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Humidity Data for 9975 Shipping Packages with Softwood Fiberboard

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Summary

The 9975 surveillance program is developing a technical basis to support extending the storage period of 9975 packages in K-Area Complex beyond the currently approved 15 years. A key element of this effort is developing a better understanding of degradation of the fiberboard assembly under storage conditions. This degradation is influenced greatly by the moisture content of the fiberboard, which is not well characterized on an individual package basis.

Direct measurements of humidity and fiberboard moisture content have been made on two test packages with softwood fiberboard and varying internal heat levels from 0 up to 19W. Comparable measurements with cane fiberboard have been reported previously. With an internal heat load, a temperature gradient in the fiberboard assembly leads to varying relative humidity in the air around the fiberboard. However, the absolute humidity tends to remain approximately constant throughout the package, especially at lower heat loads.

The moisture content of fiberboard varies under the influence of several phenomena. Changes in local fiberboard temperature (from an internal heat load) can cause fiberboard moisture changes through absorption or evaporation. And the moisture level within the package is constantly seeking equilibrium with that of the surrounding room air, which varies on a daily and seasonal basis as the package is not hermetically sealed. Fiberboard degradation at elevated temperature will produce water as a byproduct (as the cellulose breaks down, it is converted to water and carbon dioxide). Conversely, as degradation rates increase at elevated temperature, more of the water created from degradation will partition into the air (vs the fiberboard), leading to an increase in moisture escaping the package. This should partially limit the impact of the increased temperature.

One indicator of the moisture condition within a 9975 package might be obtained by measuring the relative humidity in the upper air space, by inserting a humidity probe through a caplug hole. This humidity correlates with the moisture level of the bottom fiberboard layers, although the trend starts to break down for extreme fiberboard moisture levels (above saturation), and for heat loads above 15W. The upper air gap relative humidity is currently measured during field surveillance of 9975 packages, but this is not done in the storage location. Once the package is moved to a different ambient temperature, the humidity measurement will no longer reflect fiberboard moisture conditions until equilibrium is re-established. NMM should consider whether surveillance measurements of the relative humidity (and temperature) of the upper air gap should be measured before the package is moved from the storage environment, or if there is value in making such measurements on additional (non-surveillance) packages. Such data could help assess the potential for mold growth or accelerated fiberboard degradation. However, without greater knowledge of the overall moisture content of a given package, application of such data could be limited.

Background

The 9975 surveillance program [1] includes elements to predict the service life of 9975 shipping packages used to store special nuclear materials in the K-Area Complex (KAC). One key area of inquiry is the potential degradation of the fiberboard overpack in these packages. The fiberboard contains moisture which can migrate within the package during storage under the influence of internal temperature gradients [2-4]. The moisture content of as-manufactured fiberboard is less than 10 wt% [5]. However, moisture is added when the fiberboard layers are laminated with water-based glue, and the moisture content can change during storage and handling as the fiberboard approaches equilibrium with the humidity of the surrounding environment. This process can continue after the overpack assembly is loaded into the 9975 package, although the 9975 drum provides a significant degree of isolation from the environment such that the rate of moisture exchange is greatly reduced [6]. In addition, moisture is created as the fiberboard degrades under the influence of elevated temperature and/or humidity [7].

The concentration of moisture can be sufficient to support the growth of mold. In addition, some constituents present in the fiberboard (such as chlorides) can leach out and concentrate with the moisture at levels that could lead to degradation of the stainless steel drum. Such behaviors have been observed in test packages with a nominal initial moisture level and a 19 watt internal heat load [3, 4], the maximum heat load for which the 9975 is approved. In order to better understand the degree to which packages in storage are susceptible to this behavior, two instrumented packages with softwood fiberboard assemblies have been prepared and tested to measure internal humidity profiles with varying ambient temperature, fiberboard moisture content and internal heat load. It is hoped that data from this effort will provide understanding to relate measurement of the relative humidity in the upper air space with moisture conditions in the fiberboard.

The test sequence duplicates work completed previously for cane fiberboard assemblies [8].

Temperature / Humidity Profile Data in Test Packages

Two 9975 test packages had been previously modified to allow placement of an internal heater (in a dummy 3013 container), several thermocouples throughout the package and several additional features for monitoring package component performance [2, 3]. The fiberboard in these packages was replaced, and the packages were further modified to provide channels for a humidity probe along the fiberboard ID and OD surfaces. These channels extend through the drum lid and are sealed with tape between measurements, to maintain normal patterns of air circulation within the package. The inner channel is fitted with a plug that extends from the lid into the lower fiberboard assembly to facilitate alignment and prevent air circulation from the inner air space to the upper air space above the upper fiberboard assembly. The configuration of these channels and placement of the thermocouples on the fiberboard are illustrated in Figure 1.

The fiberboard assemblies used for this effort are new assemblies fabricated from softwood fiberboard. Both had a typical initial moisture content of ~7 % wood moisture equivalent (WME), or ~9.4 wt% moisture. One assembly was placed into test with this moisture condition, while the other was held in a high humidity environment (enclosed in a plastic bag with a water source) until an average moisture content of ~8.2 %WME (~10.7 wt%) was reached. This moisture level was targeted to provide an example of elevated moisture similar to that which might be expected in service. It was expected that moisture segregation during testing will produce a range of moisture content throughout the fiberboard.

The packages are placed on a steel pallet (borrowed from KAC), to approximate the conditions for heat transfer through the drum bottom experienced in KAC. Once testing of each package begins, the package remains closed with periodic monitoring of internal temperature and humidity until an equilibrium condition is reached. Temperatures at the thermocouple locations (Figure 1) are recorded automatically. On approximately a weekly basis, a probe is placed into each channel and held at different elevations in 3 inch increments to record the temperature and relative humidity profiles along the fiberboard ID and OD surfaces.

The initial test condition includes no internal heat load for baseline data, followed by a 5 watt internal heat load. Internal temperatures fluctuate due to varying room ambient temperature. However, temperature and humidity gradients developed and stabilized within a few weeks. An insulating blanket was subsequently placed on the side and top of the drum to provide a slightly elevated temperature environment, thus extending the temperature range over which data could be collected. Fiberboard temperatures at the six thermocouple locations are shown for each package in Figures 2 and 3.

Once equilibrium data were collected with the 5W heat load (both with and without the insulating blanket) the package was opened to make limited direct measurements of the fiberboard dimensions and moisture content, and then the heat load was increased sequentially to 10, 15 and 19W and the process repeated at each heat load. Both temperature and relative humidity values can vary within the package, especially with an internal heat load. However, it is observed that the absolute humidity (a function of temperature and relative humidity) tends to be much more uniform throughout the package. Accordingly, typical absolute humidity data are used to illustrate the fiberboard behavior for the different environments in the two test packages. The following equation provides a conversion to absolute humidity with accuracy within 0.3% for temperatures up to 60 °C [9].

$$AH = 13.253*(10\^{}(7.5914*T/(T+240.73)))*RH/(273.15+T)$$

Where AH = absolute humidity (g/m3)
T = temperature (°C)
RH = relative humidity (%)

Typical profiles for all three parameters (temperature, relative humidity and absolute humidity) are shown in Figures 4 and 5.

In each package, the axial gap increased following each successive heat load. At the same time, the larger axial fiberboard dimensions tend to decrease as the heat load increases, although this trend varies at the lower heat loads for the drier package. This is seen in Figure 6 which compares the change in the lower fiberboard assembly height and the axial gap. The increase in axial gap is largely countered by a decrease in the height of the lower assembly.

Discussion

Following each internal heat load cycle, the packages were opened for inspection. In both packages, the fiberboard moisture content shifted such that the outer and lower regions gained water while the inner regions lost water. Package LE RH1 SW, with a nominal moisture content, had a maximum individual moisture reading of 18.2 %WME (21.2 wt%) on the bottom of the lower assembly after the 19 watt heat load. This moisture level is below the saturation level for cellulose (~28 wt%). Average moisture values for each region of fiberboard in LE RH1 SW are summarized in Figure 7a. No indication of mold growth was observed in this package.

