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THERMAL PROPERTIES OF FIBERBOARD OVERPACK MATERIALS IN THE 9975 SHIPPING PACKAGE

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ABSTRACT

The 9975 shipping package incorporates a cane fiberboard overpack for thermal insulation and impact resistance. Thermal properties (thermal conductivity and specific heat capacity) have been measured on cane fiberboard and a similar wood fiber-based product at several temperatures representing potential storage conditions. While the two products exhibit similar behavior, the measured specific heat capacity varies significantly from prior data. The current data are being developed as the basis to verify that this material remains acceptable over the extended storage time period.

INTRODUCTION

In support of a Savannah River Site (SRS) program of surveillance and monitoring for long term plutonium (Pu) storage, thermal conductivity and specific heat capacity measurements were performed on cane fiberboard overpack materials taken from a 9975 shipping package. Replicate tests were performed at three different temperatures and a range of values were obtained. The average thermal conductivity (k) values (perpendicular to the plane of the fiberboard) range from 0.059 W/mK at 25°C to 0.073 W/mK at 91°C. Average k values (parallel to the plane of the fiberboard) range from 0.103 W/mK at 25°C to 0.114 W/mK at 91°C. Average specific heat capacity values range from 600 J/kgK at 25°C to 697 J/kgK at 91°C. These values are considered baseline values for the 9975 overpack material in the storage facility. Additional testing will be performed to evaluate degradation of materials under actual storage conditions.

BACKGROUND

The 9975 is a certified radioactive material shipping package designed and sponsored by the Savannah River National Laboratory (SRNL). Celotex[®] cane fiberboard is used as overpack material in the 9975 for its insulating, criticality

control, and energy absorption properties. Celotex[®] is specified as cane fiberboard manufactured per ASTM Specification C208-95 and is required to be regular grade wall sheathing with a nominal density of 240 kg/m³ (15 lb/ft³). The ASTM specification requires a maximum thermal conductivity of 0.058 W/mK at a mean temperature of 24 ± 3°C (75 ± 5°F). Knight-Celotex is the only company that produces fiberboard from a sugar cane by-product (bagasse) and bonding agents. Knight-Celotex produces Celotex[®] cane fiberboard at its plant in Marrero, Louisiana.

The thermal integrity of the shipping package was based on work by Sanchez et al.[1] Sanchez performed thermal testing on cane fiberboard from a 6M shipping package. The 6M fiberboard is specified in Title 49 of the Code of Federal Regulations (49 CFR) section 178.354-3(c)(1). The 49 CFR 178.354-3(c)(1) states that the “insulation is to be made of industrial cane fiberboard having a density of at least 15 lb/ft³ (0.24 g/cc).” This is a generic specification and allows variation in the type of cane fiberboard used in the 6M package. The 9975 package requirements for the cane fiberboard are more prescriptive and all cane fiberboard material used in 9975 packages meets the generic 6M requirement.

CANE FIBERBOARD TEST MATERIALS

Cane fiberboard thermal conductivity samples 30.5 cm x 30.5 cm x 3.8 cm (12 inch x 12 inch x 1.5 inches) were machined at SRS from material taken from 9975 packages that had been drop tested in 2000. The samples of undamaged material were oriented so that thermal conductivity measurements could be performed in both the perpendicular (heat flow across the glue joints) and parallel (heat flow in line with the glue joints or radial) directions. These two sample orientations are shown in Fig. 1. Three samples each weighing less than 10-mg were prepared for testing specific heat

capacity. The material used in these tests had been stored for approximately 4 years in a closed non-environmentally controlled warehouse. Because the dropped packages did not contain heat sources while in storage, their thermal history is less severe than that of the Pu storage 9975 packages, which have internal heating dependent upon the amount and types of materials contained. Thus, the Celotex® from the dropped 9975 packages is expected to represent packages near the beginning of storage life in the storage facility.

In order to calculate the density of the samples used for testing thermal conductivity and specific heat capacity, dimensions and weights were measured at SRNL prior to shipping. Dimensions were measured using digital calipers accurate to ± 0.001 cm. Weight measurement accuracy was ± 0.003 kg. Cane fiberboard density calculations were performed with results as shown in Table 1. Error propagation in the density calculations was calculated at ± 1 kg/m³.

