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J. F. Eichelberger

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These reports are not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of MLM topical reports.

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Hugh Kinser
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PLASTICS RESEARCH

Work is being directed at the development of methods of producing plastics and adhesives having characteristics superior to those currently being used at Mound Laboratory and elsewhere. Variations in plastic formulations and molding procedures are being studied in an effort to produce plastic which consistently meet specifications.

ADHESIVES Work performed on six new formulations shows the effect obtained when diols and triols are used to cure Adiprene * L-213. The diols-Adiprene L-213 systems have longer pot-lives and the films produced are more elastomeric than triol systems. The diols and triols are of relatively low molecular weight and only a small amount supplies the necessary hydroxyl equivalent for cure. Hence, the systems are very viscous and present difficulties in degassing operations. The films obtained in all formulations except AD136 were characterized by the presence of bubbles formed after film casting was complete. AD136 was free of bubbles because it was necessary to heat the AD L-213 1, 1, 1 trimethylol propane mixture to get complete mixing and solution. The hot mixture degassed easily. All other formulations tended to foam during degassing operations, then their viscosities increased and tended to prevent gas removal.

As mentioned in last month's report, a survey of all previous work on polyurethanes is being made. In the course of this re-evaluation of data, additional information of polyol cured polyurethane systems has been obtained and allowed the establishment of approximate molecular weight data for Adiprenes L-100, L-167, and L-213. Also hydroxyl equivalents of all available polyols have been calculated.

Formulation	AD 132	AD 133	AD 134	AD 135	AD 136	AD 137
AD L-213	25 g	25 g	25 g	25 g	25 g	25 g
1,4 BDO[1]	2.0g	3.0g	--	--	--	1.0g
1,5 PDO[2]	--	--	3.0g	--	--	--
1,2,6 HTO[3]	--	--	--	3.0g	--	--
1,1,1 TMP[4]	--	--	--	--	3.0g	2.0g
Pot Life (hours)	1 ½	1	1	¼	¼	2+
Cure Time (days)	3	3	2	2	2	1
Film characteristics	Foamy	Foamy	Foamy	Foamy	Clear	Foamy
Plastic qualities	tough elastic	soft semi tacky elastic	tough elastic	tough elastic	tough semi elastic	tough elastic

[1] Butanediol

[2] Pentanediol

[3] Hexanetriol

[4] 1,1,1 trimethylpropane

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* Trademark of E. I. duPont de Nemours

Processes are being developed for separating and purifying radioelements and potential sources of supply are being evaluated.

IONIUM PROJECT A survey is being made of potential sources of kilogram quantities of ionium (thorium-230) in the United States. Preliminary examination of samples from uranium mills and processing plants has indicated several promising sources. These materials are being examined further.

Four of the solid samples were found to have ionium concentrations of one part per million or greater. It was reported last month that, when one of these samples (24C) was digested with perchloric acid, approximately three times as much ionium was recovered as when the sample was digested with nitric acid alone. The remaining three samples have now been re-analyzed after preliminary perchloric acid digestions. The results are shown in Table 1.

Table 1

ANALYSIS OF SOLID SAMPLES FOR IONIUM

Sample	Ionium Found (ppm) by HNO_3 Digestion	Ionium Found (ppm) by Perchloric Acid Digestion
24C	2.3	7.0
18E	2.8	3.3
24AR	1.1	1.1
28A (Airport cake)	3.7	3.8

The digestion procedure was modified as follows:

> Weigh 2-5g of sample into a 50-ml centrifuge tube. Leach the solid with 20-ml of 3 N HCl, stirring for several minutes to insure adequate contact of the phases. Centrifuge, and pour off the supernate to a storage bottle. Repeat the HCl leach, and retain the solution. To the residue add 5-ml of 10 M NaOH and mix. Dilute the slurry to 20-ml, warm on a water bath, centrifuge, and discard the supernate. Repeat the NaOH leach and discard the solution. Leach the residue with 20-ml of 3 N HCl. Stir, centrifuge, and retain the supernate. Transfer the residue, as a slurry in 5-ml of concentrated HNO_3 and 10-ml of 70% $HClO_4$. Evaporate the slurry to perchloric acid fumes, and continue heating for one hour. After cooling, transfer the slurry to a 50-ml centrifuge tube with 10-ml of water. Centrifuge and transfer the supernate to the storage bottle. Wash the insoluble residue with 20-ml of water, and add the wash to the storage bottle. Dilute the solution in the storage bottle to a convenient volume. Analyze an aliquot of the solution by the tributyl phosphate-cerium fluoride procedure previously described.