Package LE RH2 SW, with a slightly elevated moisture content, exceeded the saturation point on the bottom fiberboard layer after 19 watts, with a maximum moisture reading of 75.4 %WME. Average moisture values for each region of fiberboard in LE RH2 SW are summarized in Figure 7b. No indication of mold growth was observed in this package.

Reference 10 identifies several packages that were removed from storage in KAC with elevated moisture and/or mold present in the fiberboard. Mold was observed but appeared dormant in two of these packages with measured moisture content up to 19 %WME. Mold appeared active in one package with measured moisture content up to 24 %WME. (All of these packages contained cane fiberboard.) This suggests that the local moisture content needs to be at least ~20 %WME (16 wt%) for active mold growth. Similarly, Reference 11 identifies that most fungal activity is inhibited when the equilibrium relative humidity drops below 70%, which corresponds to 16 wt% moisture for typical softwoods. The data indicate that the higher heat load conditions within these two packages were conducive to mold growth. It is assumed that the absence of observed mold is a result of the relatively short duration spent at these conditions. However, it should be assumed that packages in long-term storage with heat loads approaching 19W are capable of developing mold in the lower fiberboard layers.

The average absolute humidity is plotted as a function of temperature in Figure 8. OD and ID results for each package are plotted separately. Some of the scatter in these graphs results from the varying internal heat load, especially for the ID surface. Additional scatter is introduced as a result of occasional transient conditions, such as changes in the ambient temperature. A similar slope is seen at each internal heat load for each package in Figure 8, although the data subset for each heat load is offset slightly from the other heat loads. This might result from changes in the relative rates of water loss and water generation at each condition, or the relative partitioning of water between the fiberboard and air at a given temperature.

Fiberboard and Moisture

A given moisture content will register differently on the moisture meter for cane and softwood fiberboard, making it necessary to convert the measurements from %WME to wt% to make quantitative comparisons. The conversions have been previously developed and reported [12, 13] and are applicable only to moisture content below saturation (~28 wt%):

wt% moisture = 0.67 * %WME + 2.6 (cane fiberboard)

wt% moisture = 1.02 * %WME + 2.3 (softwood fiberboard)

When manufactured, ASTM C208-95 specifies a maximum fiberboard moisture content of 10 wt% (~7.5 %WME for softwood fiberboard, ~11 %WME for cane fiberboard). However, there are no requirements to control the moisture content subsequently. During fabrication of the fiberboard assembly, the layers are laminated with water-based glue, which can increase the moisture content of the assembly.

Since most of the 9975 packages in storage were manufactured, loaded with a modest heat load and placed in storage within a relatively short time, it is likely that the majority have remained close to their initial fiberboard moisture value, although a range of moisture levels will exist. This is supported by destructive examination data from the 9975 surveillance program for which typical fiberboard moisture content ranges from about 9 to 16 %WME for cane fiberboard (~9 to 13 wt%). This range corresponds to ~6.6 to 10.5 %WME for softwood fiberboard. Similar values are often seen in field surveillance of 9975 packages in KAC, but since these measurements exclude much of the lower fiberboard assembly, they are less conclusive as to the overall moisture content.

Published data for wood in an outdoor atmosphere show the equilibrium moisture content will vary seasonally between 12 and 14 wt% (~9.5 – 11.5 %WME for softwood fiberboard, ~14 – 17 %WME for cane fiberboard) in this area (taken as an average of reported behavior for Columbia, SC and Augusta, Ga) [14]. Since cellulose is the primary constituent of wood and fiberboard (both cane and softwood based) products, it is expected that fiberboard will behave similarly. However, with indoor storage and at least modest climate control (heating and cooling), the higher humidity levels will be reduced somewhat. Therefore, it is expected that conforming packages will not experience significant moisture gain from the environment while in approved storage conditions.

The chemical formula for cellulose is $(C_6H_{10}O_5)_n$. As fiberboard degrades, the cellulose it contains is converted to CO_2 and water. In this reaction, an additional 6 moles of oxygen (from the air) are required to produce 6 moles of CO_2 and 5 moles of water from 1 mole of cellulose. If the water is in liquid form or absorbed into the remaining cellulose, then there is no net increase in volume of gaseous compounds. However, some of the water will likely evaporate as the fiberboard comes into equilibrium with moisture in the air. This added gas volume will create a driving force to move air (including humidity) out of the package. If all the available hydrogen converts to water, then each gram of degraded fiberboard will produce 0.56 gram water.

In addition to internal gas creation, atmospheric pressure changes outside of the drum will drive air in or out of the drum toward an equilibrium condition. There are several potential leak paths through which these exchanges might occur, including between the drum flange and lid, around the caplugs, and through the rolled bottom edge of the drum.

In summary, there are several mechanisms that can affect the moisture content of the fiberboard within a 9975 package. Each of these mechanisms can be active at different times during fabrication, handling and storage of the package. They include:

- Fabrication of the fiberboard assembly with water-based wood glue can elevate the moisture content.

- The fiberboard moisture content will increase or decrease to approach equilibrium with the surrounding environment. However, with the drum providing a degree of isolation from the room ambient environment, this change will be very slow.
- With an internal heat load and temperature gradient across the fiberboard, the local relative humidity within the package will vary even though the absolute humidity tends to remain constant. For a given moisture content of the adjacent air, the fiberboard moisture content will decrease as the temperature increases (and relative humidity decreases). Therefore, the fiberboard moisture content will change with the local fiberboard temperature.
- Daily or seasonal fluctuations in the ambient temperature will affect the drum surface temperature. If the relative humidity adjacent to the drum interior surface is high, a decrease in room temperature can cause moisture to condense on the drum interior. This condensation will preferentially run to the bottom of the drum and be absorbed by the bottom fiberboard layers.
- As the fiberboard slowly degrades in service (at a rate determined by temperature and moisture level), additional water is produced as a byproduct. As additional volume of gas byproduct is produced, a corresponding amount of (humid) air will escape from the drum.

For packages in storage, the heat load is known, and the ambient temperature can be estimated. However, the overall moisture content of the fiberboard is not known for most packages. For higher fiberboard moisture content, fiberboard degradation rates will increase (if the temperature goes above a threshold value) and the likelihood of mold growth will increase. Measurement of the upper air gap relative humidity through a caplug hole is one means to gain some information about the fiberboard moisture content. However, measurement of this parameter within the surveillance program (after the package has been removed from the storage environment) has shown that relative humidity values can change significantly when the package is moved to an area with different ambient temperature. In such cases, the humidity measurement will not reflect fiberboard moisture conditions until equilibrium is re-established.

Implications for Packages in Storage

The overall behavior of moisture within softwood fiberboard is similar to that seen for cane fiberboard [8]. With both fiberboard materials, the moisture within the air spaces throughout the package tends to maintain a constant value of absolute humidity, while the relative humidity will vary inversely to the temperature gradient within the package. The moisture absorbed within the fiberboard will come to equilibrium with the adjacent air, but this equilibrium level varies with temperature.

Figure 9 shows the correlation between absolute humidity measured in the upper air gap and the moisture content along the fiberboard OD surface for both softwood and cane fiberboard. Three separate plots are shown for fiberboard moisture content at different elevations. In this figure, moisture content is expressed as wt% to allow direct comparison between the two materials. For a given overall fiberboard moisture content, both the moisture content at the fiberboard OD and the absolute humidity in the upper air gap increase as the heat load increases. This pattern starts to break down for heat loads above 15W, and where local fiberboard moisture exceeds saturation.

NMM should consider whether surveillance measurements of the relative humidity (and temperature) of the upper air gap should be made before the package is moved from the storage environment, and if there is value in making such measurements on additional (non-surveillance) packages in-situ. Such data could help assess the overall moisture content of the package and the potential for mold growth or accelerated degradation. However, with only limited knowledge of the overall moisture content of a given package, application of such data could be difficult.

Several trends are common in all four test packages (with cane and softwood fiberboard).
- In each test package, the overall weight decreases as the heat load increases. This weight loss comes primarily from the fiberboard and is a result of moisture loss (Figure 10).
- The average absolute humidity in each package increases with the heat load up to 15W, and then decreases at 19W (Figure 11).
- The average absolute humidity increases with the insulating blanket vs the bare package (Figure 11).
- The average fiberboard temperature increases with the heat load, and increases further when an insulating blanket is added to the package (Figure 12). This effect is lessened in going from 15W to 19W due to a coincident seasonal decrease in ambient temperature.
- The temperature gradient across the fiberboard is approximately constant for a given heat load, with or without the insulating blanket.