TEST DESCRIPTION

An independent ISO 9002 Certified laboratory was contracted to perform thermal conductivity measurements on six (30.5 cm x 30.5 cm x 3.8 cm) cane fiberboard blocks provided by SRNL. The laboratory is a leading manufacturer of thermophysical property testing instruments and has a laboratory to perform research and property testing. ISO 9002 certification ensures that appropriate Quality Assurance programs and processes are being used. All testing was performed per American Society for Testing and Materials (ASTM) standards and National Institute of Standards and Technology (NIST) certified reference materials are used for instrument calibration. Specific tests were computer controlled.

Specific heat capacity measurements were performed on separate, smaller samples prepared by the laboratory from material supplied by SRNL. A Fox Model 300, heat flow meter apparatus designed to use flat slab specimens, was used to perform the thermal conductivity measurements per ASTM C518. This is a comparative measurement method since specimens of known thermal transmission properties are used for comparison with the test samples and for calibration purposes. The heat flow meter unit establishes steady state one-dimensional heat flux perpendicular to the test specimen (direction as shown by arrow in Figure 2) between two parallel plates at constant but different temperatures. Appropriate calibration of the heat flux transducers with a glass fiber composite material standard, and measurement of the plate temperatures combined with the plate separation measurement allows for Fourier's law of heat conduction to be used for thermal conductivity calculations.

The laboratory estimated the accuracy of the thermal conductivity tests at $\pm 5\%$ of the test values. The system was calibrated using an in-house glass-fiber composite reference standard. An initial test was performed at ambient temperature ($\sim 25^\circ\text{C}$). The sample temperature was subsequently raised to 52°C and then 91°C , testing after stabilization at each step. The test was repeated on five (5) additional cane fiberboard

blocks for a total of eighteen (18) measurements. The measurements were taken three times at each temperature. Two different orientations were tested.

Samples weighing less than 10-mg were used for specific heat capacity testing. The samples were removed without any portion of the glue joints. These tests were performed with a TA Model 2920 differential scanning calorimeter per ASTM E1269. This test method consists of heating the test material at a controlled rate in a zero humidity nitrogen atmosphere at the desired temperature. Differences in heat flow into the fiberboard test sample and a synthetic sapphire disk reference, due to energy changes in the material, was continually monitored and recorded. Estimated accuracy of the specific heat capacity tests was $\pm 5\%$ of the test values.

Figure 2 shows the cane fiberboard samples used for testing in the axial direction.

TEST RESULTS

Thermal conductivity (k) and specific heat capacity (Cp) values from the cane fiberboard samples are recorded in Table 1 and shown graphically in Figure 3 and Figure 4. Average values for k perpendicular to the plane of the fiberboard are 0.059 W/mK at 25°C , 0.063 W/mK at 52°C , and 0.073 W/mK at 91°C . The average k value at 25°C is 0.001 units above the maximum thermal conductivity value allowed by ASTM C208 (0.058 W/mK). Average values for k parallel to the plane of the fiberboard are 0.103 W/mK at 25°C , 0.107 W/mK at 91°C , and 0.114 W/mK at 91°C . Thermal conductivity is clearly temperature dependent. The parallel k values are 1.6 to 1.7 times higher than the perpendicular k values. The scatter in the thermal conductivity data among the three samples is shown in Figure 5. The k values at room temperature vary by a maximum of 0.005 with a 9% difference between the maximum and minimum values.

Average Cp values were measured as 600 J/kgK at 25°C , 620 K/kgK at 52°C , and 697 J/kgK at 91°C . The variability in the specific heat capacity values are shown in Figure 6. The values measured from a sample from the bottom of the cube appear to be significantly higher than the samples taken from the middle and top of the cube.

Density values of the 9975 cane fiberboard ranged from 265 to 311 kg/m³, see Table 1. The lower density value was obtained on a sample slightly lighter in color than the other two.

