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White Paper: Demonstration of Equivalency of Cane and Softwood Based CelotexTM for 9975 Packaging

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Acronyms

ASTM American Society of Testing and Materials

DOE Department of Energy

HAC Hypothetical Accident Conditions NCT Normal Conditions of Transport

pcf pounds per cubic foot

PCV Primary Containment Vessel

ppm parts per million PVAc polyvinyl acetate

SARP Safety Analysis Report for Packaging

SCV Secondary Containment Vessel TGA Thermogravimetric Analyzer

Purpose

The purpose of this White Paper is to demonstrate that softwood-based Celotex[™] from the Knight-Celotex Danville Plant has performance equivalent to cane-based Celotex[™] from the Knight-Celotex Marrero Plant for transportation in a 9975 package.

Background

Cane-based CelotexTM has been used extensively in various DOE packages as a thermal insulator and impact absorber. Cane-based CelotexTM for the 9975 was manufactured by Knight-Celotex Fiberboard at their Marrero Plant in Louisiana. However, Knight-Celotex Fiberboard shut down their Marrero Plant in early 2007 due to impacts from hurricane Katrina and other economic factors. Therefore, cane-based CelotexTM is no longer available for use in the manufacture of new 9975 packages. Knight-Celotex Fiberboard has CelotexTM manufacturing plants in Danville, VA and Sunbury, PA that use softwood and hardwood, respectively, as a raw material in the manufacturing of CelotexTM (see Figure 1).



Figure 1: From Left to Right. Hardwood-based Celotex TM from Sunbury Plant, Softwood-based Celotex TM from Danville Plant, and Cane-based Celotex TM from Marrero Plant.

Discussion

The 9975 SARP, Revision 1 currently under review specifies cane fiberboard, CelotexTM brand, 0.5" thick, Type IV, Grade 1 per ASTM C208-95, 14 to 16 pcf density [1,2]. All Knight-Celotex premium fiberboard insulating sheathing, previously produced at Marrero, LA, and currently being produced at the Danville, VA and Sunbury, PA, meet ASTM C208-95 (reapproved 2001) for Type IV, Grade 1 fiberboard. However, of the two wood-based CelotexTM products, only softwood-based CelotexTM from the Danville Plant meets the density requirement of 14 to 16 pcf specified for 9975 fabrication (see Attachment 1).

The following discussion compares the attributes of cane- and softwood-based CelotexTM as credited in the 9975 SARP, Revision 1 to show equivalency between the two products. The discussion is broken into five topical areas as it relates to CelotexTM performance. These topical areas are the chemical, structural, thermal, criticality, and shielding properties of the material.

Chemical

Fiberboard, whether produced from softwood or sugarcane bagasse (i.e. biomass following juice extraction of the sugarcane stalk), is a lignocellulosic biomass comprised primarily of cellulose, hemicellulose, and lignin. Other minor constituents of softwood and sugarcane bagasse are water insoluble extractives which include terpenes, fatty acids, aromatic compounds, oils, and waxes. Both softwood and sugarcane bagasse may contain approximately 1-7% extractives [3-6]. The average composition of the primary constituents (i.e. cellulose, hemicellulose, and lignin) for sugarcane bagasse and softwood is detailed below in Table 1.

Table 1: Average Weight Percent Composition of Sugarcane Bagasse and Softwood

	Sugarcane Bagasse [7]	Softwood [3, 8]
Cellulose	26.6-54.3	40-51.4
Hemicellulose	22.3-29.7	25-29
Lignin	14.3-24.5	19.2-31

The cellulose and hemicellulose reported for softwood falls entirely within the range for sugarcane bagasse. There is also significant overlap in lignin composition for the two materials. In addition, as part of the CelotexTM manufacturing process, up to 10% starch, in the form of corn starch, may be added to the biomass as a binding agent regardless of whether the fiberboard is cane or softwood based (see Attachments 2 & 3). As a point of note, clay, carbon black, wax, and adhesive can be applied as a moisture barrier as part of the normal manufacturing process. If present, the moisture barrier is removed prior to 9975 fabrication.

The chemical composition can vary significantly, even in the same kind of woody biomass, due to habitat and climate [9]. As shown above, there are large variances in the chemical composition of softwood and sugarcane bagasse biomasses with softwoods having the smallest variances. Additionally, Knight-Celotex may use newsprint material as part of their normal cane- and softwood-based fiberboard manufacturing process. However, larger quantities of newsprint have been historically used in cane-based CelotexTM as compared to softwood-based CelotexTM. Softwood-based CelotexTM is a more consistent material than cane-based CelotexTM due to the tighter limits of its individual constituents (i.e. cellulose, hemicellulose, & lignin) and minimal use of newsprint. Therefore, softwood-based CelotexTM is a suitable replacement for cane-based CelotexTM in regards to their chemical constituents.