It seems likely that the relative insolubility of sample 24C is the result of calcining. The other samples, which were not calcined, showed no significant increase in ionium recovery due to the more vigorous digestion procedure.

Isotopic ratios are not yet available. Samples have been sent to Oak Ridge National Laboratory for determination of total thorium by neutron activation analysis.

HALF-LIFE OF RADIUM-223 The decay of a specially purified radium-223 sample has been followed by alpha counting for 109 days. Preliminary results for the first 83 days were reported last month. The half-life has been recalculated to include the new data, and the results are again reported as a function of the resolution time of the gas flow proportional counter (Table 2).

Table 2

HALF-LIFE OF RADIUM-223 AS A FUNCTION OF COUNTER RESOLVING TIME (T) (109 DAYS - 37 OBSERVATIONS)

T (Min. x 10^7)	Half-life (Days)	Probable Error (Days)
1.50	11.4782	0.0054
1.60	11.4519	0.0059
1.65	11.4387	0.0064
1.7	11.4250	0.0070

It will be noted that the data in Table 2 do not converge to a minimum value of the probable error (within the range of resolution times selected). A plot of resolution time against probable error indicates that the minimum probable error would occur at a substantially lower resolution time than any which have been previously found. However, examination of the deviations from the least squares determinations indicates strongly that the resolution time lies within the range shown. It seems probable, therefore, that excessive weight is being given to the later observations, where the influence of a long-lived impurity would be greatest.

The counting rate of the sample has not decreased to less than 0.2 per cent of the initial counting rate. At the present rate (approximately 3000 counts per minute), the coincidence correction due to the resolution time of the counter is negligible. On the other hand, the influence of any trace impurity, such as actinium-227 or radium-226, is greater. While it is unlikely that any significant amount of actinium-227 remains in the sample, the separation procedure did not allow for the possibility that radium-226 may be present. Counting of the sample will be continued until the presence or absence of long-lived impurities can be established.

A composite sample, estimated to contain approximately 130 microcuries of actinium-227, has been prepared from residues and tailings accumulated during the FY-1952 processing of actinium. The actinium, probably not in equilibrium with its decay products, was purified of iron by anion exchange. However, upon evaporation to dryness, the sample was found to contain a substantial amount of non-radioactive impurities of unknown nature and origin.

When the sample was evaporated in concentrated nitric acid, a white precipitate developed, which was readily soluble in distilled water. This behavior is characteristic of barium nitrate. The precipitate, which was estimated to represent approximately 100 micrograms of barium, carried the bulk of the radium-223.

The remaining impurities were largely soluble in ammonium hydroxide. Upon filtration of the hydroxide, a small brown precipitate, resembling ferric hydroxide, was recovered. This precipitate appeared to contain the bulk of the actinium-227 and thorium-227.

The presumed barium nitrate crystals and the hydroxide precipitate were redissolved and combined. The solution was evaporated to dryness, and it was found that only a slight residue remained. The residue was taken up in dilute nitric acid in preparation for an anion exchange separation of radium-223. The gamma activity of the sample was approximately 13 times as great as that from a standard sample containing 10 microcuries of actinium.

The sample was treated by the same procedure as that used for the run with eight microcuries of actinium-227. (MLM-CF-60-12-99, p. 68-70). The radium-223 fraction was evaporated to dryness on a hot plate. A dark residue was noted, indicating that there had been some attack on the Dowex-1 resin. The residue was taken up in concentrated nitric acid and again evaporated to dryness. A white residue remained, equivalent in amount to the residue previously presumed to be barium nitrate. This residue dissolved readily and quantitatively in distilled water.

The actinium was recovered from the anion column by elution with nine normal hydrochloric acid. Upon evaporation to dryness, it was found that there was no appreciable residue. This repurified actinium-thorium fraction will be retained for later use as a source of radium-223.