COMPARISON WITH SANCHEZ DATA

The data obtained from the recent thermal tests was compared with the Sanchez data.[1] The thermal conductivity values (perpendicular direction) in this test at room temperature, 25°C , are approximately 9.4% higher than the comparable Sanchez data as shown in Table 1.[1] At 90°C , the new values are only 8.5% higher than the Sanchez value at 86°C . Thermal conductivity decreases above 260°C as the cane

fiberboard becomes charred and densification occurs. Measurements were made on material in the as-received condition to represent baseline properties. Sample conditioning was not described by Sanchez [1] and no reference was provided regarding the source of the fiberboard or a detailed description of its properties, except that contained in the title of the report, DOT 6M containers. Thus, it is assumed that the tested fiberboard came from a 6M package and meets the intent of the generic requirement of the 15 lb/ft³ (240 kg/m³) density. Likewise, the Cp data generated for this paper was compared with the Sanchez data. Recent cane fiberboard specific heat capacity values (Table 1) are substantially lower than those reported by Sanchez, 600 average versus 1280 J/(kgK) at 25°C. The Sanchez data are shown in Fig. 4. Two of the values (1st and 2nd columns) are the same at room temperature and vary slightly at 52°C, within 2% of each other. The third column values are 9% higher than the other two. In Figure 4, the average cane fiberboard values obtained in the current tests are plotted versus the Sanchez values. The scatter in the data is seen in Figure 6. Regardless of the temperature, the Sanchez values are approximately twice that of these recently measured values. Both the independent laboratory and Sanchez used the differential scanning calorimetry method to measure specific heat capacity. Specific details of the Sanchez test were not revealed and therefore a direct comparison and correlation of the data is not possible.[1]

In trying to correlate the data, prior purchases of 6M containers were reviewed for material information. Following the purchase of 6M shipping packages by WSRC in 2002, it was confirmed that the new 6M used cane fiberboard. The 6M supplier had purchased ½ inch x 4 foot x 8 feet and 2 inch x 2 foot x 4 foot cane fiberboard from a lumber company in Knoxville, TN. Two inch thick cane fiberboard is only supplied as roof insulation board. Conversation with a representative from that lumber company revealed that the 6M supplier requested high density cane fiberboard in their recent order. High density cane fiberboard is currently marketed by Knight-Celotex as High Density Fiberboard Roof Insulation. The high density product meets ASTM C-208 requirements for Type II Roof Insulation Board, Grade 1 with a “maximum thermal conductivity of 0.055 W/mK at a mean temperature of 75 ± 5°F (24 ± 3°C), a minimum tensile strength (perpendicular to the surface) of 500 lb/ft² (23.9 kPa), and 50 lb/in² (345 kPa) parallel to the surface with a density greater than 15 lb/ft³ (240 kg/m³).”

The measured density of the 6M fiberboard used in the Sanchez tests was 270 kg/m³ (16.9 lb/ft³) versus 265-311 kg/m³ (16.5-19.4 lb/ft³) for these cane fiberboard tests and the minimum 240 kg/m³ required by ASTM C-208. Since minimal property information for the 6M insulating material was provided, we have postulated why the test values could be different. One reason for the lower density could be that the 6M cane fiberboard was not glued and that solid samples were used or that the fiberboard contained less moisture compared with the 9975 cane fiberboard used in this test program. Also, as-manufactured cane fiberboard densities are known to vary

according to the manufacturer depending on the weather and the time of year.

ADDITIONAL FIBERBOARD TESTING AND LITERATURE DATA

Prior to testing cane fiberboard obtained from dropped 9975 packages, an additional set of samples were fabricated from fiberboard which was purchased from the same lumber company in Knoxville, TN, who confirmed that the Type IV Grade 1 cane wall sheathing was from the Knight-Celotex Marrero plant. The density of the as-machined blocks, received from the vendor was 345-355 kg/m³ (21.5 – 22.2 lb/ft³). Using similar 5 cm cube samples, also machined by the vendor, the thick glue layer between the fiberboard sections was removed with the average density of the remaining fiberboard measured as 272 kg/m³ (17 lb/ft³). These fiberboard samples were subsequently examined by a Knight-Celotex representative, and visually identified as not being a cane fiberboard product produced by their company. This material will be further identified in this paper as wood based fiberboard. Photographs of these samples are shown in Fig. 7. Notice the thicker glue joints and the finer texture of the wood based fiberboard compared to the 9975 samples shown in Fig. 7. The differences in this material as noted by Knight-Celotex necessitated retesting of fiberboard with assurance that the samples were from a 9975 shipping package. The wood based fiberboard data are reported to show that while differences exist between available fiberboard materials, their temperature behavior follows a similar trend.

