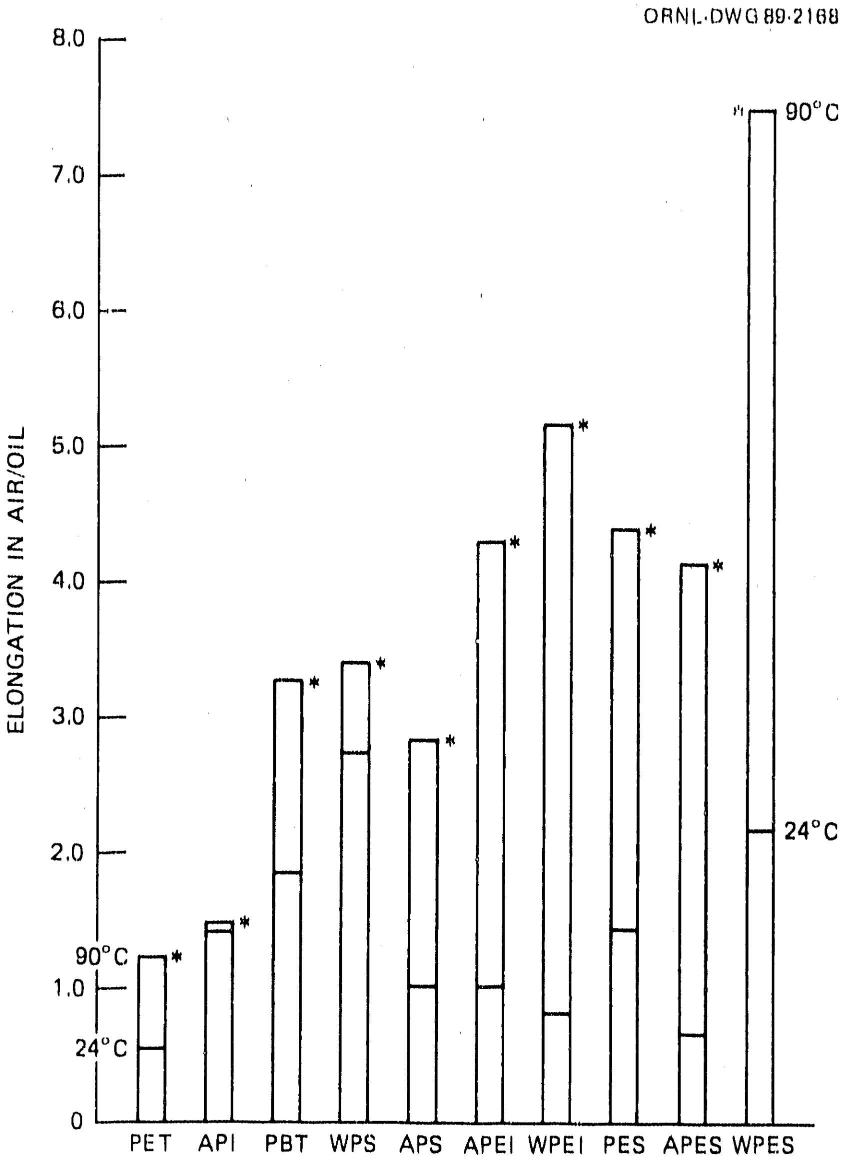

Fig. 25. Comparison of fracture elongation of several polymers in oil and air.

elongation in oil than in air, two were unaffected, and five had lower elongation at fracture in oil than in air.

The results at a test temperature of 90°C are represented by a horizontal line with an asterisk. Under these conditions, the elongation ratio was >1 for all polymers, i.e., the fracture elongation was lower in oil than in air. PET and API were affected the least, and the elongation ratio varied from 3 to 7 for all other polymers. Thus, the fracture strain was less in oil than in air for most polymers.

TENSILE PROPERTIES OF KRAFT PAPER-POLYPROPYLENE LAMINATED PRODUCT

Samples of a product developed for Electric Power Research Institute were provided for evaluation. The material was in the form of strips 25.4 mm (1 in.) wide with Kraft paper on the two outer surfaces and polypropylene film in the center. When the samples were aged in the Dussek fluid, they were easy to delaminate into layers. Thickness measurements indicated that the paper was 0.0254 mm (1 mil) on each side and the polypropylene was 0.0508 mm (2 mils), for a total thickness of 0.1016 mm (4 mils). In the thicker material the paper was 0.0381 mm (1.5 mils) on each side and the polypropylene was 0.0762 mm (3.0 mils), for a total thickness of 0.1524 mm (6 mils).

The strain measuring system being used was not accurate enough to measure the exact fracture strain of the laminate. The paper usually fractured at about 4% strain. Under some conditions (usually at 25°C) the polypropylene broke at the same time. At elevated temperatures and after aging in the Dussek fluid, the paper broke and the propropylene remained intact and stretched several hundred percent before fracturing.

Because the strengths of the two components vary differently with temperature, the fracture loads (listed in Table 9) do not make much sense when converted to stresses. On the basis of the fracture loads, several conclusions seem to be supported by the data:

- The thicker material is stronger than the thinner material.
- The fracture stress of each material decreases slightly with increasing test temperature.
- Aging in the Dussek fluid and then testing at 25°C in air reduces the fracture strength. The reduction in strength due to aging 4343 h at 90°C is about 50%.