The fractions recovered from the anion exchange separation of radium-223 have not yet been analyzed, but it is estimated that the yields were comparable to those reported for the eight-microcurie run.

DETERMINATION OF COINCIDENCE CORRECTION A method is being developed for determining the coincidence correction in proportional alpha counters by following the decay of a short-lived radioisotope. The radioisotope being used for this purpose is lead-211, a 36-minute beta-emitter, which decays to bismuth-211 and polonium-211, two alpha-emitters having half-lives of 2.16 minutes and 0.56 second, respectively.

Thirteen runs have been made in the Nuclear Measurements Corporation PC-1 counter (No. 64). The data have been turned over to the Computer Analysis group for least squares determinations of the half-life and resolution time. Included in the series were three runs in which the counting interval was 30 minutes, and the data from these runs have been analyzed manually (Table 3). The results shown represent the best fit of the counting data for arbitrarily selected values of the resolution time. In each case, a higher probable error was obtained when the resolution time was either higher or lower than that given.

The grand mean half-life of lead-211 was calculated from the individual values in Table 3, weighted inversely as the squares of their probable errors. The half-lives calculated from each run were plotted as functions of the resolution times arbitrarily selected, and the resolution time corresponding to the grand mean half-life was taken to be the best value in each run. The probable errors indicated in Table 4 are proportional to the probable error of the half-life. The grand mean resolution time was calculated from the individual values in Table 4, weighted inversely as the squares of their probable errors.

Table 3

HALF-LIFE OF LEAD-211
(30 MINUTE COUNTING INTERVALS)

Run No.	n*	T X 10^7 (min.)	$T_{1/2}$ (min.)	Prob. Error (min.)
0216	12	0.7	36.133	0.011
0220	12	0.8	36.119	0.010
0227	9	0.55	36.211	0.015
			---	---
		Grand Mean	36.141	0.007

n* - number of observations

Table 4

Run No.	T X 10^7 (min.)	Prob. Error of T X 10^7 (min.)
0216	0.650	0.038
0220	0.708	0.028
0227	0.791	0.029
	--	--
Grand Mean	0.727	0.066

It should be noted that the half-lives indicated in Table 3 were calculated from equally weighted data. The computer program includes a weighting factor for the precision of each count, and the half-life is therefore subject to some correction.

The grand mean value of T can be assumed to be slightly high, as shown in last month's report, because the counting interval was a large fraction of the half-life. This value is also subject to later correction.

REACTOR FUELS AND MATERIALS DEVELOPMENT

Reactor fuels and materials development includes research, development and design incident to the development of materials or techniques which lead to higher performance and/or lower costs for full scale power reactors.

Plutonium Alloy Research *Plutonium, which has a high neutron efficiency, is being considered for use in reactors of the fast breeder type. Mound Laboratory has been given the responsibility for acquiring data on some of the proposed fuel systems, and for maintaining technical cognizance of fuel cycle problems associated with the fast breeder reactors. Research has been initiated to determine the density, viscosity, thermal capacity, thermal conductivity and phase equilibria of plutonium and plutonium alloys proposed as fuels for these reactors.*

