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Humidity Data for 9975 Shipping Packages with Cane 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 cane fiberboard and varying internal heat levels from 0 up to 19W. 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.

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. Fiberboard degradation at elevated temperature will produce water as a byproduct. 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.

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. However, the data indicate that for the higher internal heat loads (15 and 19 watts), a large variation in internal moisture conditions produces little or no variation in the air space relative humidity. Therefore, this approach does not appear to be sensitive to fiberboard moisture variations at the higher heat loads which are of most interest to maintaining fiberboard integrity.

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 cane 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 is being repeated with softwood fiberboard assemblies, which will be reported at a later date.

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 came from packages that had been removed from service due to NCR conditions not relating to the fiberboard (9975-03892 and 9975-03449) and are fabricated from cane fiberboard. Both had a typical initial moisture content of ~8 – 9 % wood moisture equivalent (WME), or ~8 – 8.6 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 a moisture content of ~13 – 15 %WME (~11.3 – 12.6 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 two locations (at the bottom of the ID surface, and on the upper assembly OD surface) 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 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 [8].

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. Since the baseline axial gap was not recorded, and the lower fiberboard assembly was not measured after 5 watts, the Figure 6 data are referenced to the 10 watt condition.

Discussion

For packages in storage, the heat load is known, and the ambient temperature can be estimated. However, the moisture content of the fiberboard is not known for most packages. When manufactured, ASTM C208-95 specifies a maximum fiberboard moisture content of 10 wt% (~11 %WME for cane fiberboard). However, there are no requirements to control the moisture content subsequently. There are several mechanisms that can affect the moisture content of the fiberboard within a 9975 package, including:

- The fiberboard moisture content will tend 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.
- After manufacture, the fiberboard layers are laminated with water-based glue, which provides an increase in the moisture content of the assembly.
- 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 in the annulus between the drum and fiberboard is high, a decrease in room temperature can cause moisture to condense on the drum inside surface. 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.

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, with a nominal moisture content, had a maximum individual moisture reading of 36 %WME (on the bottom of the lower assembly) after the 19 watt heat load. This moisture level is just below the saturation level for cellulose (~39 %WME, or 28 wt%). Average moisture values for each region of fiberboard in LE RH1 are summarized in Figure 7a. No indication of mold growth was observed in this package, although it might be expected had the higher heat load conditions been maintained for a longer time.

Package LE RH2, with a slightly elevated moisture content, exceeded the saturation point on the bottom fiberboard layer after 10 watts, with a maximum moisture reading of 42.3 %WME. Average moisture values for each region of fiberboard in LE RH2 are summarized in Figure 7b. Mold was noted near the bottom of the lower fiberboard assembly after 15 watts (Figure 8).

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 (~9 to 13 wt%). Similar values are also seen in field surveillance of 9975 packages in KAC, but since these measurements do not include 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% (~14 – 17 %WME for cane fiberboard) in this area (taken as an average of reported behavior for Columbia SC and Augusta, Ga) [9]. 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 average absolute humidity is plotted as a function of temperature in Figure 9. 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 significant changes in the ambient temperature in the 24 hours before measurement. A similar slope is seen at each internal heat load for each package in Figure 9, although the data for some heat loads is offset from that of the other heat loads. This might result from changes in the relative rates of water loss and water generation at each condition.

During these tests, the ambient temperature generally varied between 65 and 85 °F, which is consistent with the overall average KAC storage facility ambient temperature of 74 °F [10]. Reference 11 identifies that the fiberboard degradation rate generally becomes negligible below ~120 °F, and increases above this threshold temperature. The hotter fiberboard regions of these test packages exceeded ~120 °F when the internal heat load was 15 watts or greater, with maximum temperatures of ~140 °F. This comparison would apply to packages in KAC along the edges of an array subject to the room ambient temperature. Packages deeper in the array can be up to 11 °F hotter [10]. For such packages within the array, the hottest fiberboard region could be ~120 °F with an internal heat load as low as ~10 watts. Note, however, that the degradation rates at these temperatures are relatively low and would apply only to a limited region along the fiberboard ID surface.

Figure 2 shows the lower ID region of package LE RH1 was ~130 °F or greater for 18 weeks. During this period, the upper ID thermocouple read about 10 degrees cooler, and the thermal gradient through the sidewall was about 40 degrees. Considering just this sidewall region of the lower fiberboard assembly, this conservatively defines a volume of approximately 2860 cm^3 (or ~800 g of dry fiberboard) that was ~120 °F or greater. Assuming an average temperature within this volume of 125 °F, the weight of this region is decreasing at 0.19 %/year [11], or 1.5 g/year. Over the 18 week period, this package would have lost ~0.5 g from this region due to fiberboard degradation. The chemical formula for cellulose is $(C_6H_{10}O_5)_n$. If this compound is completely degraded, CO_2 and water are produced. If all the available hydrogen is converted to water, then the half-gram of degraded fiberboard produced 0.28 gram water. The amount of water production during this limited period is very small. However, for a package in storage with higher internal temperatures, the amount of water produced over the long term could be appreciable.