Table 9. Ultimate tensile strength of Kraft paper-polypropylene product under various conditions

Thickness (mils)	Test conditions	Average fracture load (lb)
4	25°C, air	8.9
6	25°C, air	12.8
4	70°C, air	6.3
6	70°C, air	12.0
4	90°C, air	6.7
6	90°C, air	9.0
4	110°C, air	6.7
6	110°C, air	8.4
4	25°C, Dussek	8.9
6	25°C, Dussek	13.1
4	100 h; 90°C, Dussek; 25°C, air	8.2
6	100 h; 90°C, Dussek; 25°C, air	11.4
4	1030 h; 90°C, Dussek; 25°C, air	7.3
6	1030 h; 90°C, Dussek; 25°C, air	10.0
4	4343 h; 90°C, Dussek; 25°C, air	4.1
6	4343 h; 90°C, Dussek; 25°C, air	7.3
4	24 h; -196°C, Dussek; 25°C, air	8.7
6	24 h; -196°C, Dussek; 25°C, air	12.8
4	13 h; -78°C, Dussek; 25°C, air	7.9
6	13 h; -78°C, Dussek; 25°C, air	12.3

TENSILE PROPERTIES OF POLYBUTYLENE TEREPHTHALATE

Polybutylene terephthalate (PBT) is a polymer in which there seems to be considerable interest as an insulating material. We evaluated three thicknesses of this material (0.25, 0.38, and 0.51 mm), and the results for an air test environment are summarized in Table 10. The material becomes weaker with increasing thickness. The yield and ultimate tensile strengths are shown as a function of temperature for the 0.25-mm-thick material in Fig. 26. This polymer was very isotropic with almost equivalent properties in the MD and the TD. The strength decreased rapidly with increasing test temperature, so that the strength above 100°C was very small. The fracture strains for these same samples are shown in Fig. 27. The fracture strain for this material was between 75 and 100% at 25°C and increased to several hundred percent with increasing temperature.

Table 10. Tensile properties of polybutylene terephthalate (PBT) in an air environment at temperatures of 24 through 185°C

Thickness		Orientation	Temperature (°C)	Yield strength		Ultimate tensile strength		Elongation (%)
mm	mils			MPa	ksi	MPa	ksi	
0.25	10	MD	24	45.5	6.6	52.4	7.6	87
0.25	10	TD	24	44.8	6.5	49.5	7.2	94
0.38	15	MD	24	43.4	6.3	47.6	6.9	32
0.38	15	TD	24	44.1	6.4	47.6	6.9	102
0.51	20	MD	24	37.9	5.5	42.7	6.2	94
0.51	20	TD	24	35.8	5.2	40.7	5.9	122
0.25	10	MD	70	18.6	2.7	29.0	4.2	225
0.25	10	TD	70	20.0	2.9	29.0	4 2	195
0.38	15	MD	70	17.9	2.6	25.5	3.7	200
0.38	15	TD	70	18.6	2.7	26.2	3.8	180
0.51	20	MD	70	17.2	2.5	20.0	2.9	125
0.51	20	TD	70	15.2	2.2	21.4	3.1	185
0.25	10	MD	90	17.2	2.5	23.4	3.4	220
0.25	10	TD	90	15.2	2.2	25.5	3.7	175
0.38	15	MD	90	14.5	2.1	22.1	3.2	180
0.38	15	TD	90	15.9	2.3	22.1	3.2	140
0.51	20	MD	90	12.4	1.8	18.6	2.7	150
0.51	20	TD	90	13.1	1.9	17.9	2.6	170
0.25	10	MD	110	6.9	1.0	20.7	3.0	250
0.25	10	TD	110	9.7	1.4	17.9	2.6	205
0.38	15	MD	110	9.7	1.4	16.6	2.4	240
0.38	15	TD	110	11.0	1.7	17.9	2.6	170
0.51	20	MD	110	9.0	1.3	12.4	1.8	205
0.51	20	TD	110	7.6	1.1	12.4	1.8	180

Table 10. (continued)

Thickness		Orientation	Temperature (°C)	Yield strength		Ultimate tensile strength		Elongation (%)
mm	mils			MPa	ksi	MPa	ksi	
0.25	10	MD	135	3.4	0.5	11.7	1.7	300
0.25	10	TD	135	2.8	0.4	15.2	2.2	380
0.38	15	MD	135	6.9	1.0	12.4	1.8	260
0.25	10	MD	160	1.4	0.2	7.6	1.1	440
0.25	10	TD	160	1.4	0.2	12.4	1.8	620
0.38	15	MD	160	1.4	0.2	9.0	1.3	380
0.38	15	TD	160	1.4	0.2	11.7	1.7	530
0.51	20	MD	160	0.7	0.1	6.2	0.9	540
0.51	20	TD	160	0.7	0.1	5.5	0.8	380
0.25	10	MD	185	0.7	0.1	2.8	0.4	240
0.25	10	TD	185	0.7	0.1	5.5	0.8	370
0.38	15	MD	185	0.6	0.08	2.8	0.4	230
0.38	15	TD	185	0.6	0.08	4.8	0.7	365
0.51	20	MD	185	0.4	0.06	2.8	0.4	300
0.51	20	TD	185	0.3	0.04	1.4	0.2	270