These observations suggest that when surrounded by air of constant absolute humidity, the equilibrium fiberboard moisture content decreases as the temperature increases. In other words, as the fiberboard temperature increases, more of the moisture it contains (and more of the moisture created by fiberboard degradation) is partitioned to the surrounding air. This sets up a feedback mechanism to partially limit fiberboard degradation at higher temperatures. As the absolute humidity within the package increases, there is a larger driving force for moisture to escape the package. This can happen by two mechanisms: as more water vapor is released into the air, the increased pressure inside the drum will force some air to escape, and with higher humidity in the air, more moisture will escape as the package "breathes" with cyclic ambient room pressure variation. The net effect of these mechanisms is manifest in the observation that the average absolute humidity increases with heat load up to 15W and then decreases at 19W.

At the highest heat loads, this effect may lead to lower total fiberboard moisture content as the degradation rate increases, which may act to limit the increase in degradation rates. Note, however, this pattern may be disrupted (at least temporarily) if condensation or other mechanisms allow liquid water to collect in the lower fiberboard layers, as has been observed in some packages.

Conclusions

Two test packages have been assembled to provide a correlation between humidity and fiberboard moisture levels within the package, and moisture gradients throughout the fiberboard assembly for different internal heat loads. This effort has examined packages with softwood fiberboard and internal heat levels ranging from 0 to 19W. A similar effort previously looked at the same correlation with cane fiberboard. The fiberboard in each package had a different initial moisture content, and developed a gradient in relative humidity related to the internal heat load.

The absolute humidity tends to remain approximately constant within the package for a given condition. A correlation is seen between the relative humidity in the upper air space and the moisture content of the fiberboard assembly, although this correlation is relatively crude without knowledge of the overall moisture content of the package. The moisture content of the fiberboard will tend to increase in the bottom layers for higher heat loads and higher overall moisture content.

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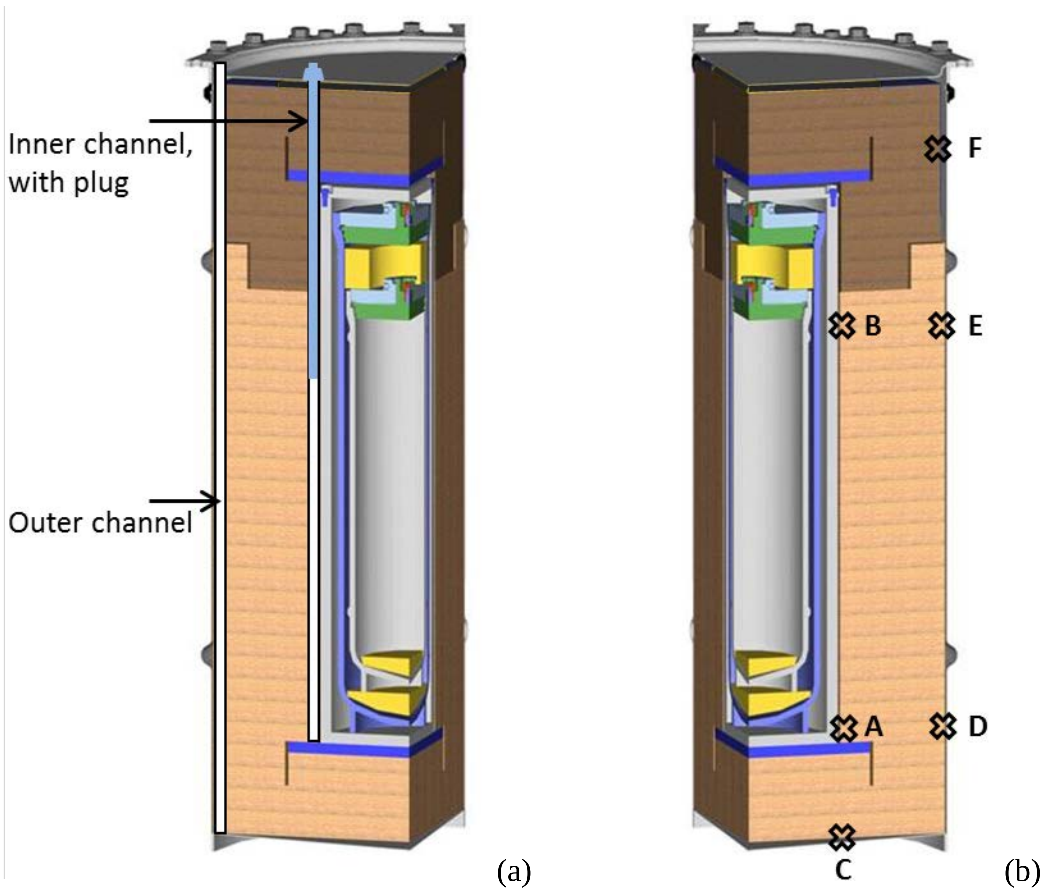


Figure 1. Location of channels to measure relative humidity (a), and location of thermocouples in humidity test package fiberboard assembly (b)

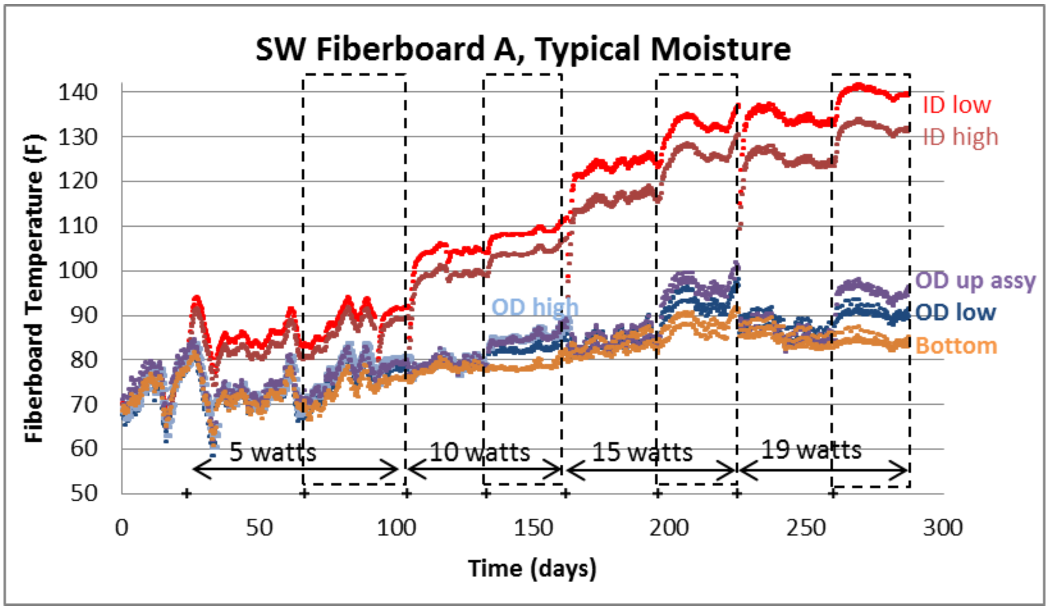


Figure 2. Temperature history for the six thermocouple locations within LE RH1 softwood fiberboard assembly. An insulating blanket was placed around the package during the periods within the dashed boxes.