The wood based fiberboard data is listed in Table 2. The wood fiberboard thermal conductivity is slightly lower than the cane fiberboard at all temperatures (Fig. 3). Moreover, while the specific heat capacity values are higher than the cane fiberboard they are still approximately 58% of the Sanchez values (Fig. 4).

In SRS testing of cane fiberboard in 1995, thermal conductivity was measured to be 0.057 W/mK at 24°C. This value, in Table 2, is similar to the new data (0.056-0.061 W/mK) from 5 cm thick glued material. However, source and quality information on the fiberboard tested in 1995 was not documented. The SRS internal report noted that thermal conductivity measured in a direction parallel to the surface was about twice the value of thermal conductivity perpendicular to the surface. Furthermore, the National Institute of Standards and Technology website lists a lower thermal conductivity value of 0.047 W/mK at approximately 24°C with reference to Celotex® insulation board (3.6 cm thick).[2] This test was performed in 1953 with no reference to density, test methods, or fiberboard quality. Comments were not provided on whether the Celotex® was glued or tested as a solid piece. It appears that there is no absolute value of thermal conductivity for cane fiberboard and that thermal values should be reported as a range from minimum to maximum depending on density.

The specific heat capacity of solid wood with zero moisture content is 1200 J/kgK at 75°F (24°C) and rises to 1800 J/kgK at 75°F with 20% moisture.[3] Thus, the heat capacity of wood, containing water, is greater than that of dry wood. The specific heat capacity measurements developed for this report are for dry nitrogen conditions. This provides a possible explanation for the difference in Cp values between the 9975 cane fiberboard and the SNL data. The moisture level of the Sanchez test material is unknown.

Based on wood research, specific heat capacity can be corrected for moisture content by summation of the specific heat capacity of dry cellulose fiber plus that of water plus an additional adjustment factor per Equation (1).[3] Using the annual average relative humidity of 70% for the Southeast (Atlanta, GA), the moisture content of solid wood is 13% per references 3-4. Correction of the measured values of the specific heat capacity of cane fiberboard yields a higher value of 1013 J/kgK per the following calculation using Equation (1).

$$C_p = (C_{p_0} + 0.01MC_{p_w} / (1+0.01M)) + A_c \quad (1)$$

- Where M = moisture content in %
 Cp₀ = heat capacity of dry wood in kJ/kgK
 = 0.1031 + 0.003867t
 Cp_w = heat capacity of water = 4.19 kJ/kgK
 A_c = adjustment factor accounting for the additional energy in the wood-water bond = M(B₁ + B₂t + B₃M)
 t = temperature, °K (273 + 25)
 B₁ = -0.06191
 B₂ = 2.36 x 10⁻⁴
 B₃ = -1.33 x 10⁻⁴

Reference 4 recognizes that the relationship between specific heat capacity and moisture content is practically independent of density or wood species. Accordingly, since solid wood and cane fiberboard both contain primarily cellulose, the equation (1) relationship is probably similar, but not exact. The specific heat capacity of cane fiberboard is expected to be higher than solid wood due to its porosity and its accompanying moisture content. Thus, the high specific heat capacity levels reported by Sanchez could be the result of high moisture content, although doubtful since the climate in Albuquerque is typically drier than at SRS.

CONCLUSIONS AND RECOMMENDATIONS

Newly measured cane fiberboard thermal conductivity is similar to that from Sanchez and previous SRS data.

Differences in individual values could be due to density differences, moisture content prior to the room temperature tests, and the quality of the fiberboard (including texture and production differences). However, specific heat capacity values were substantially different than that reported by Sanchez. Additional testing of the 6M fiberboard is recommended to aid in understanding these differences. Since there could be potential moisture differences between the Sanchez laboratory and SRS cane fiberboard, future samples will be conditioned prior to testing to determine how moisture content affects thermal properties.

ACKNOWLEDGMENTS

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REFERENCES

[1] L. C. Sanchez, R. S. Longenbaugh, M. Moss, G. M. Haseman, W. E. Fowler, and E. P. Roth, “Thermal analysis of the 10-gallon and the 55-gallon DOT-6M containers with thermal boundary conditions corresponding to 10CFR71 normal transport and accident conditions, SAND87-1896, Sandia National Laboratories, Albuquerque, NM, March 1988.