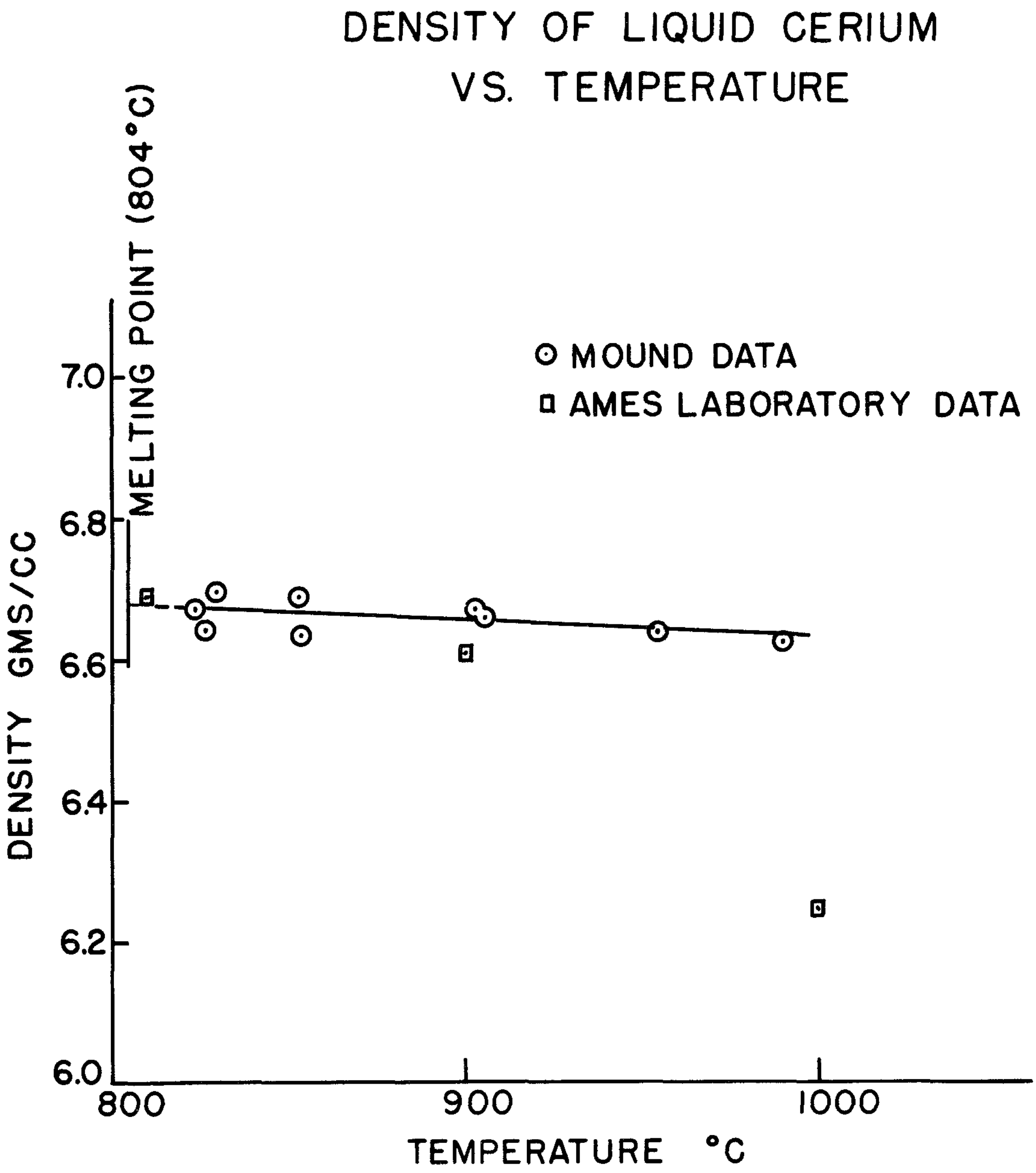
Density The density of liquid cerium has been determined from 825 to 1000°C by the vacuum pycnometer method. Tantalum pycnometers with approximately 0.5 cubic centimeters of volume were used. The results are shown in Table 5 and Figure 1. The line drawn in Figure 1 is a least-squares fit to the data with a 0.5 per cent maximum deviation between the line and any point. Also shown are three density values as given by Ames Laboratory (ISC-659) in which the sessile-drop method was used. Except for the values at 1000°C, the Mound Laboratory and Ames Laboratory values are in fairly close agreement.

The thermal coefficient of cubical expansion for liquid cerium, 33×10^{-6} cm^3/cm^3/°C, is very small for a liquid and only slightly larger than the coefficient for cubical expansion of solid cerium, 21×10^{-6} cm^3/cm^3/°C. The density of the liquid at the melting point, 804°C, is found by extrapolation in Figure 1 to be 6.68 g/cm^3. The density of solid metal has never been measured up to the melting point; however, if a constant expansion coefficient is assumed from room temperature to 804°C, the density would be 6.62 g/cm^3, or if a 10 per cent volume contraction is assumed when beta cerium converts to gamma, as suggested by Trombe and Foex, Comptes Rendus, 217, 501 (1943), then the density would be 6.71 g/cm^3. In either case, the volume change upon fusion is unusually small, less than 0.5 per cent.

