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Demonstration of Equivalency of Cane and Softwood Based CelotexTM for Model 9975 Shipping Packages

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ABSTRACT

Cane-based CelotexTM has been used extensively in various Department of Energy (DOE) packages as a thermal insulator and impact absorber. Cane-based CelotexTM fiberboard was only 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 fiberboard is no longer available for use in the manufacture of new shipping packages requiring the material as a component. Current consolidation plans for the DOE Complex require the procurement of several thousand new Model 9975 shipping packages requiring cane-based CelotexTM fiberboard. Therefore, an alternative to cane-based CelotexTM fiberboard is needed. Knight-Celotex currently manufactures CelotexTM fiberboard from other cellulosic materials, such as hardwood and softwood. A review of the relevant literature has shown that softwood-based CelotexTM meets all parameters important to the Model 9975 shipping package.

INTRODUCTION

This paper demonstrates softwood-based CelotexTM fiberboard from the Knight-Celotex Danville Plant has performance equivalent to cane-based CelotexTM for transportation in a Model 9975 shipping package. Continued fabrication of the Model 9975 shipping package is important to the DOE Complex Sites which have been given direction to consolidate DOE-STD-3013 compliant packaged materials to Savannah River Site (SRS). Although other certified shipping packages may exist which could meet the transportation needs of a consolidation mission, the SRS storage location requires the use of Model 9975 shipping packages. Without an alternative to cane-based CelotexTM fiberboard, repair of existing packages or fabrication of new packages with a design specification of cane-based CelotexTM fiberboard would be impacted.

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 CelotexTM from Sunbury Plant, Softwood-based CelotexTM from Danville Plant, and Cane-based CelotexTM from Marrero Plant.

DISCUSSION

The 9975 Safety Analysis Report for Packaging (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 pounds per cubic foot (pcf) specified for 9975 fabrication [3].

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 [4-7]. The average composition of the primary constituents (i.e. cellulose, hemicellulose, and lignin) for sugarcane bagasse and softwood is detailed below in Table 1.

	Sugarcane Bagasse [8]	Softwood [4, 9]	
Cellulose	26.6-54.3	40-51.4	
Hemicellulose	22.3-29.7	25-29	
Lignin	14.3-24.5	19.2-31	

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

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 per the manufacturer's MSDS. 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 [10]. 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 [11]. 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 [4]. 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 polyvinyl acetate (PVAc) glue used in laminating the sheets of CelotexTM [12]. 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 Normal Condition of Transport (NCT) and Hypothetical Accident Condition (HAC) testing as defined by 10CFR71 [1, 13]. 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 Primary Containment Vessel (PCV)/Secondary Containment Vessel (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.

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 as defined within ASTM C209-07 [14]. However, ASTM C208-95 (reapproved 2001) does not have any requirements as far as the compressibility of fiberboard. Slow strain rate testing conducted on softwood-based CelotexTM has shown significant agreement with compression testing of cane-based CelotexTM in the parallel and perpendicular orientations [15-16]. The culmination of all required ASTM C208-95 (reapproved 2001) testing, some limited slow strain rate testing of softwood-based CelotexTM, and density limitations as prescribed by the SARP, it is judged that softwood-based CelotexTM would not behave significantly differently than cane-based CelotexTM under NCT and HAC test.

<u>Thermal</u>

For the NCT insolation test, as described in 10CFR71.71(c)(1), thermal analytical modeling was conducted for purposes of the 9975 SARP with the prescribed insolation heat loads [1, 13]. 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) [17]. 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).

The thermal response of the 9975 shipping package with softwood-based CelotexTM is expected to be the essentially the same as cane-based CelotexTM. The thermal conductivity of fiberboard is required to be less than 0.40 BTU·in./h·ft²·°F at 75 ± 5 °F per ASTM C208-95 (reapproved 2001). Based on softwood-based CelotexTM testing, the thermal conductivity of softwood-based CelotexTM is within ASTM C208-95 (reapproved 2001) and within the variation reported for cane-based CelotexTM [1, 15, 18]. Similarly, the specific heat capacity of softwood-based CelotexTM is within the variation reported for cane-based CelotexTM [1, 15, 18]. Due to these similarities, along with

chemical composition and density, 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 10CFR71.73(c)(4), testing of a 9975 package was conducted as discussed in SARP, Revision 1, Appendix 3.5 [1,13]. 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 thermogravimetric analyzer (TGA) to study the pyrolysis characteristics of various biomasses [17]. In particular, bagasse and subabul wood, a softwood indigenous to Mexico, were ground to less than 250 μ m 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_{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_{eff} (i.e. an increase on k_{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 canebased 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.

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