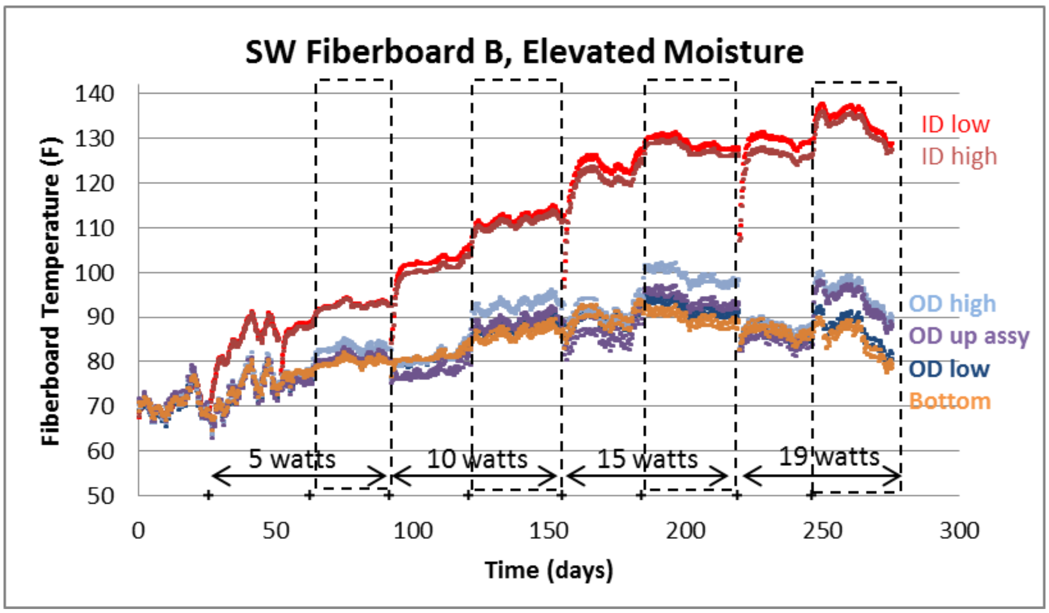
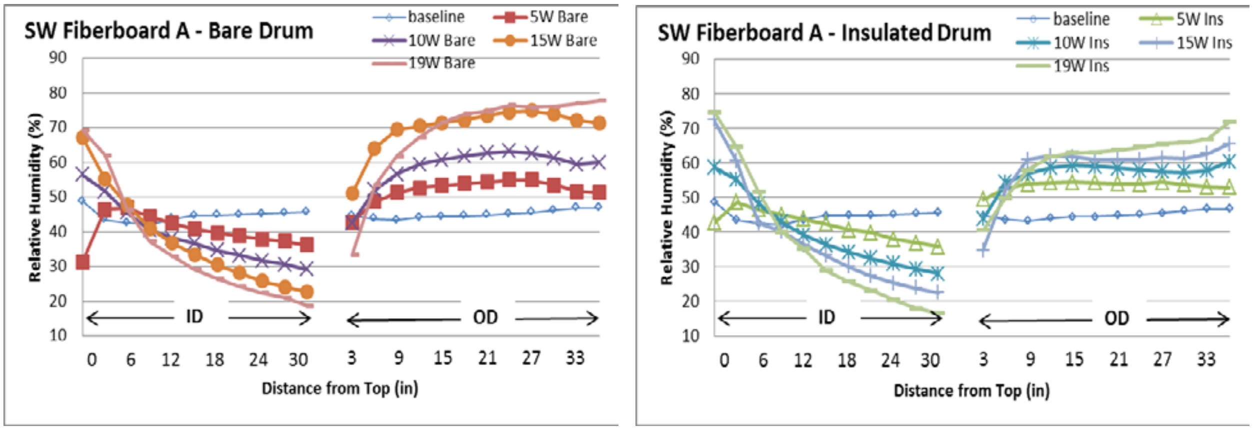
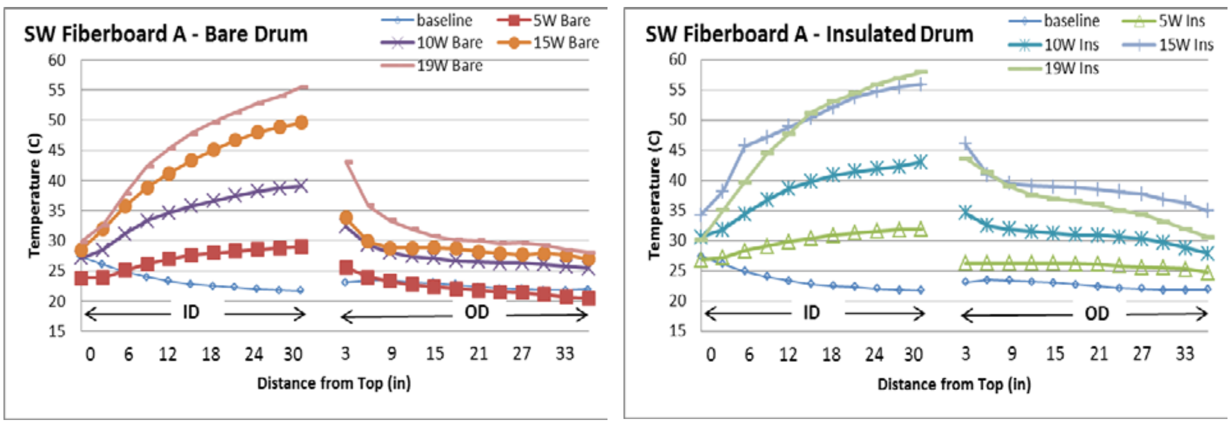


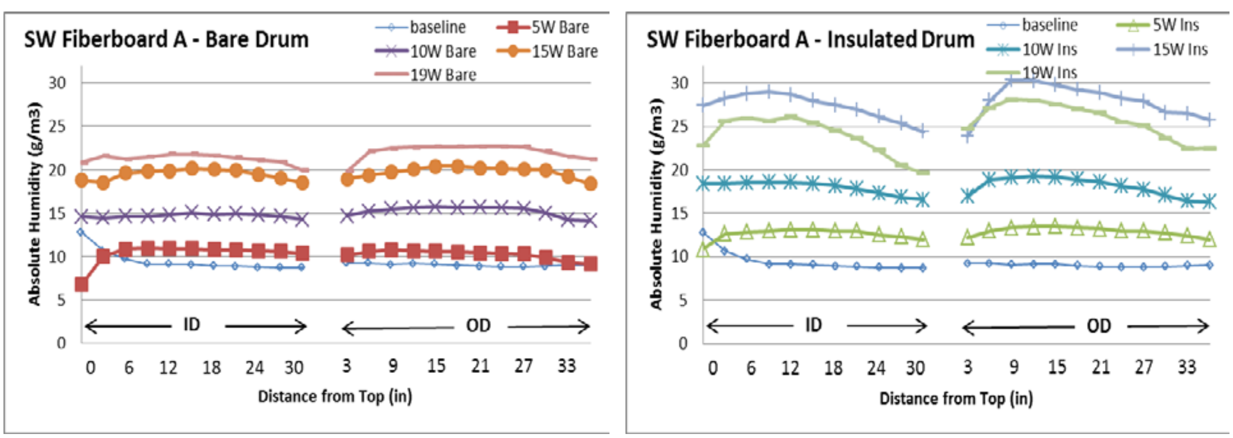
Figure 3. Temperature history for the six thermocouple locations within LE RH2 softwood fiberboard assembly. An insulating blanket was placed around the package during the periods within the dashed boxes.