Table 5

DENSITY OF LIQUID CERIUM

Temperature °C	Density g/cm^3
824	6.672
827	6.644
830	6.699
853	6.693
854	6.637
903	6.675
906	6.663
955	6.640
990	6.628


DENSITY OF LIQUID CERIUM
VS. TEMPERATURE
MELTING POINT (804°C)
⊙ MOUND DATA
□ AMES LABORATORY DATA
DENSITY GMS/CC
7.0
6.8
6.6
6.4
6.2
6.0
800
900
1000
TEMPERATURE °C

8830

Viscosity The viscosities of molten lanthanum and praseodymium metals have been investigated from their melting points to 994 and 996°C, respectively, in the Mound Laboratory oscillating cup viscometer. The physical properties of the rare earth liquid metals are being measured for comparisons with the properties of the transuranium liquid metals.

A high purity lanthanum metal rod weighing 75.4617 grams was vacuum cast and welded inside a tantalum viscosity cup. The viscosity was measured at three temperatures from 931 to 994°C with the results shown in Table 6.

Table 6

Metal	Temperature °C	Viscosity (centipoise)
Lanthanum	931	2.24
Lanthanum	949	2.08
Lanthanum	994	2.02
Praseodymium	968	1.2
Praseodymium	996	1.3

A 99 plus per cent pure praseodymium rod weighing 60.3824 grams was vacuum cast and welded inside a tantalum viscosity cup. The viscosity was measured at two temperatures, 968 and 996°C with the results shown in Table 6. Since no values for the density of molten praseodymium have been found, an estimate was made on the basis of the densities reported for liquid cerium and lanthanum metals. Inasmuch as a one per cent error in density corresponds to a one per cent error in the viscosity calculation, the values reported for praseodymium are only as accurate as the estimate of its liquid density.

The viscosities of liquid lanthanum and praseodymium are normal in the sense that most other liquid metals fall in this range. Since lanthanum and praseodymium bracket cerium in this periodic chart of the elements, it is interesting that the viscosity of liquid cerium is so high that it could not be measured in the Mound Laboratory viscometer as it is presently designed. Tantalum tubing for making larger diameter tantalum cups for measurement of the viscosity of cerium is in transit.

The investigation of graphite liners for viscosity cups was continued by sealing 138.8038 grams 99.999 plus per cent pure lead in a graphite lined tantalum cup. Viscosity determinations were made at eight different temperatures from 342 to 702°C. The results are under investigation.

Twenty-nine new tantalum crucibles have been fabricated with an increase of one-quarter inch in their length. The additional length will permit a larger ratio of height to radius. An evaluation of this means of minimizing end effects will be made with a determination of the viscosity of liquid bismuth to 1000°C.

Simultaneously, a new torsion fibre, 0.005 inch diameter, 92 per cent platinum, 8 per cent tungsten alloy, is being evaluated. A 0.010 inch diameter fibre of the same composition will be used to re-evaluate the viscosities of lanthanum and praseodymium.

High Temperature Calorimeter The fabrication of parts for the high temperature calorimeter is nearing completion. The three parts which have yet to be made are:

1. The "Lava" guide caps through which the alumina lead-out insulators will pass, and which will be located on the top and the bottom of the temperature controller forms.
2. The "Lava" bearing plates which will support the heater-thermal-sleeve assembly and eliminate any weight on the calorimeter bridge and heater wires.
3. The radiation shields which will protect the soldered electrical connections at the top of the vacuum enclosure.

A multiple terminal hermetic seal header has been made and will be used to bring the wiring from the calorimeter through the hood wall to the control console. The cylindrical metal baffle tube has been bored out so that the nickel block no longer fits snugly inside. The block is now centered in the baffle tube by means of ceramic spacers located at the top and bottom edges of the nickel block. If the tube were in contact with the nickel block, the block controller would try to regulate the block and the baffle tube temperature. With the separation, better control of the block temperature will be possible.

Metallography An expanded annealing facility has been installed so that a total of 10 tube furnaces for handling encapsulated metal buttons are now available.