ORNL-DWG 90-8107

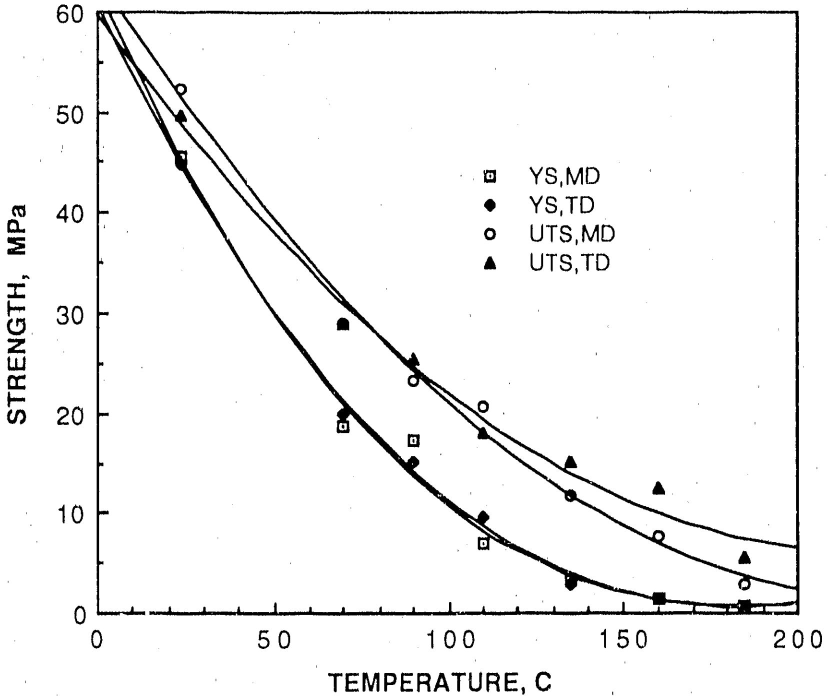

Fig. 26. Yield and ultimate tensile strengths of 0.25-mm-thick polybutylene terephthalate in air at various temperatures.

ORNL-DWG 90-8108

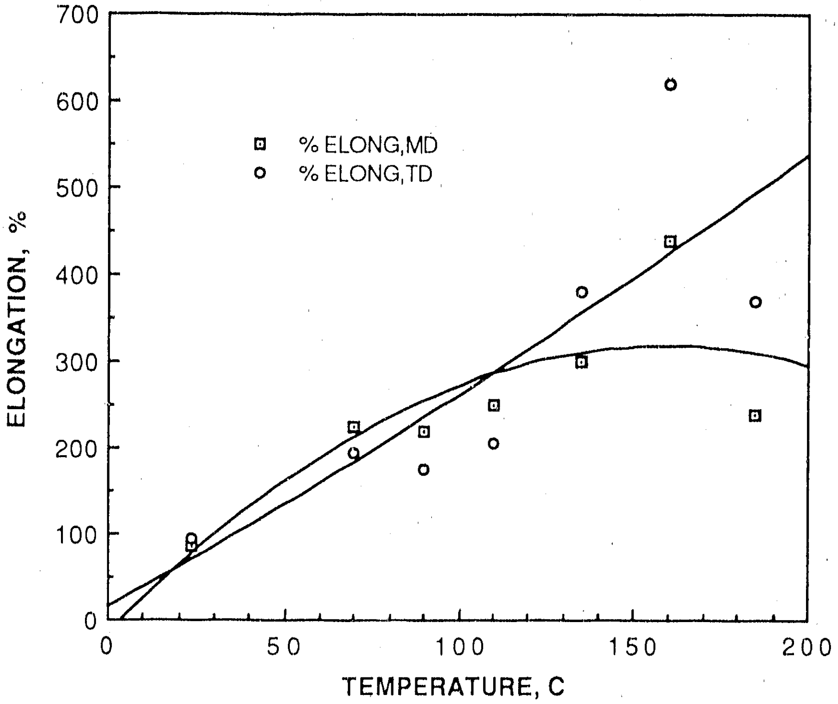

Fig. 27. Elongation of 0.25-mm-thick polybutylene terephthalate when tensile-tested at various temperatures.

A few tensile tests of PBT were run in transformer oil at 24 and 90°C, and these results are summarized in Table 11. At a test temperature of 24°C, the yield and ultimate tensile strengths were about equivalent to those obtained in an air environment (Table 10). At a test temperature of 90°C, the yield and ultimate tensile strengths were 25 to 50% lower in oil (Table 11) than in air (Table 10). At 24°C, the fracture strain in oil was consistently about 30% of that measured in air. The effect of oil on the fracture strain at 90°C was not as clear. At 90°C in air, the fracture strains fell between 150 and 200%. At 90°C in oil, the fracture of three samples was below the ranges for an air test environment, and three samples had fracture strains above the 150 to 200% range for air.

CREEP TESTS IN DRY NITROGEN

The results of creep tests reported previously indicated that, in general, polymers have an initially high creep rate followed by a period of decreasing creep rate.[4] It appears that if the stress is low enough, the creep rate will continue to decrease until it approaches zero. The tests reported this time were run without the complex history of varying test temperature that was used in the previous creep tests.

The creep test results in Table 12 show very similar trends to those noted before. The strain that occurred during the first 50 to 100 h was very dependent upon the stress level. However, the long-term creep rate was of the order of 10^{-4} to 10^{-5} %/h and was not very dependent upon the stress level. Thus, the problem with dimensional stability was primarily a concern during the early hours of loading. There were significant strength differences between the various polymers. Polyimide and polyethersulfone had the highest creep strengths at 90°C.