(a) Relative humidity profiles

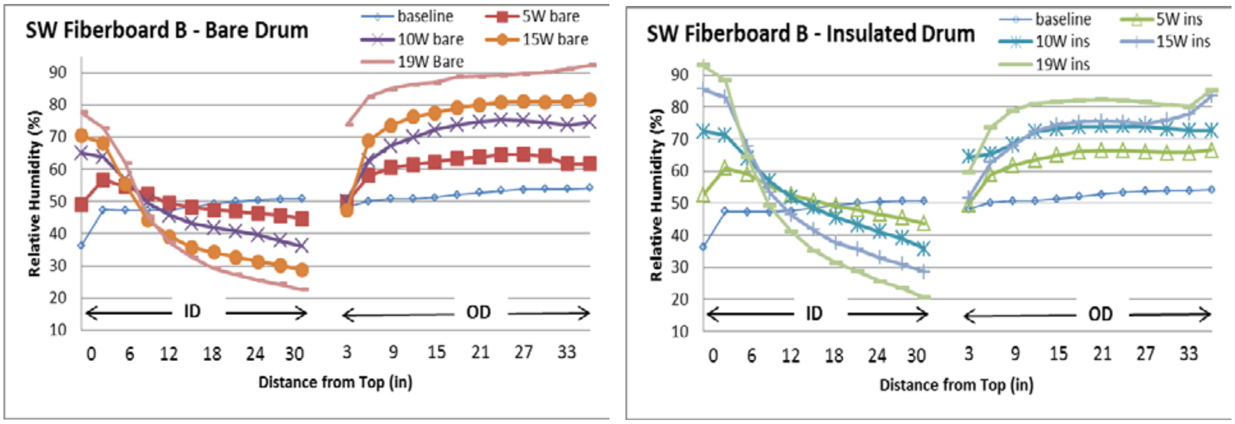


(b) Temperature profiles

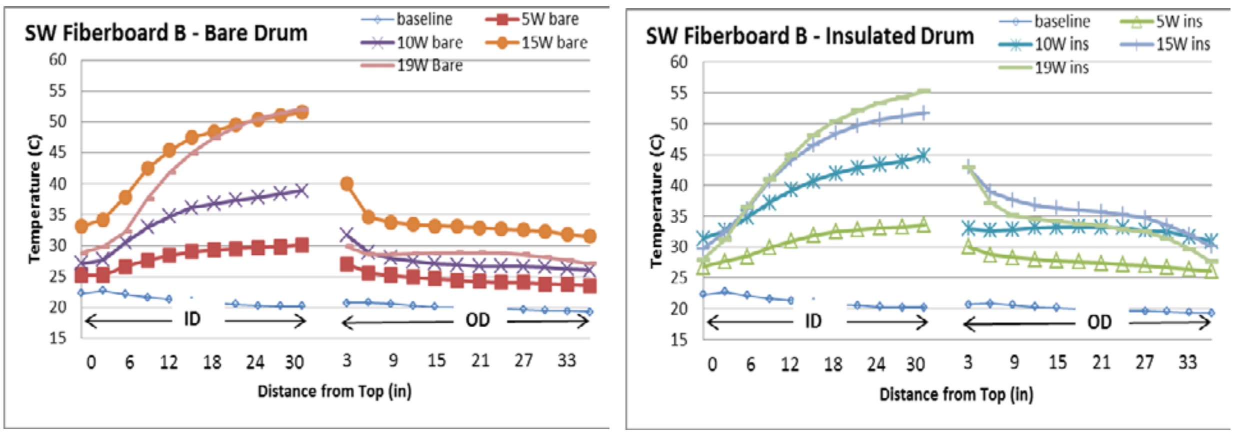


(c) Absolute humidity profiles

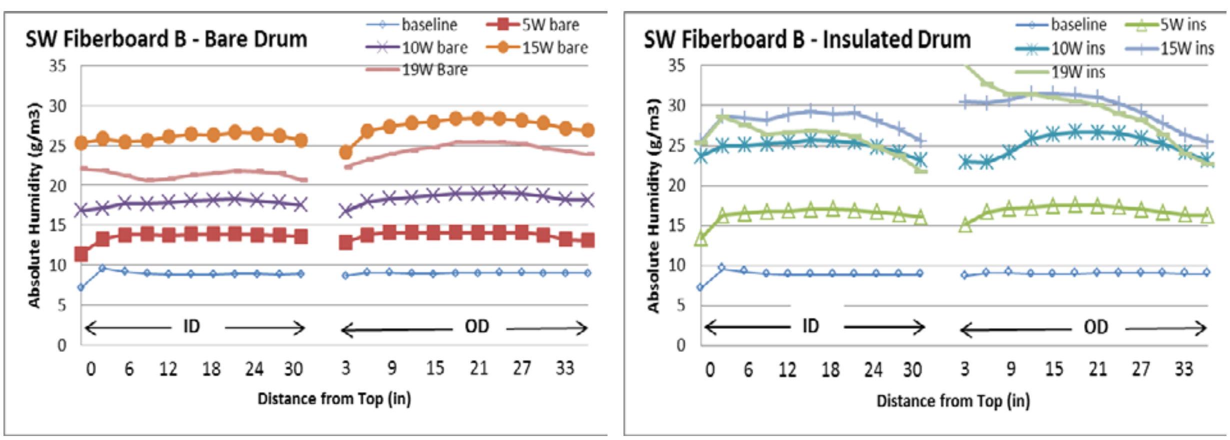
Figure 4. Profiles for package LE RH1 softwood A fiberboard (nominal moisture content). Each graph shows typical measurements along the ID and OD surfaces of the fiberboard. Profiles are graphed separately for measurements on the bare package and the insulated package for clarity.



(a) Relative humidity profiles



(b) Temperature profiles



(c) Absolute humidity profiles

Figure 5. Typical profiles for package LE RH2 softwood B fiberboard (elevated moisture content). Each graph shows typical measurements along the ID and OD surfaces of the fiberboard. Profiles are graphed separately for measurements on the bare package and the insulated package for clarity.

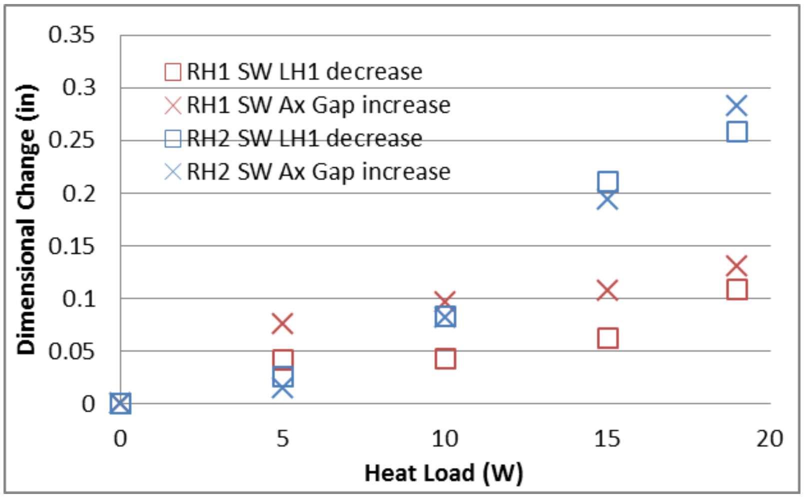


Figure 6. Decrease in lower fiberboard assembly height (LH1) compared to axial gap increase.

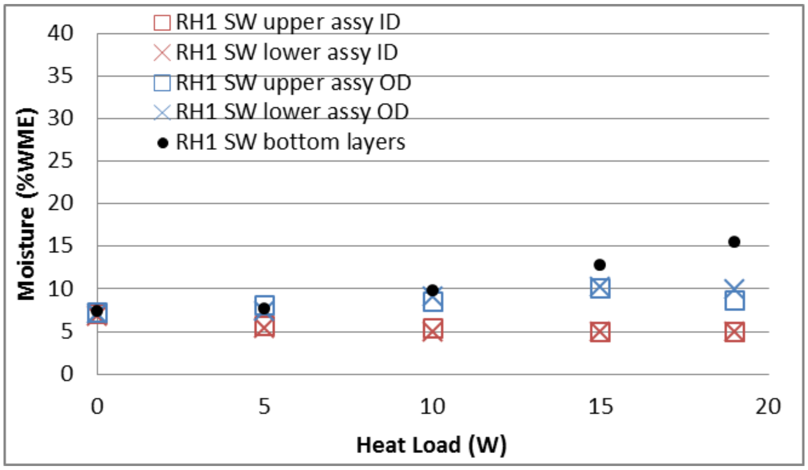
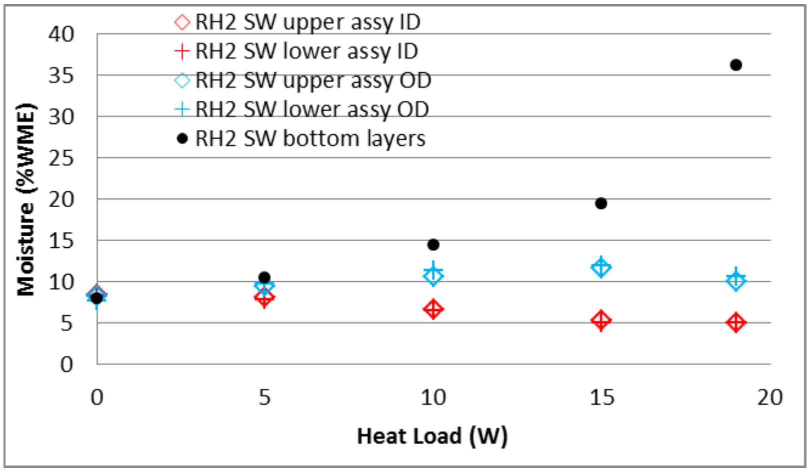