Another area of concern, in regards to biomass chemistry, is that of chloride content due to its role in stress corrosion cracking of stainless steels. There has been limited testing of leachable chlorides in cane-based CelotexTM with reported results varying from 415 ppm to 944 ppm [10]. Knight-Celotex uses what the industry refers to as a wet form process at all of their CelotexTM manufacturing plants. As the name implies, water is used to wash the biomass and is extracted during the board forming operation [3]. This washing and water extraction process would tend to remove the leachable chlorides. It is judged that softwood fiberboard would not have substantially more leachable chlorides than cane-based fiberboard.

The final area of concern is the formation of lead carbonate on the lead shielding of the 9975 package. The formation of lead carbonate in previous 9975 packages is primarily attributed to the off-gassing of the PVAc glue used in laminating the sheets of CelotexTM [11]. Since the basic chemical constituents and their proportions are similar between softwood and sugarcane bagasse, there is no expectation that softwood-based CelotexTM would significantly increase the reaction rate of lead carbonate formation as compared to cane-based CelotexTM.

Structural

The 9975 package has met the acceptance criteria for NCT and HAC testing as defined by 10CFR71 [1, 12]. The testing included NCT and HAC test (i.e. 30-ft. free drops and puncture), where the cane-based CelotexTM acted as an impact absorber. Additionally, dynamic structural analysis was successfully conducted for a PCV/SCV assembly without an outer drum and CelotexTM at a 55-ft. vertical and horizontal drop. In this analysis, the CelotexTM is not credited as an impact absorbing material for the HAC free drop events. However, it is important to note that whether CelotexTM is manufactured from sugarcane bagasse or softwood, the fiberboard has to meet mechanical property requirements specified in ASTM C208-95 (reapproved 2001) [2]. These mechanical property test requirements include minimum transverse strength, minimum parallel and perpendicular to surface tensile strengths, minimum modulus of rupture, and maximum deflection at specified minimum load. These tests are defined within ASTM C209-07 [13]. Although ASTM C208-95 (reapproved 2001) does not have any requirements as far as the compressibility of fiberboard, the culmination of all required

testing, the manufacturing process being the same for all Knight-Celotex plants, and density limitations as prescribed by the SARP, it is judged that softwood-based Celotex[™] would not behave significantly differently than cane-based Celotex[™] under compression.

Thermal

For the NCT insolation test, as described in 10 CFR 71.71(c)(1), thermal analytical modeling was conducted for purposes of the 9975 SARP with the prescribed insolation heat loads [1, 12]. A temperature limit of 250 °F was imposed for the cane-based CelotexTM under the NCT event. This temperature limit was established based on extended thermal testing as presented in SARP, Revision 1, Appendix 3.16, where at temperatures below 250 °F, weight loss was fairly constant due to primarily moisture evaporation. The NCT thermal modeling resulted in a cane-based CelotexTM temperature of 257 °F, which was considered to have a negligible consequence compared to the temperature limit of 250 °F.

The testing, as described in SARP, Revision 1, Appendix 3.16, is consistent with literature in regards to moisture being the primary constituent in various biomasses undergoing volatilization at temperatures less than 373 K (212 °F) [14]. At temperatures between 373 K (212 °F) and 523 K (482 °F) the extractives decompose creating volatile vapors. Cellulose, hemicellulose, and lignin decompose producing char and volatiles at temperatures above 523 K (482 °F). In addition, due to the low thermal conductivity of fiberboard (i.e. \leq 0.40 BTU·in./h·ft²·°F per ASTM C208-95 (reapproved 2001)), variations of thermal conductivity between cane- and softwood-based CelotexTM would result in similar thermal responses for the package. Due to the similar chemical composition and density of cane- and softwood-based CelotexTM and the maximum thermal conductivity specification of ASTM C208-95 (reapproved 2001) (i.e. \leq 0.40 BTU·in./h·ft²·°F), there is no reason to expect the two types of CelotexTM to behave differently during the NCT insolation test [2].

For the HAC thermal test, as described in 10 CFR 71.73(c)(4), testing of a 9975 package was conducted as discussed in SARP, Revision 1, Appendix 3.5 [1,12]. The test resulted in a char layer forming in the cane-based CelotexTM extending from its exterior to a depth of 1.4 to 2.3 inches. Similar to the justification for the NCT insolation test, due to the similar chemical composition, density, maximum thermal conductivity requirement of ASTM C208-95 (reapproved 2001), there is an expectation the two types of CelotexTM would behave similarly during the HAC test [2].