CREEP TESTS IN OIL

The major shortcoming of the creep test units with an oil environment was that the oil was exposed to air. The oil was changed about once per month, but some darkening due to oxidation was still noted. The experimental equipment is being altered to enclose the test samples in a

Table 11. Tensile properties of polybutylene terephthalate (PBT) in transformer oil

Thickness		Orientation	Temperature (°C)	Yield strength		Ultimate tensile strength		Elongation (%)
mm	mils			MPa	ksi	MPa	ksi	
0.25	10	MD	24	48.3	7.0	52.4	7.6	38
0.25	10	TD	24	49.6	7.2	54.5	7.9	31
0.38	15	MD	24	44.8	6.5	47.6	6.9	12
0.38	15	TD	24	46.9	6.8	49.6	7.2	30
0.51	20	MD	24	41.4	6.0	46.2	6.7	19
0.51	20	TD	24	40.0	5.8	42.7	6.2	22
0.25	10	MD	90	11.0	1.6	18.6	2.7	67
0.25	10	TD	90	14.5	2.1	19.3	2.8	320
0.38	15	MD	90	13.1	1.9	17.2	2.5	34
0.38	15	TD	90	12.4	1.8	18.6	2.7	300
0.51	20	MD	90	7.6	1.1	12.4	1.8	226
0.51	20	TD	90	9.0	1.3	14.5	2.1	70

Table 12. Creep behavior of several polymers in dry nitrogen at 90°C

Material	Stress		Intercept strain (%)	Minimum creep rate (%/h)	Test time (h)	Final strain (%)
	MPa	ksi				
PET, EL, MD	19.4	2.81	2.0	5.5×10^{-5}	28,053	3.5
PET, C, MD	27.6	4.0	8.7	8.5×10^{-5}	22,779	11.0
PET, C, TD	27.6	4.0	4.9	3.6×10^{-5}	22,779	6.0
PET, C, MD	19.2	2.78	2.7	5.0×10^{-5}	28,053	4.0
PET, C, MD	6.9	1.0	0.5	2.0×10^{-4}	22,779	5.0
PI, MD	39.2	5.68	0	8.6×10^{-5}	28,053	2.5
API, MD	39.2	5.68	0.2	1.2×10^{-4}	28,053	3.5
API, MD	27.6	4.0	0	1.5×10^{-5}	28,053	4.0
APES, MD	34.5	5.0	0	1.1×10^{-4}	28,053	3.0
WPES, MD	20.7	3.0	0	1.1×10^{-4}	28,053	3.0
PES, MD	20.7	3.0	0	1.0×10^{-4}	28,053	2.5
APES, MD	20.7	3.0	0	8.9×10^{-5}	28,053	2.5
APS, MD	17.2	2.5	0.12	5.9×10^{-4}	332.5	42
APS, MD	10.3	1.5	0	9.7×10^{-5}	28,053	3.0
WPS, MD	10.3	1.5	0	4.7×10^{-5}	28,053	2.0
PBT, MD	17.2	2.5	12.7	4.8×10^{-5}	22,779	48
UHMWPE, MD	5.2	0.75	6.6	5.6×10^{-3}	2,003	65
APEI, MD	21.4	3.1	0.35	1.8×10^{-3}	671	14
APEI, MD	10.3	1.5	0	1.2×10^{-4}	28,053	3.0
WPEI, MD	10.3	1.5	0	8.9×10^{-5}	28,053	2.0

dry nitrogen environment, but this conversion has not been completed and all of the data reported came from the old equipment.

Limited data from the creep tests in nitrogen and oil are used in Table 13 to show comparative effects of the two environments. Because of the scatter inherent in testing polymers, it is unlikely that enough tests have been run to accurately determine the creep properties in the two environments.

The results available for polyethylene terephthalate, polyimide, and polyetherimide failed to show any influence of environment. Test results for polyethersulfone, polysulfone, polybutylene terephthalate, and ultrahigh-molecular-weight polyethylene show that the creep rupture strength of these materials was lower in oil than in nitrogen. This environmental effect would need to be taken into account if the affected polymers were to be used in an oil environment.

MULTISTRESS AGING

Test Results for Polyethylene Terephthalate

The test conditions for type C film polyethylene terephthalate are summarized in Table 14.[7] The test material was 0.0254 mm (0.001 in.) thick, and all specimens were oriented with the longitudinal axis in the machine direction of the sheet. Duplicate specimens were stressed from 20.7 MPa (3 ksi) to 41.4 MPa (6 ksi). The experimental approach taken with the first set of specimens was to apply the dc potential to all specimens for a time and then run for a time without any applied potential. The strain-time (creep) curves were examined to determine whether the applied potential affected the creep rate. The creep curves for duplicate PET tests at two stress levels are shown in Figs. 28 and 29. The few slope changes in these curves were associated with changes in the test temperature. The applied potential sequence is indicated in Table 14, and in Figs. 28 and 29. There is no detectable change in the creep rate due to the potential. The dc potential applied in these tests was 46,000 V, or about one-half the measured breakdown voltage of the film.

Table 13. Comparison of creep behavior of several polymers in nitrogen and transformer oil at 90°C[a]

Polymer	Environment	Stress		Test time (h)	Rupture	Strain (%)
		MPa	ksi			
Polyetherfulfone	Nitrogen	35	5	28,053	No	3.0
	Oil	35	5	70	Yes	
	Oil	28	4	1,169	No	1.8
Polysulfone	Nitrogen	10	1.5	28,053	No	3.0
	Oil	14	2.0	164	Yes	
	Oil	6.9	1.0	1,152	No	3.2
Polybutylene terephthalate	Nitrogen	17	2.5	48	Yes	
	Oil	14	2.0	0	Yes	
	Oil	6.9	1.0	1,774	Yes	
Ultrahigh-molecular-weight polyethylene	Nitrogen	5.2	0.75	2,003	Yes	
	Oil	5.2	0.75	0.15	Yes	
	Oil	3.4	0.50	486	Yes	
	Oil	2.1	0.30	14,155	No	2.0

[a] Polyethylene terephthalate, polyimide, and polyetherimide displayed no differences in creep behavior in the two environments and are therefore not included in the table.