Figure 7. Average moisture values for each fiberboard region

(a) LE RH1 SW



(b) LE RH2 SW

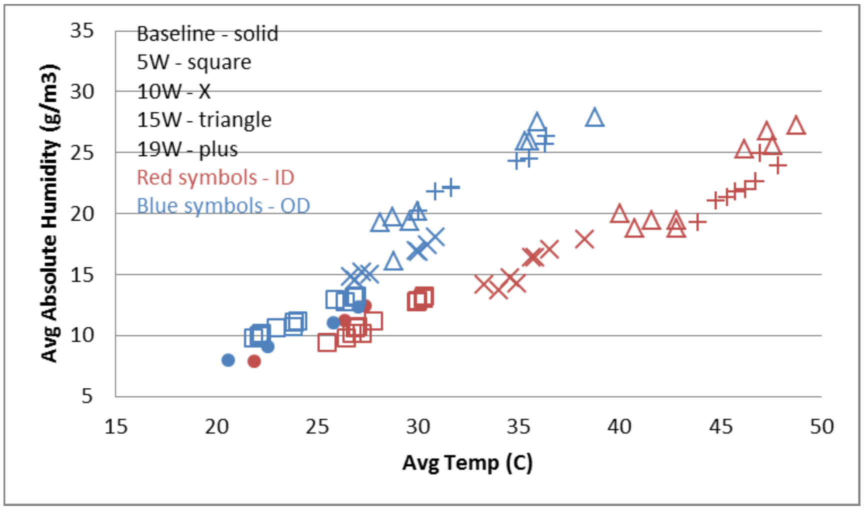
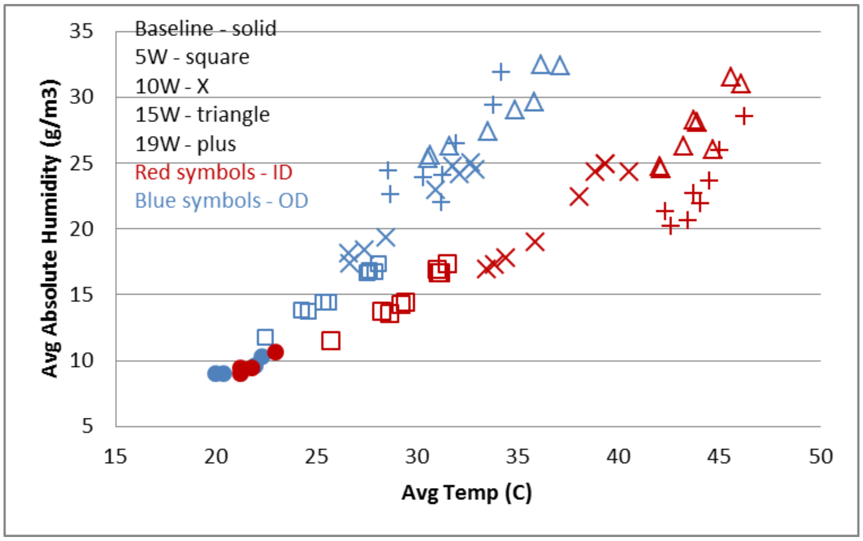


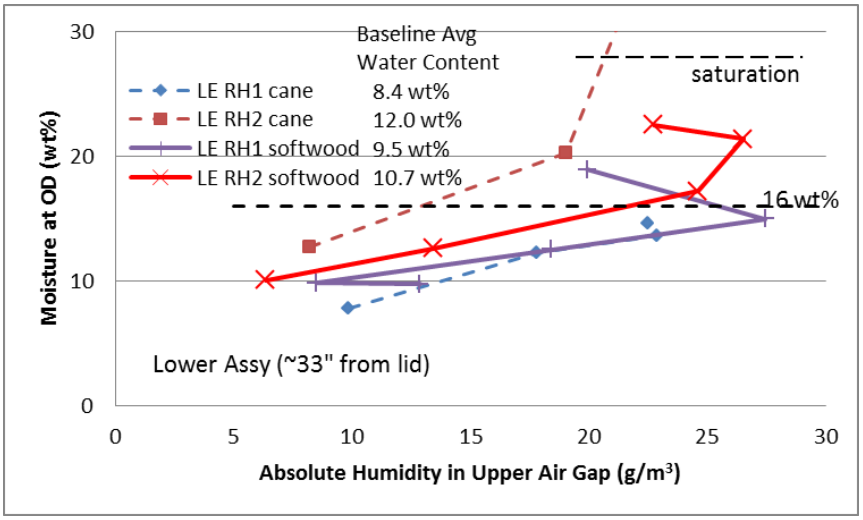
Figure 8. Average absolute humidity vs temperature for all LE RH1 SW data (a) and all LE RH2 SW data (b). Results are shown separately in each graph for OD and ID, and for each heat load.

(a) LE RH1 softwood

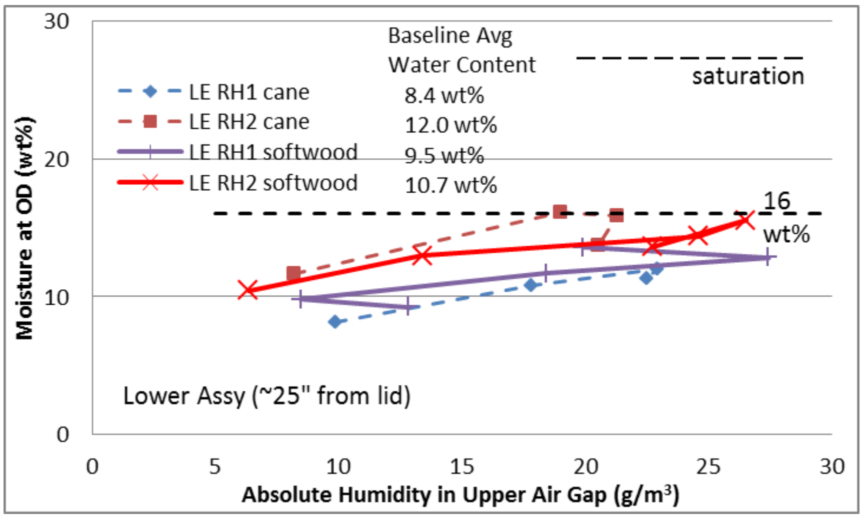


(b) LE RH2 softwood

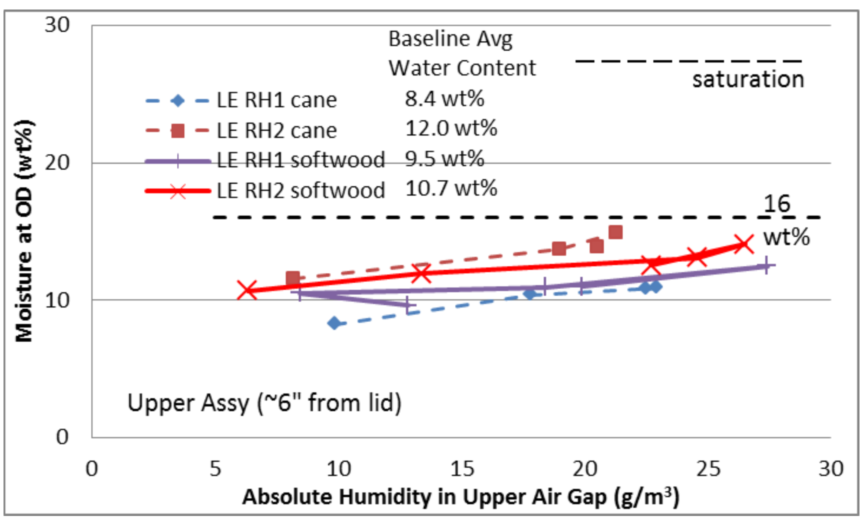
Figure 9. Correlation between absolute humidity in the upper air gap and the fiberboard moisture content at the OD surface for softwood and cane fiberboard.