[2] R. R. Zarr, Editor, “NIST Heat Transmission Properties of Insulating and Building Materials,” NIST Standard Reference Database 81, National Institute of Standards and Technology, Gaithersburg, MD, <http://srdata.nist.gov/insulation/UT>.

[3] W. Simpson and A. TenWolde, “Physical Properties and Moisture Relations with Wood,” in *Wood Handbook – Wood as an Engineering Material*, Gen. Tech. Rep FPL-GTR-113, Madison, WI, U.S. Department of Agriculture, Forest Service, Forest Products Laboratory, Chapt. 3, p. 3-17 and 3-21 (1999).

[4] W. T. Simpson, “Drying and Control of Moisture Content and Dimensional Changes,” in *Wood Handbook – Wood as an Engineering Material*, Gen. Tech. Rep FPL-GTR-113, Madison, WI, U.S. Department of Agriculture, Forest Service, Forest Products Laboratory, Chapt. 12, p. 12-4 (1999).

of a 2 inch cube sample cut from the cane fiberboard insert in a dropped 9975 package.

Table 1. Measured and reference [1] temperature dependent thermal conductivity, specific heat capacity, and density data.

| Temp. | | 9975 Cane Fiberboard Data from Current Testing | | | Sanchez Data [[1]] |
|--|-------|--|-------|-------|--------------------|
| °F | °C | #1 | #2 | #3 | |
| Thermal Conductivity (W/mK), perpendicular direction | | | | | |
| 77 | (25) | 0.061 | 0.059 | 0.056 | 0.053 |
| 125 | (52) | 0.065 | 0.063 | 0.060 | |
| 187 | (86) | | | | 0.059 |
| 196 | (91) | 0.073 | 0.073 | 0.073 | |
| 295 | (146) | | | | 0.063 |
| 439 | (226) | | | | 0.065 |
| 532 | (278) | | | | 0.051 |
| Thermal Conductivity (W/mK), parallel direction | | | | | |
| 77 | (25) | 0.101 | 0.103 | 0.106 | |
| 126 | (52) | 0.106 | 0.107 | 0.109 | |
| 196 | (91) | 0.113 | 0.114 | 0.116 | |
| Specific Heat Capacity (J/kgK) | | | | | |
| 77 | (25) | 580* | 580* | 640* | 1280 |
| 125 | (52) | 590* | 580* | 690* | |
| 187 | (86) | | | | 1506 |
| 196 | (91) | 640* | 630* | 820* | |
| 295 | (146) | | | | 1745 |
| 439 | (226) | | | | 2046 |
| 532 | (278) | | | | 2063 |
| Density (kg/m ³) | | | | | |
| 77 | (25) | 311 | 311 | 265 | 270 |
| 295 | (146) | | | | 286 |
| 439 | (226) | | | | 297 |
| 532 | (278) | | | | 313 |

* Specific heat capacity samples (without glue) were taken from top (1), middle (2) and bottom (3) locations

Table 2. Wood based fiberboard [2] and previous SRS temperature dependent thermal conductivity, specific heat capacity, and density data.

| Temperature | | Wood based fiberboard Data | | | 1995 SRS Data |
|--|------|----------------------------|-------|-------|---------------|
| °F | °C | #1 | #2 | #3 | |
| Thermal Conductivity (W/mK), perpendicular direction | | | | | |
| 75 | (24) | | | | 0.057 |
| 77 | (25) | 0.058 | 0.057 | 0.055 | |
| 125 | (52) | 0.059 | 0.057 | 0.056 | |
| 180 | (82) | | | | 0.063 |
| 194 | (90) | 0.064 | 0.064 | 0.06 | |
| Specific Heat Capacity (J/kgK) | | | | | |
| 77 | (25) | 720 | 830 | 760 | |
| 122 | (50) | 720 | 880 | 750 | |
| 194 | (90) | 780 | 990 | 850 | |
| Density (kg/m ³) | | | | | |
| 75 | (24) | | | | 251 |
| 77 | (25) | 355 | 355 | 345 | |