An additional facility is under construction to provide more space for plutonium alloy preparation and handling . Twelve linear feet of drybox space with circulating dry air is included with three linear feet of fume hood space. Two vacuum systems are being installed along with a two-station induction heater. Low voltage heating can be added if desired. The main advantage of the facility is that the plutonium metal can be cleaned, weighed and passed over to the melting apparatus without removing it from the dry air atmosphere. The fume hood will be used for nonradioactive work and for encapsulation of buttons for annealing.

Preliminary qualitative tests are being made to study the "wetting" properties of plutonium alloys on tantalum. Small discs of 0.006 inch tantalum foil, 5/8 inch in diameter, were crimped around the outer edge. The discs were heated in a high vacuum to approximately 1100°C for outgassing prior to introduction of the plutonium.

The plutonium specimens were heated to temperatures of 800 to 1000°C in a vacuum of 3×10^{-5} millimeters of mercury. In the first trial with pure plutonium, the liquid ran around the crimped edge but did not wet the flat surface of the tantalum. With a 43 atom per cent cobalt alloy, the wettability of the plutonium was considerably increased as the liquid covered all of the tantalum except for a small spot in the center of the disc. These discs will be sectioned to determine the thickness of the alloy and the corrosion of the tantalum. Also, the wettability of the copper and gold alloys will be examined.

Plutonium-Bearing Glass Fibers *At the request of the Reactor Fuels and Materials Development branch of the AEC, a cooperative experiment in the fabrication of plutonium-bearing glass fibers is being carried out by Rensselaer Polytechnic Institute, Owens-Corning Fiberglas Corporation and Mound Laboratory.*

Leaching tests in water, 0.1 normal hydrochloric acid and 0.5 normal nitric acid were continued on fibers of an experimental glass containing silicon dioxide, aluminum oxide, calcium oxide, sodium oxide, potassium oxide and ten weight per cent plutonium oxide. Leaching in room temperature water for 2206 hours resulted in loss of 0.15 per cent of the plutonium oxide from the glass fibers; and leaching in 0.1 normal hydrochloric acid at room temperature for 2183 hours resulted in loss of 0.24 per cent of the plutonium oxide from the glass fibers; and leaching in 0.5 normal nitric acid at room temperature for 1363 hours resulted in loss of 0.65 per cent of the plutonium oxide from the glass fibers.

Leaching tests were continued on fibers of an experimental glass containing 15 weight per cent plutonium oxide dissolved in the same parent glass composition as above. After 666 hours of leaching in room temperature water, 0.065 per cent of the plutonium oxide has been removed from the glass fibers. This loss is comparable to the loss from the glass fibers containing ten weight per cent plutonium oxide when leached for the same length of time under the same conditions. These results indicate that the rate of dissolution of plutonium oxide from the glass fibers by the leaching solution is not related to the plutonium oxide concentration in the glass. It is dependent instead on the concentration of the alkali metal oxides in the glass. The glass composition described above has the highest chemical durability of all glasses evaluated to date. At present eight additional parent glass compositions are under test to determine their ability to dissolve plutonium oxide and their chemical durability. These glasses all represent variations in the chemical content of the above composition.

A device for measuring tensile strength of plutonium bearing glass fibers has been assembled. It consists of a very sensitive precalibrated spring through which the test fiber is loaded at a constant rate until failure occurs. In preliminary tests of the apparatus, fibers of an experimental glass containing 56 weight per cent silicon dioxide, 24 weight per cent sodium oxide and 20 weight per cent plutonium oxide were employed. These fibers, about 14 microns diameter and selected from a freshly coiled specimen, gave a tensile strength of 87,000 psi. Fibers of the same composition, but having remained for one month in laboratory air, gave a tensile strength of 61,000 psi. The loss of tensile strength was attributed to surface deterioration of the hygroscopic fibers. However, both tensile values may be low due to an apparent source of error noted during the tests. This resulted from the inability to align the test fiber with the load axis. When the fiber was placed in tension, a slight bend occurred near each end where the fiber was secured. Failure invariably occurred near one end of the test fiber, and the error introduced by this condition resulted in low values for the tensile strength. Refinements in the method of mounting the test fiber will be made to improve the accuracy of the tensile values.