Table 14. Summary of test information for tests on polyethylene terephthalate[a]

Test	Mechanical stress		Strain when test terminated (%)
	MPa	ksi	
25005	20.7	3	5.1
25009	20.7	3	5.3
25010	27.6	4	8.1
25006	27.6	4	11.1
25007	34.5	5	16.9
25011[b]	34.5	5	11.0
25012	41.4	6	20.2
25008	41.4	6	26.4

[a] Tests were run at 90°C in N61 grade Exxon Univolt transformer. The potential-time cycle in terms of time interval (h) and potential (4000 V dc during "on" cycle and zero during "off" cycle) was as follows:

Time interval	*Potential*
0-1525	On
1525-1870	Off
1870-2200	On
2200-2525	Off
2525-2700	On
2700-3020	Off
3020-3370	On
3370-3690	Off
3690-4050	On

All tests except 25011 were run 4050 h.

[b] Test ruptured at 280 h.

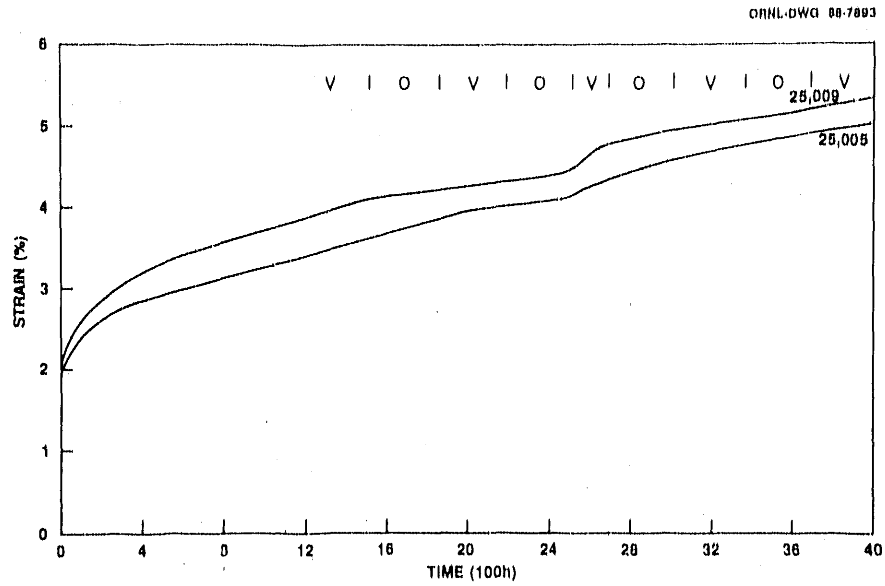

Fig. 28. Stress-time correlations for two samples of polyethylene terephthalate stressed at 20.7 MPa in N61 oil at 90°C. Both samples were exposed to 4000 V dc in the pattern noted on the graph (O = off, V = voltage on). Reference test number to Table 14 for more details.

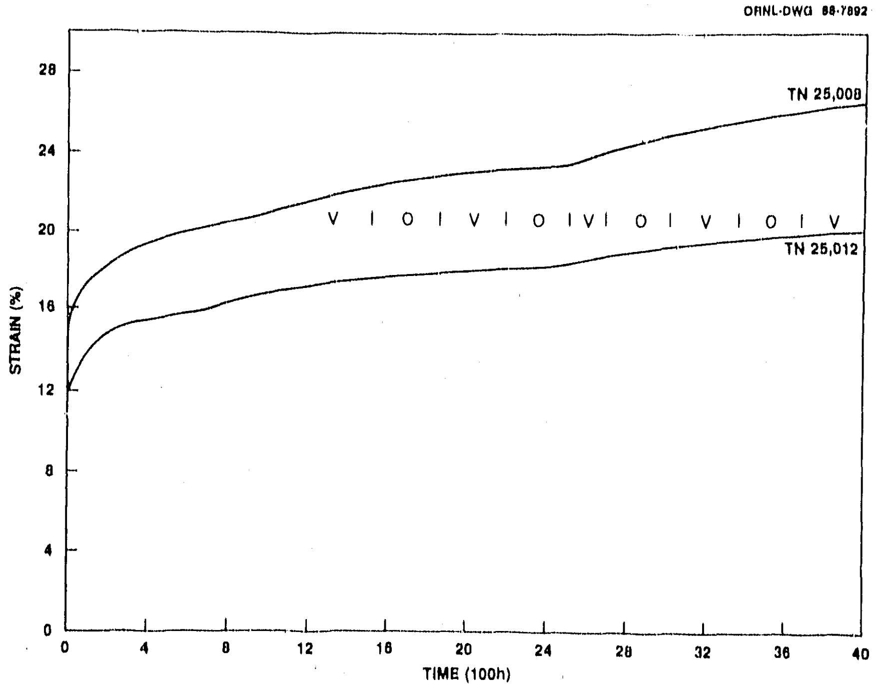

Fig. 29. Stress-time behavior for two samples of polyethylene terephthalate stressed at 41.4 MPa in N61 oil at 90°C. Both samples were exposed to 4000 V dc in the pattern noted on the graph (O = off, V = voltage on). Reference test number to Table 14 for more details.

Post-test Examination of Polyethylene Terephthalate

Following the experiment with PET samples, it was noted that the insulation of one type of high-voltage lead had reacted slightly with the oil. This wire was replaced with wire having a compatible insulation, but the insulation of the first wire had dissolved in the oil and deposited on the samples. The samples were cleaned in alcohol and viewed in a low-power stereo microscope. Pits were present in several specimens. The silver was removed from several samples by immersion in dilute nitric acid, and the samples were again viewed. The pits were mostly associated with the silver strip, but it was not clear whether they would have formed had the wire insulation not contaminated the oil. There was no obvious variation in the concentration of pits between the samples. There is no obvious reason why test 25011 failed in such a short time. The fracture looks normal, and there are no visible flaws. However, the rupture time of this test is anomalous.