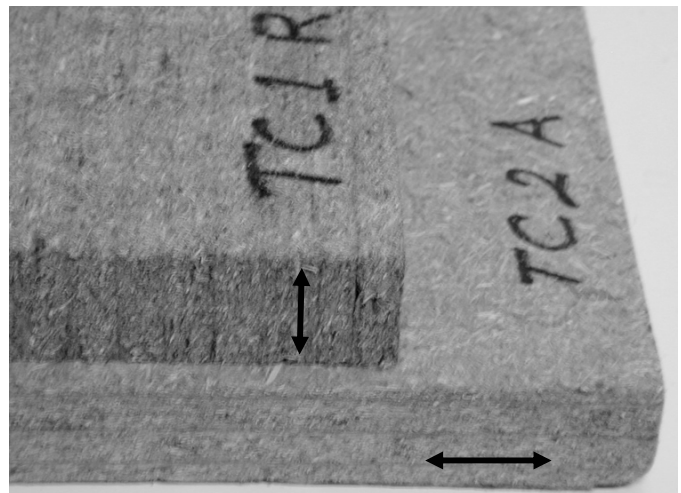


Figure 1. Perpendicular and parallel orientations (shown by glue joints) of the cane fiberboard blocks used for thermal conductivity measurements. Block thickness is 3.8 cm.

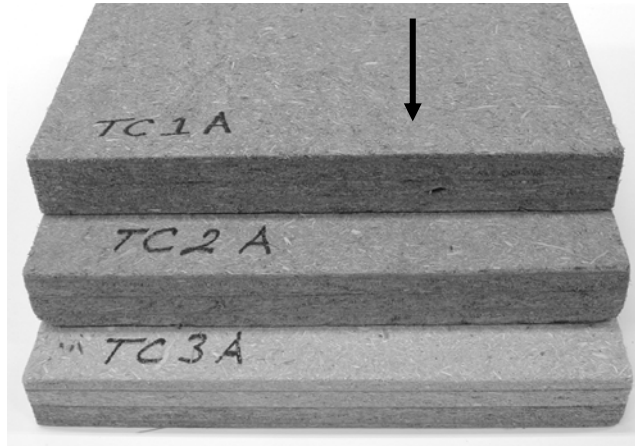


Figure 2. 30.5 x 30.5 x 3.8 cm glued cane fiberboard samples which were taken from a 9975 package. Arrow shows the expected heat flow direction during thermal conductivity testing.

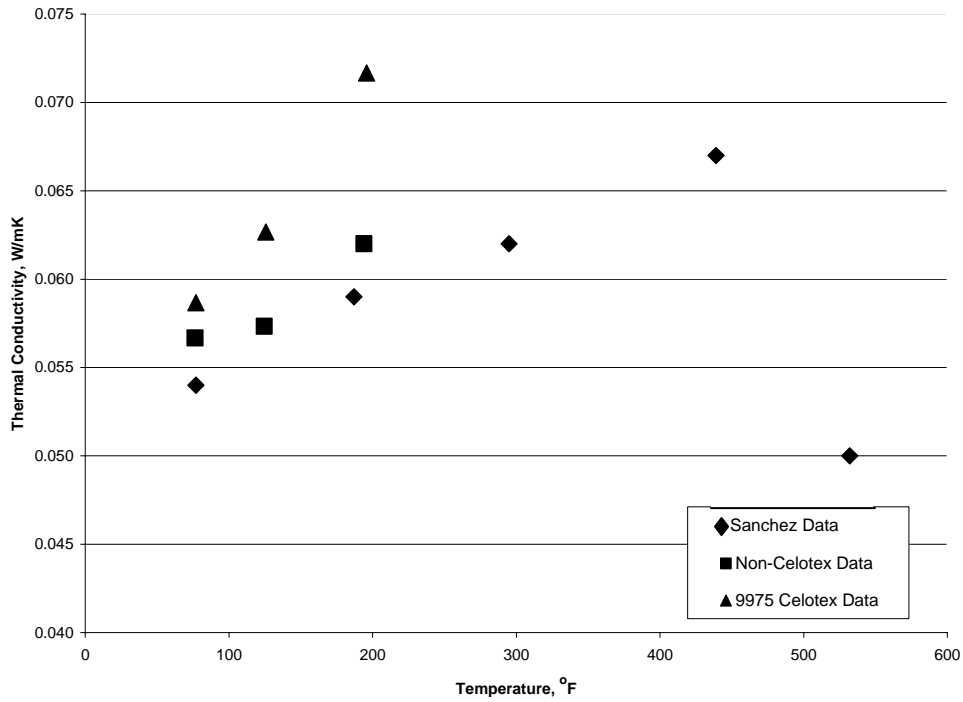


Figure 3. Temperature dependent thermal conductivity values (perpendicular or axial direction) from the recent 9975 cane fiberboard tests with comparison to the Sanchez [1] and wood based fiberboard (Non-Celotex) data.