Tests have been conducted with a TGA to study the pyrolysis characteristics of various biomasses [14]. In particular, bagasse and subabul wood, a softwood indigenous to Mexico, were ground to less than 250 µm particles and tested in a TGA with a heating rate of 50 K/min. The results indicated that bagasse yields 79.7 wt% volatiles and 20.3 wt% char compared to subabul wood yielding 76.3 wt% volatiles and 23.7% char. Bagasse had a maximum rate of decomposition at 677 K (759 °F) and an initial decomposition at 483 K (410 °F). In comparison, subabul wood had a maximum rate of

decomposition at 683 K (770 °F) and an initial decomposition at 498 K (437 °F). The maximum rate of decomposition for both materials was 0.9 wt%/K. Based on this information, the thermal decomposition of softwoods is similar to bagasse.

Criticality

The effect of the HAC sequential test events on the criticality evaluation is discussed in the 9975 SARP, Revision 1 [1]. The HAC events have a higher $k_{\rm eff}$ than NCT events with similar fissile contents, even though the NCT arrays modeled are infinite compared to HAC arrays which are 5x5x2. This is due to the loss of spacing from drop and fire-event testing of the 9975 package. The criticality evaluation reduced the cane-based CelotexTM 9975 package dimensions from the drop and fire test data. In addition, charred cane-based CelotexTM was assumed to be removed from the 9975 package model. As discussed in previous sections, softwood-based CelotexTM should behave in a similar manner (i.e. within the safety margin provided in the criticality evaluation) to cane-based CelotexTM under HAC. Therefore, no negative impacts to $k_{\rm eff}$ (i.e. an increase on $k_{\rm eff}$) are anticipated with the use of softwood-based CelotexTM.

Shielding

The 9975 SARP, Revision 1 evaluated shielding of the 9975 package for determination of gamma and neutron dose rates under NCT and HAC. As for HAC, the CelotexTM properties are of no consequence since the modeling assumed total loss of packaging outside of the SCV. However, the NCT models did assume CelotexTM at a 0.20 g/cm³ cellulose density. This is based on a fiberboard density of 12.5 pcf. Since the chemical make-up of softwood-based CelotexTM is similar to cane-based CelotexTM and the fiberboard density is specified to be 14-16 pcf regardless of the base material, there is no impact to the shielding evaluation with the use of softwood-based CelotexTM in the 9975 package.

Conclusion

This paper has evaluated the impact of the use of softwood-based CelotexTM for a replacement for cane-based CelotexTM in terms of its chemical, structural, thermal, criticality, and shielding properties. In all aspects important to the 9975 package for transport, softwood-based CelotexTM from the Knight-Celotex Danville Plant is a suitable replacement for cane-based CelotexTM. It is the position of this paper that softwood- and cane-based CelotexTM, conforming to ASTM C208-95 (reapproved 2001), are equivalent materials and softwood-based CelotexTM should be approved as "equivalent" for use in fabrication of Model 9975 radioactive material packages.

References

- 1. *Safety Analysis Report for Packaging Model 9975*, Washington Savannah River Company, WSRC-SA-2002-00008, Revision 1, Aiken, SC (Pending Approval).
- 2. Standard Specification for Cellulosic Fiber Insulating Board, ASTM C208-95, American Society for Testing and Materials (Reapproved 2001).
- 3. Irving S. Goldstein, ed., *Wood Technology: Chemical Aspects*, American Chemical Society, Washington, D.C., pp. 20-21 & 196-198 (1977).
- 4. Adilson R. Goncalves, Elisa Esposito, and Priscila Benar, *Evaluation of Panus Tigrinus in the Delignification of Sugarcane Bagasse by FTIR-PCA and Pulp Properties*, Journal of Biotechnology, Volume 66, pp. 177-185, (1998).
- 5. P. Rezayati-Charani, J. Mohammadi-Rovshandeh, S. J. Hashemi, and S. Kazemi-Najafi, *Influence of Dimethyl Formamide Pulping of Bagasse on Pulp Properties*, Bioresource Technology, Volume 97, pp. 2435-2442, (2006).
- 6. J. X. Sun, X. F. Sun, R. C. Sun, and Y. Q. Su, *Fractional Extraction and Structural Characterization of Sugarcane Bagasse Hemicelluloses*, Carbohydrate Polymers, Volume 56, pp. 195-204, (2004).
- 7. Surinder Katyal, Kelly Thambimuthu, and Marjorie Valix, *Carbonisation of Bagasse in a Fixed Bed Reactor: Influence of Process Variables on Char Yield and Characteristics*, Renewable Energy, Volume 28, pp. 713-725, (2003).
- 8. Mari H. Nuopponen, Gellian M. Birch, Rob J. Sykes, Steve J. Lee, and Derek Stewart, *Estimation of Wood Density and Chemical Composition by Means of Diffuse Reflectance Mid-Infrared Fourier Transform (DRIFT-MIR) Spectroscopy*, Journal of Agriculture and Food Chemistry, Volume 54, pp. 34-40, (2006).
- 9. Mitsuru Sasaki, Tadafumi Adschiri, and Kunio Arai, *Fractionation of Sugarcane Bagasse by Hydrothermal Treatment*, Bioresource Technology, Volume 86, pp. 301-304 (2003).
- 10. C. F. Jenkins and P. R. Vormelker, *Celotex Fiberboard Insulation Chloride Concerns for KAMS 9975 Package*, SRT-MTS-994076, Washington Savannah River Company, Aiken, SC (August 1999).
- 11. K. H. Subramanian, *Corrosion of Lead Shielding in Model 9975 Package (U)*, WSRC-TR-2006-00094, Washington Savannah River Company, Aiken, SC (March 2006).
- 12. *Packaging and Transportation of Radioactive Material*, Code of Federal Regulations, Title 10, Part 71, U. S. NRC, Washington, DC (January 2006).