Test Results for Polyimide

A second test sequence was completed with polyimide specimens.[7] Type H (Kaptron) polyimide film with a thickness of 0.051 mm (0.002 in.) was used. As shown in Table 15, the samples in this experiment were run in pairs with an electrical potential applied to only one of the two samples at a given stress. Because of some problems with electrical shorts at the grips, the potential on many specimens was interrupted for short periods while repairs were made. The creep curves for two specimens stressed at 41.4 MPa are shown in Fig. 30. The two curves compare very well, showing that the creep behavior was not detectably influenced by the electric potential. The dc electrical breakdown voltage of this material was not measured but is estimated to be at least 10,000 V, and the applied potential was about one-half the breakdown voltage.

Post-test Examination of Polyimide

The silver strip was removed from the samples with dilute nitric acid, and the samples were viewed microscopically. There was no evidence of alteration of the material by the electric potential or the mechanical stress. None of the samples failed, and the strains at the time the tests were terminated varied from 1 to 2%.

Table 15. Summary of test information for polyimide[a]

Test	Mechanical stress		Total test time (h)	Potential applied (V dc)	Time spans potential applied (h)	Strain when test terminated (%)
	MPa	ksi				
25499	20.7	3	140	5000	0-140	1.37
25507	20.7	3	1053	0	0	1.91
25500	27.6	4	1193	5000	0-185	1.52
25501	34.5	5	140	0	0	1.93
25508	34.5	5	1053	5000	0-45 115-1053	1.79
25505	34.5	5	1193	5000	0-115	1.92
25502	41.4	6	1193	5000	0-185 255-1193	2.37
25506	41.4	6	1193	0	0	2.11

[a] Tests were run at 90°C in N61 grade transformer oil.

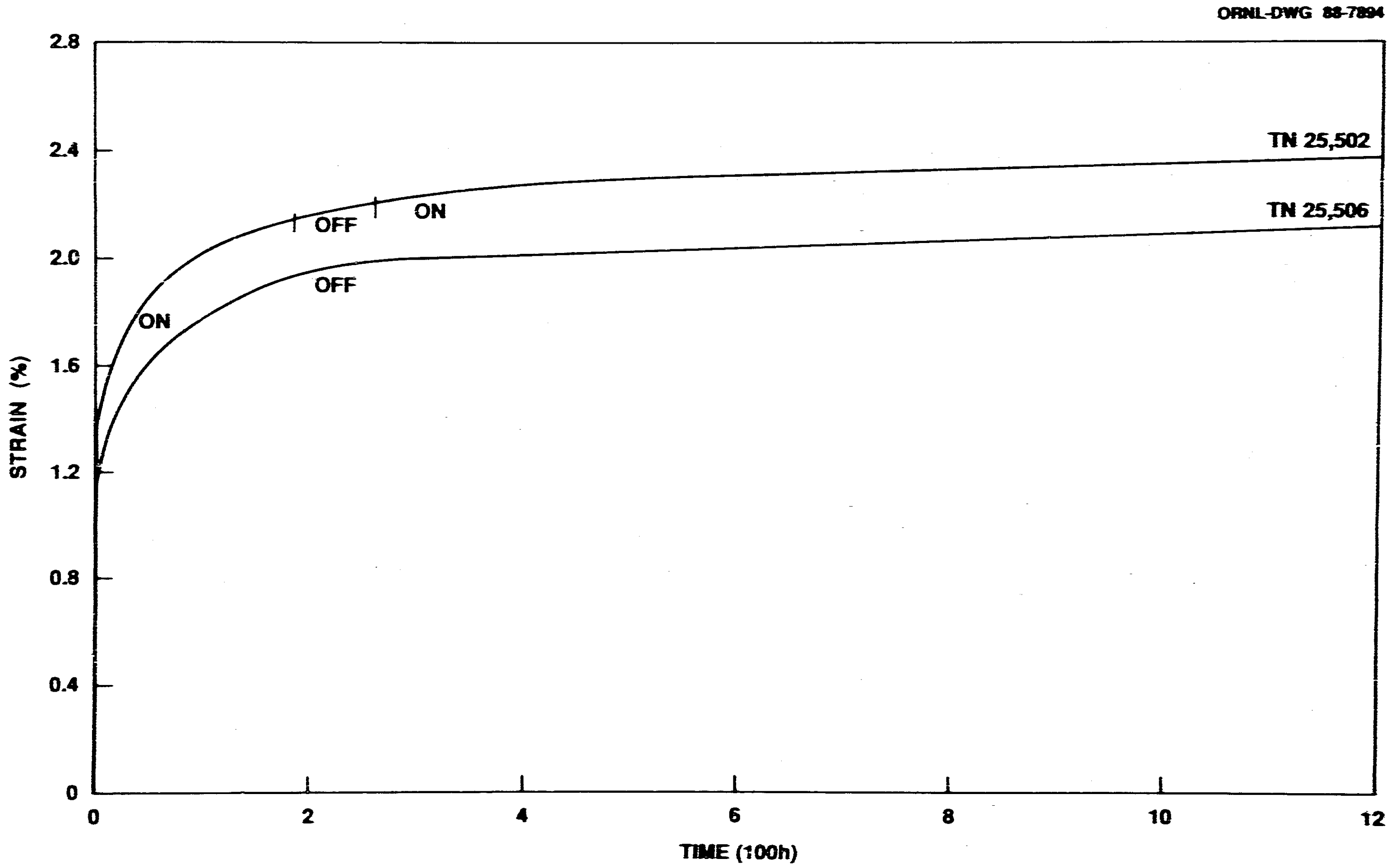

Fig. 30. Strain-time (creep) behavior of two polyimide specimens tested at 41.4 MPa stress and 90°C in N61 oil. For test 25502 the applied potential was 5000 V dc; the times when the potential was on or off are noted on the curves. Test 25506 did not have an applied potential. See Table 15 for more details.