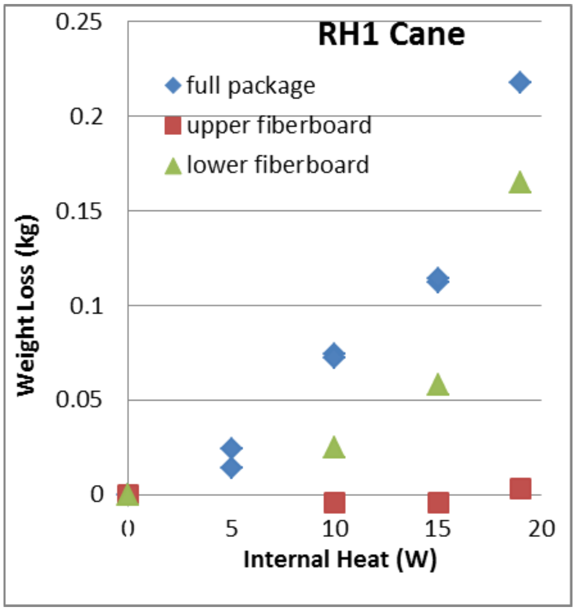
(a) Lower assembly OD, ~33” below the lid



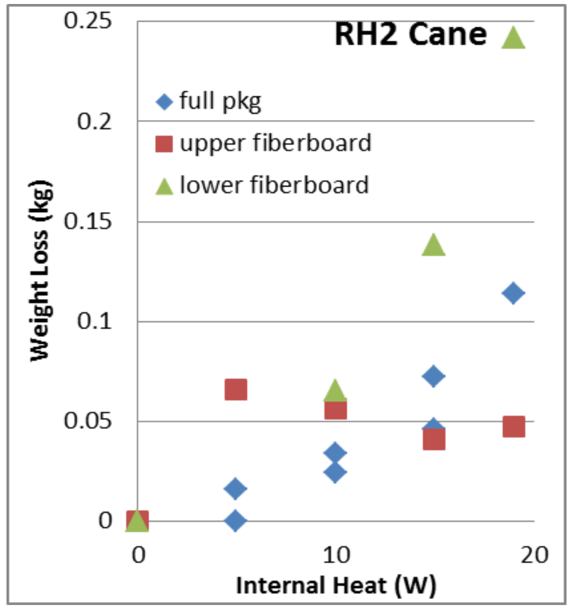
(b) Lower assembly OD, ~25” below the lid



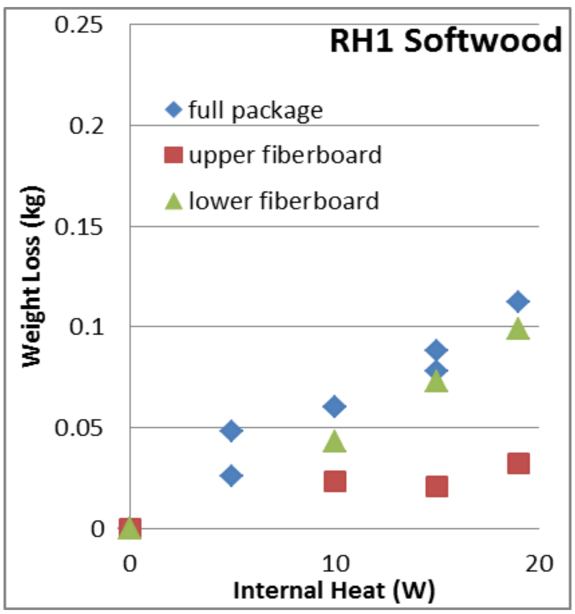
(c) Upper assembly OD, ~6” below the lid



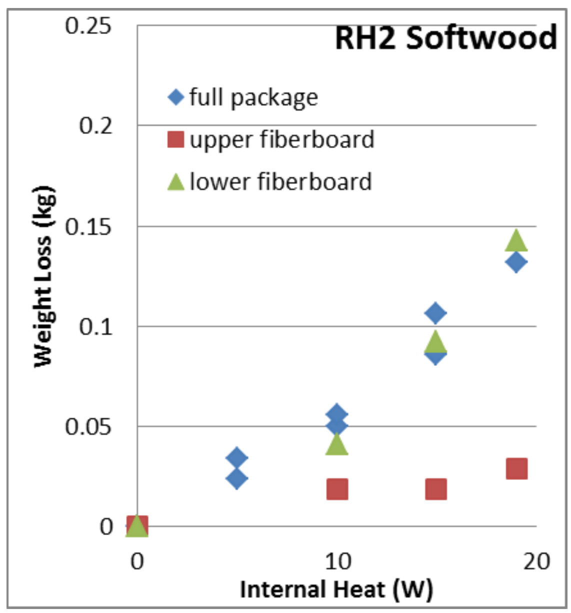
(a) Pkg RH1 (cane, nominal moisture)



(b) Pkg RH2 (cane, elevated moisture)

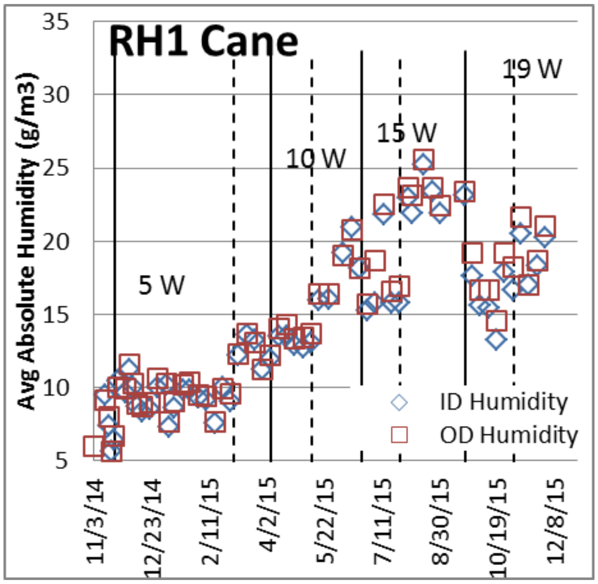


(c) Pkg RH1 (softwood, nominal moisture)

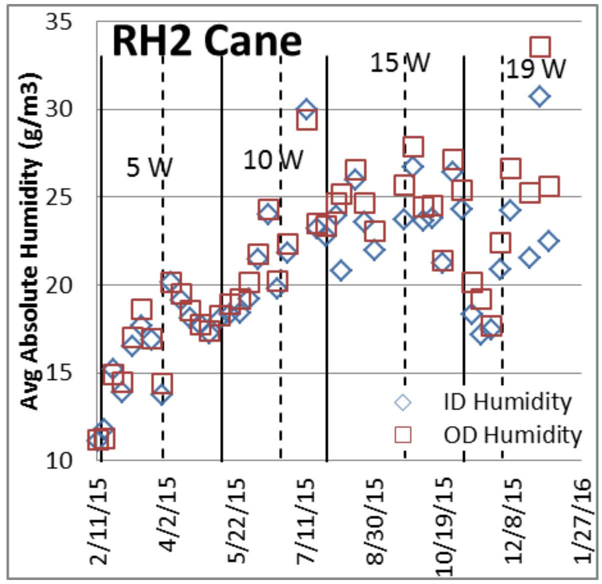


(d) Pkg RH2 (softwood, elevated moisture)

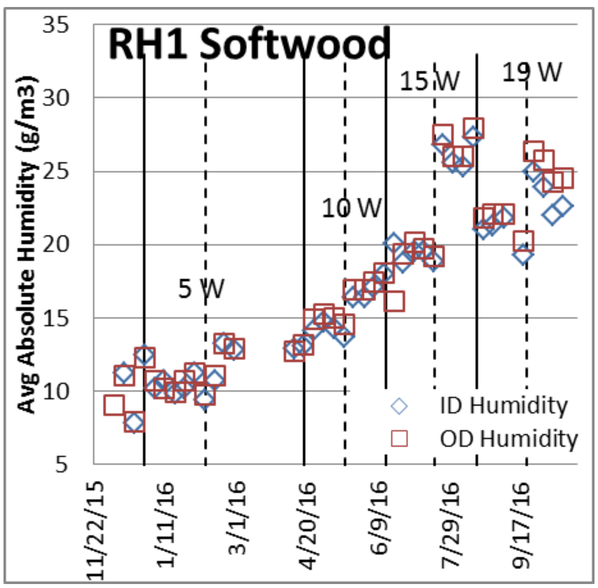
Figure 10. Weight loss for each package based on total package weight and each fiberboard assembly weight.