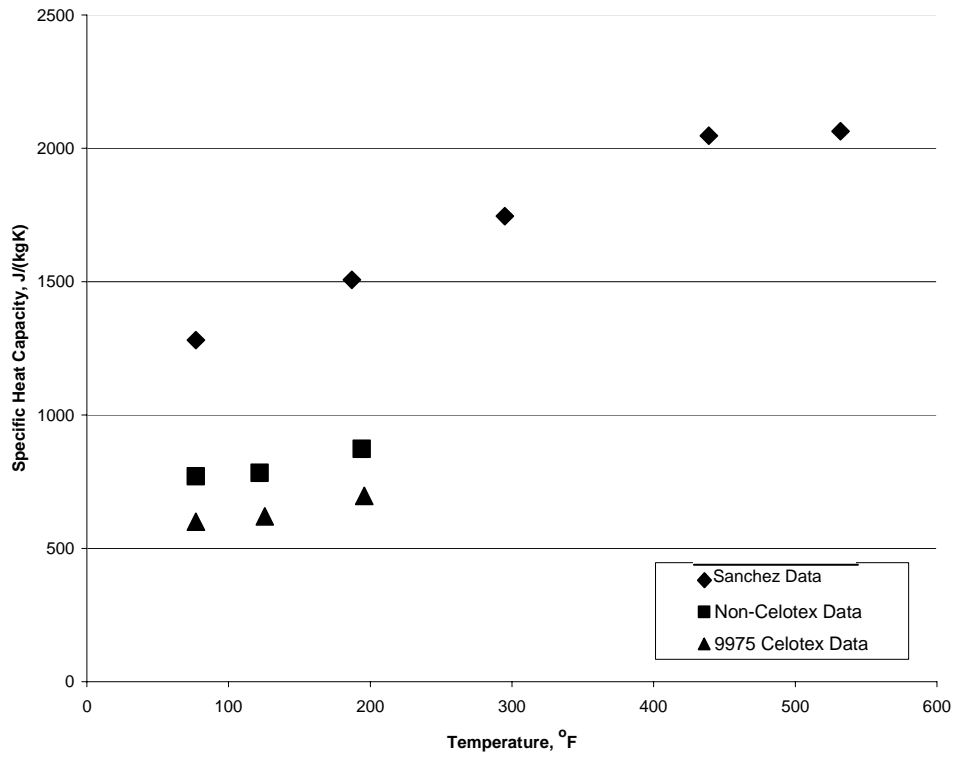


Figure 4. Temperature dependent specific heat capacity values for recent 9975 cane, Sanchez [1] fiberboard tests, and wood based fiberboard (Non-Celotex).