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- 13. Standard Test Methods for Cellulosic Fiber Insulating Board, ASTM C209-07, American Society for Testing and Materials (2007).
- 14. K. Raveendran, Anuradda Ganesh, and Kartic C. Khilar, *Pyrolysis Characteristics of Biomass and Biomass Components*, Fuel, Volume 75, pp. 987-998 (1996).

Attachment 1



"Christiansen, Craig" <cchristainsen@aknightcomp any.com> 10/22/2007 09:55 AM To <jason.varble@srnl.doe.gov>, <allen.smith@srnl.doe.gov> cc

Subject FW: density of premium sheathing

Guys I am still looking into the different tensile strengths of the board from the plants. Below is the weight of are board out of are Danville VA plant.

bcc

Craig Christiansen
Technical Support
cchristiansen@aknightcompany.com
Knight Industries, LLC
One Northfield Plaza, Suite 400
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(O) 847 999 2022
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Technical Support: 866-850-8836

www.knightcelotex.com

From: Clatterbuck, Tracey

Sent: Saturday, October 20, 2007 5:41 AM

To: Christiansen, Craig

Subject: RE: density of premium sheathing

For Premium Sheathing we are at 15 lbs/FT3 which is a weight of 625lbs/ft2.

Thanks TC White Paper: Demonstration of Equivalency of
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Attachment 2



Knight-Celotex Fiberboard™
a Knight Company

Material Safety Data Sheet Marrero Plant

SECTION I - IDENTIFICATION

Knight-Celotex LLC MSDS Number: 0006-84D One Northfield Plaza Suite 410 Effective Date: 04/19/2002

Northfield IL 60093-1215

Technical Questions: 1-800-596-9699

Trade Name: Fiberboard Insulation Board (Plain)

SECTION II - INGREDIENTS

Ingredient	CAS	%* Approx	OSHA PEL	ACGIH	Comments
	Number			TLV	
01 Cellulose	9004-34-6	≤99	5	5	mg/m³ Resp
02 Starch	9005-84-9	≤ 10	5	5	mg/m³ Resp
03 Other	NONE	< 1	NONE	NONE	

^{*%} Approximate

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Attachment 3



Material Safety Data Sheet Danville Plant

SECTION I - IDENTIFICATION

Knight-Celotex LLC One Northfield Plaza Suite 400 Northfield IL 60093-1215

Technical Questions: 800-596-9699

MSDS Number: 00040-85F Effective Date: 04/12/2005

Trade Name: Premium Fiberboard Insulating Sheathing, Manufactured Home USB, Coated 1 & 6 Sided

SECTION II - INGREDIENTS

Ingredient	CAS Number	%* Approx	OSHA PEL	ACGIH TLV	Comments
01 Cellulose	9004-34-6	<96	5	5	mg/m³ Resp
02 Starch	9005-84-9	<10	5	5	mg/m³ Resp
03 Clay	NONE	<2	5	5	mg/m ³ Resp
04 Carbon Black	1333-86-4	<1	3.5	3.5	mg/m³
05 Wax/Rosin	NONE	<2	NONE	NONE	
06 Laminating Adhesives	NONE	<3.5	NONE	NONE	

^{*%} Approximate