Test Results for Polyethersulfone

A group of tests is in progress on polyethersulfone; the results obtained to date are summarized in Table 16. The considerable scatter in test results for this material is thought to be due largely to surface pits. These were deep compared to the thickness of the material and often occurred in clusters. The last four tests in Table 16 indicate that applied voltage probably does not have a large effect on the rupture life. The results of the tests at higher stresses are not conclusive. These tests are continuing.

DISCUSSION OF RESULTS

Polymer films have excellent insulating properties. The program described in this report is concerned with determining the properties of several polymers so that these materials can be evaluated for engineering applications. Test samples were thermally aged at 70, 110, 135, and 160°C for times to 60,000 h. As these samples were removed from the aging fixture, the tendency for the films to stick together could be evaluated. The sticking temperatures noted in Table 2 indicate that polyimide, polyethersulfone, polyetherimide, and polyparabanic acid were resistant to sticking at 160°C. The other polymers evaluated had lower sticking temperatures. Sticking would be of particular importance where materials of differing thermal expansion bonded during service and were later thermal cycled.

Most of the polymers were tested for tensile properties following thermal aging. These tests revealed changes in strength and fracture strain. The mechanical properties of polyethylene terephthalate and polycarbonate were altered significantly by aging over the range of 70 to 160°C. However, these polymers are likely to be useful for some applications over the lower part of that temperature range. Polyimide was found to be affected little, if any, over the temperature range of 70 to 160°C. The strength of polyethersulfone, polyparabanic acid, and polyetherimide either remained constant with aging or increased slightly; however, the fracture strain of all three materials decreased about 50%

Table 16. Summary of test information for polyethersulfone in N61 grade transformer oil at 90°C

Test	Mechanical stress		Potential applied (kV dc)	Time (h)	Rupture
	MPa	ksi			
25702	34.5	5.0	5	9263.7	Yes
25702	34.5	5.0	5	0.7	Yes
25700	34.5	5.0	5	0	Yes
25704	34.5	5.0		150	Yes
25706	34.5	5.0		120	Yes
25705	27.6	4.0		9263	No
25699	27.6	4.0		35	Yes
25701	27.6	4.0	5	815	Yes
25703	27.6	4.0	5	150	Yes
25726	20.7	3.0	5	32.3	Yes
25712	20.7	3.0	5	120	Yes
25711	20.7	3.0		8139	No
25957	20.7	3.0		52.3	Yes
25958	13.8	2.0	5	6096	No
25959	13.8	2.0		6096	No
25960	10.3	1.5	5	6096	No
26205	10.3	1.5		6096	No

during aging. Polysulfone was less heat-resistant than the other materials, with a significant decrease in strength and fracture strain at all aging temperatures.

Density and electrical breakdown measurements were made on many of the samples aged about 23,000 h. The main purpose of the density measurement was to determine any change in the amount of crystalline material in a given polymer. Most polymers have appreciably different densities in their crystalline and amorphous forms. Polyethylene terephthalate showed the largest change in crystallinity, increasing from 57 to 83% in crystallinity during aging at 160°C. The other polymers underwent very small increases in density during aging.

Electrical breakdown measurements were made on all of the polymer films in the as-received condition. Small changes were often noted during aging, but the changes were insignificant until visible physical damage occurred. This damage was usually in the form of small tears that occurred in separating polymer sheets that were partially stuck together.

Many applications call for insulators at temperatures above or below 25°C. For this reason, tensile tests were run from −196 to 200°C for polyimide (PI) and polyethylene terephthalate (PET). PI exhibited a yield stress at −196°C and had good fracture elongation. PET was brittle and exhibited only a fracture stress. The tensile properties of several polymers were measured at −196, 24, and 110°C. They exhibited a variety of properties, and the selection of a polymer for a given application may very well depend upon the service temperature.

Two new products were evaluated. One was a laminate of two layers of Kraft paper and one layer of polypropylene. The product exhibited some decrease in strength with increasing temperature, most likely due to the decrease in strength of the polypropylene. This material might be used immersed in Dussek fluid (an organic), so some samples were aged 4343 h at 90°C in this fluid. The fracture strength decreased about 50%, and the product was easily delaminated.

The other new product evaluated was polybutylene terephthalate (PBT) film. The material was not very heat-resistant, with very low tensile strength above 100°C. At 90°C, PBT had yield and tensile strengths 25 to 50% lower in oil than in air. The data on fracture strain were too erratic to be conclusive.

Creep tests were run in dry nitrogen and transformer oil at 90°C. The general creep response of polymers involved an initial period of high creep rates and then continuously decreasing creep rate for many hours to a rate near zero. The initial creep period was very stress-dependent, but the latter period was of the order of 10^{-4} to 10^{-5} %/h and not very stress-dependent. The creep properties of polyethylene terephthalate, polyimide, and polyetherimide were not detectably dependent on whether the test environment was transformer oil or nitrogen. Test results for polyethersulfone, polysulfone, polybutylene terephthalate, and ultrahigh-molecular-weight polyethylene showed that the creep strength of these materials was lower in transformer oil than in nitrogen.