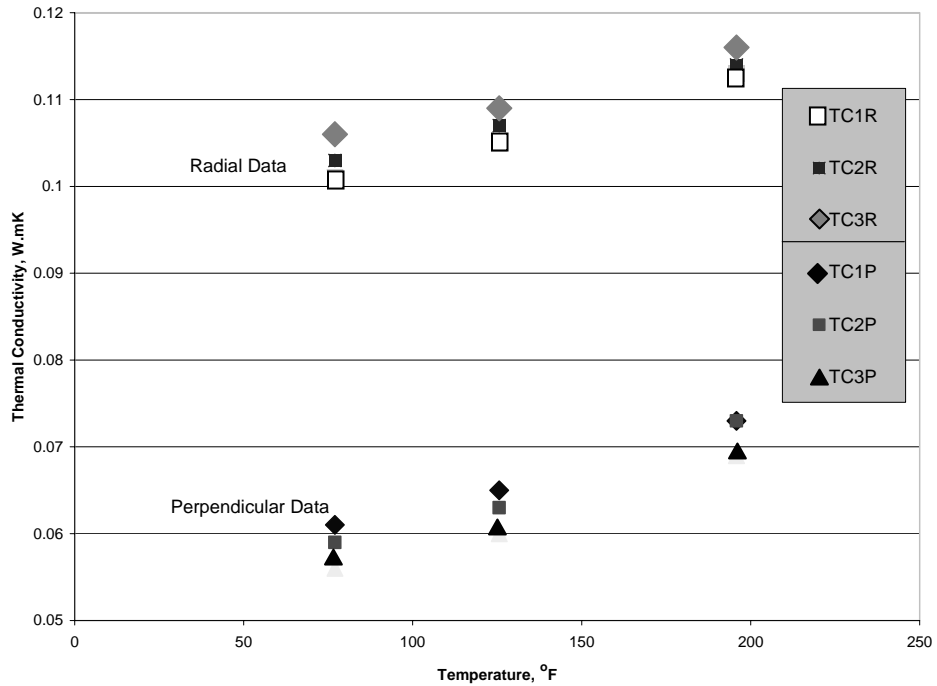


Figure 5. Variability in individual sample values from thermal conductivity tests with heat flux perpendicular and parallel (radial) to the glue joints in the 9975 cane fiberboard.

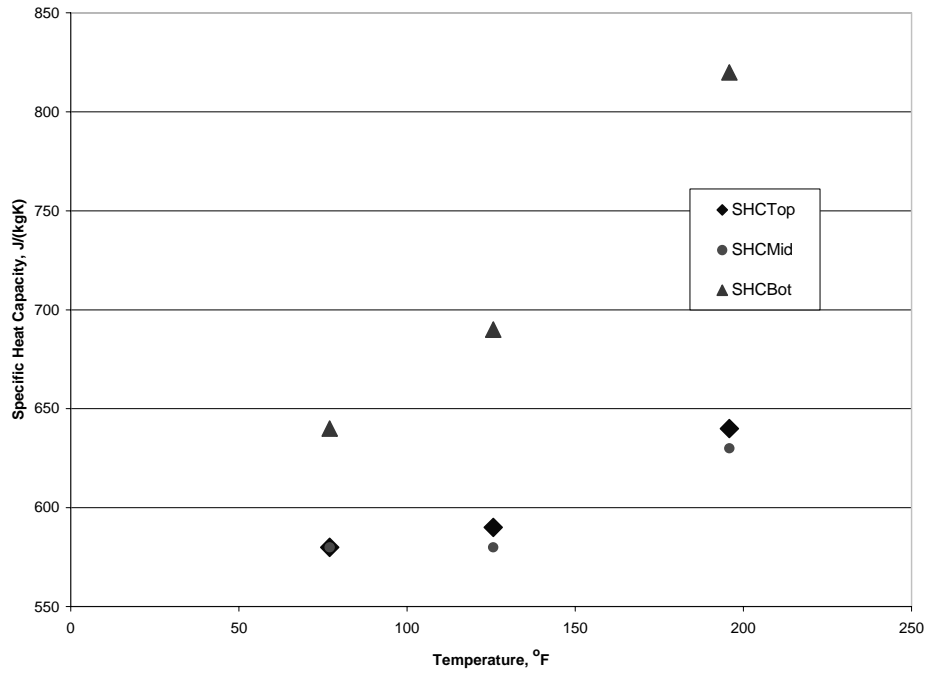
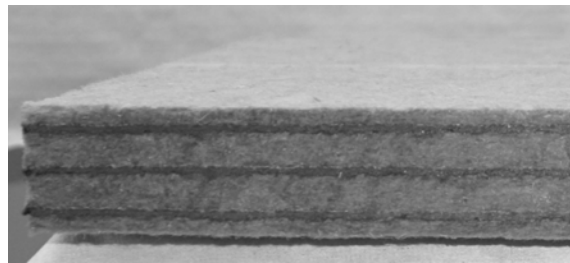


Figure 6. Variability in temperature dependant specific heat capacity values from samples taken from the top, middle, and bottom layers in the 9975 cane fiberboard sample.



A



B

Figure 7. (A) Blocks (30.5 cm x 30.5 cm x 3.8 cm) of wood based fiberboard revealing side view. Notice finer texture and thicker glue joints in these samples compared to the cane fiberboard samples in the lower photo (B). There are three glue joints in each sample.