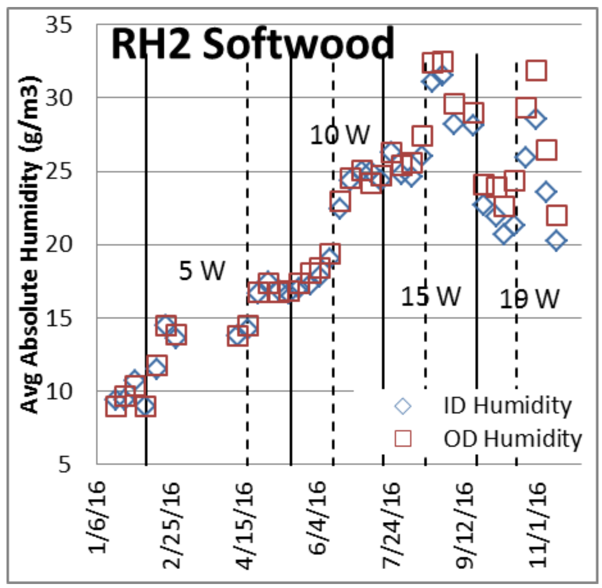
(a) Pkg RH1 (cane, nominal moisture)



(b) Pkg RH2 (cane, elevated moisture)

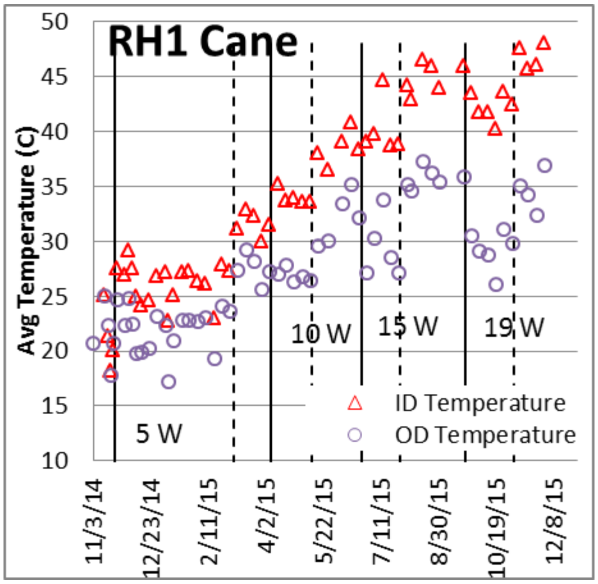


(c) Pkg RH1 (softwood, nominal moisture)

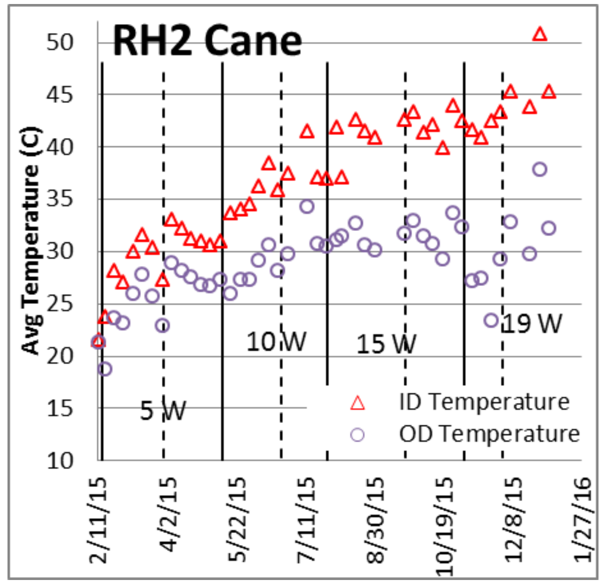


(d) Pkg RH2 (softwood, elevated moisture)

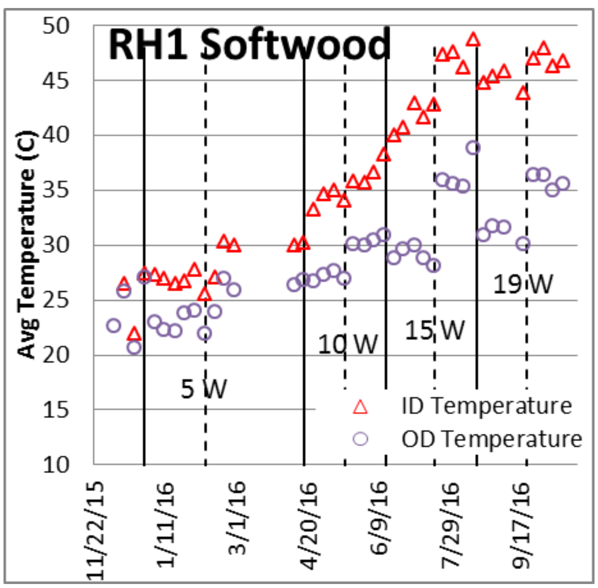
Figure 11. Absolute humidity for each package, showing separate average values along the package ID and the package OD. The solid vertical lines indicate times when the internal heat load was increased. The dashed vertical lines indicate times when an insulating blanket was added to the package.



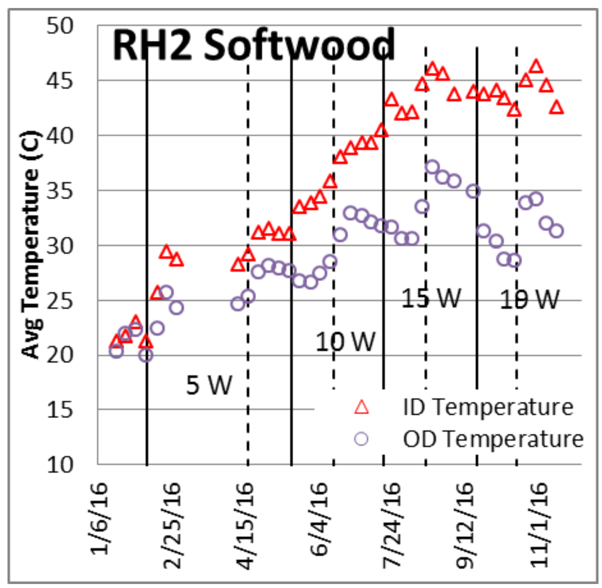
(a) Pkg RH1 (cane, nominal moisture)



(b) Pkg RH2 (cane, elevated moisture)



(c) Pkg RH1 (softwood, nominal moisture)



(d) Pkg RH2 (softwood, elevated moisture)

Figure 12. Average temperature for each package, showing separate average values along the package ID and the package OD. The solid vertical lines indicate times when the internal heat load was increased. The dashed vertical lines indicate times when an insulating blanket was added to the package.

Distribution

G. A. Abramczyk, 730-A
R. J., Bayer, 705-K
J. S. Bellamy, 730-A
W. L. Daugherty, 773-A
B. A. Eberhard, 105-K
B. L. Garcia-Diaz, 999-2W
L. F. Gelder, 999-W
T. W. Griffin, 705-K
R. J. Grimm, 705-K
E. R. Hackney, 705-K
S. J. Hensel, 705-K
J. M. Jordan, 705-K
B. B. Kiflu, 705-K
L. L. Kyriazidis, 705-K
D. R. Leduc, 730-A
P. L. Livengood, 705-K
J. W. McEvoy, 707-C
T. E. Skidmore, 730-A
D. E. Welliver, 705-K
K. E. Zeigler, 773-41A
Document Control