Equipment and experimental techniques have been developed for evaluating the influence of an applied dc potential on the deformation of polymer film samples while they are heated and under mechanical stress. The system currently has two known shortcomings. First, the transformer oil in contact with the samples is exposed to air and is subject to oxidation and contamination with moisture. This will be corrected in the future by enclosing the system in dry nitrogen. Second, the potential is applied to only 20% of the area of the test section of each sample. Applying the potential to more of the gage section causes shorting as a result of surface conduction. If there are only small effects of an applied potential on creep behavior, they might be masked in this system. Post-test examination of the sample with the silver strip removed would reveal whether any structural damage resulted from the potential even though creep effects might not have been noted. Thus, this technique is felt to be rather sensitive even though the electrical potential is applied to only 20% of the gage area.

Polyethylene terephthalate (type C) and polyimide (type H) films (both manufactured by Du Pont) were tested in separate experiments. The tests were conducted at 90°C, a dc potential of either zero or about one-half the breakdown voltage, and a range of mechanical stresses. There was no evidence in either experiment that the strain-time (creep) behavior was altered by the presence of an applied potential. Some pits were formed in the polyethylene terephthalate specimens, but these are thought to be associated with the contamination from the wire insulation that partially dissolved in the oil. This will be tested in a future experiment.

Both of the materials examined were previously creep tested without an applied potential in dry nitrogen over the temperature range of 110 to 161°C.[4] The results from the present experiments at 90°C seem consistent with the previous results. The general behavior of an initially high creep rate that decelerates during the entire test seems to be characteristic of these materials, as this behavior was noted in the previous experiments.

SUMMARY

Several polymer films were thermally aged up to 60,000 h at 70, 110, 135, and 160°C. Polyimide, polyethersulfone, polyetherimide, and polyparabanic acid were heat-resistant enough not to stick together at 160°C, but the other polymers stuck at lower temperatures. Tensile tests run on the aged polymers indicated that the properties of polyethylene terephthalate and polycarbonate were altered significantly by aging. The tensile properties of polyimide were affected little, if any, by aging. The strength of polyethersulfone, parabanic acid, and polyetherimide either remained constant or increased about 50%. Polysulfone had significant decreases in strength and fracture strain due to aging at any of the temperatures over the range of 70 to 160°C. Electrical breakdown was not affected by aging until the polymer had undergone visible physical deterioration.

Several polymers were evaluated in tensile tests over the range of −196 to 200°C. Polyimide had superior properties at both extremes, but many of the other polymers had good properties over narrower temperature ranges.

The creep behavior of polymers was characterized by a period of high initial creep and an extended period when the creep rate diminished to 10^{-4} to 10^{-5} %/h. The first period of creep was very stress-dependent, but the slower creep region was not. The creep strength at a given temperature varied considerably among the polymers. The creep and tensile strengths of some polymers were lower in transformer oil at 90°C than in dry nitrogen.

Two new products were evaluated. A laminated product having two layers of Kraft paper and one layer of polypropylene was found to decrease in yield and tensile strength when aged in an organic fluid at 90°C. A polymer film, polybutylene terephthalate, tested over a range of temperatures, was found to not be very heat-resistant and to have a very low tensile strength at 100°C. The tensile strength was less in transformer oil than in air.

A piece of equipment was developed in which samples could be exposed to mechanical, thermal, and electrical stresses. Tests thus far in transformer oil at 90°C fail to show any increase in the creep rate when a dc potential is present. The materials tested were polyethylene terephthalate, polyimide, and polyethersulfone.

ACKNOWLEDGMENTS

The author gratefully acknowledges the technical assistance of J. E. Spruiell of the University of Tennessee and R. S. Crouse and J. T. Hutton of Oak Ridge National Laboratory. Able technician support was provided by B. McNabb, Jr., T. J. Henson, and T. P. Kirkland. Financial support for the program was provided by the U.S. Department of Energy, Division of Electric Energy Systems. Local technical direction was given by P. A. Gnadt, S. J. Dale, and C. R. Brinkman. The author is grateful to D. R. James and T. G. Kollie for reviewing the manuscript. The manuscript draft was typed by B. J. Russell, and the final manuscript was prepared by G. R. Carter.

REFERENCES

1. *Fourth Annual Program Information Notice*, DOE/RA-0054, U.S. Department of Energy, Office of Electrical Energy Systems, September 1980.

2. *Compact Gas Insulated Transformer Project*, DOE/ET/29071 (to be published).

3. H. E. McCoy, Jr., *Evaluation of Polymers for Transformer Applications*, ORNL/TM-8398, Oak Ridge National Laboratorys, October 1982.

4. H. E. McCoy, Jr., *Evaluation of Polymer Films for Electrical Insulation*, ORNL-6134, Oak Ridge National Laboratory, April 1985.

5. H. E. McCoy, Jr., and C. R. Brinkman, "Evaluation of Several Polymer Films for Use as Electrical Insulators," pp. 70-85 in *Proceedings of the 17th Electrical/Electronics Insulation Conference*, Sept. 30-Oct. 3, 1985, (PUBLISHER, CITY, STATE, YEAR)

6. H. E. McCoy, Jr., and C. R. Brinkman, "Property Changes in Polymer Films Exposed to Transformer Environments," pp. 505-517 in *Environmental Degradation of Engineering Materials in Aggressive Environments*, proceedings of Second International Conference on Environmental Degradation of Engineering Materials, Sept. 21-23, 1981, Virginia Polytechnic Institute, Blacksburg, Va., ed. M. R. Louthan, Jr., R. P. McNutt, and R. D. Sisson, Jr., (PUBLISHER, CITY, STATE, YEAR)

7. C. R. Brinkman and H. E. McCoy, Jr., "Multistress Aging of Polymer Films," conference record of the 1988 IEEE International Symposium on Electrical Insulation, June 5-8, 1988, p. 192.

END

DATE FILMED

11 / 08 / 90