The water and CO_2 degradation products are gaseous, and create a driving force to move a similar volume of air plus moisture out of the drum. In addition, atmospheric pressure changes outside of the drum will drive air in or out of the drum toward an equilibrium condition. There are several 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. The intermittent stitch welds along the rolled bottom edge would create residual stresses that can open a larger leak path at the ends of these welds.

Figure 10 shows the correlation between the relative humidity measured in the upper air gap and the moisture content of the lower fiberboard layers. For each heat load, there are only two data points (one from each package) since the moisture content of the fiberboard is measured only when the package is opened for inspection. As a result, there is no indication of overall scatter in this correlation. At lower heat loads (0 and 10 watts), the slopes suggest a prediction of the moisture in the lower fiberboard layers might be made based on measuring the relative humidity in the upper air gap. However, this would not be practical at the higher heat loads (15 and 19 watts) since there is a large variation in the moisture content of the lower fiberboard layers for essentially the same humidity reading in the upper air gap. Since packages with higher heat loads are of greatest concern for concentrating moisture in the bottom fiberboard layers, the relative humidity of the upper air gap does not appear to offer a convenient indicator of this condition.

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 cane fiberboard and internal heat levels ranging from 0 to 19W. 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.

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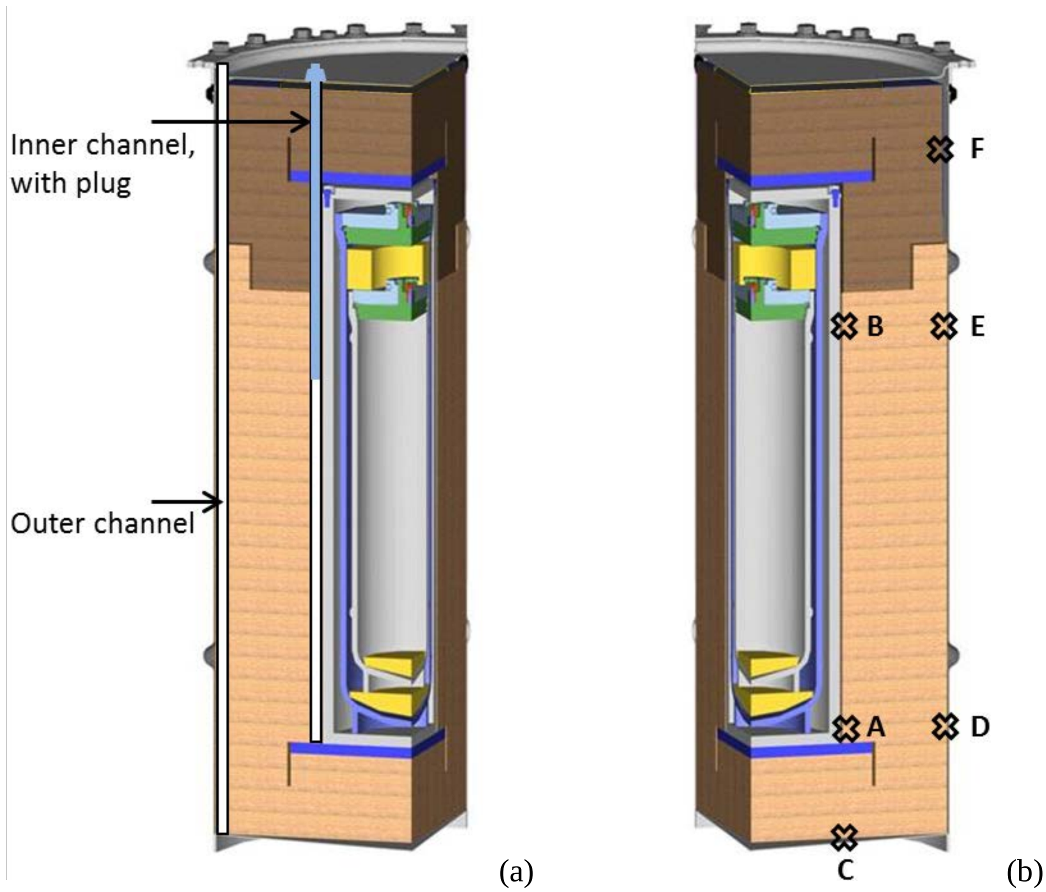


Figure 1. Location of channels to measure relative humidity (a), and location of thermocouples in humidity test package fiberboard assembly (b)

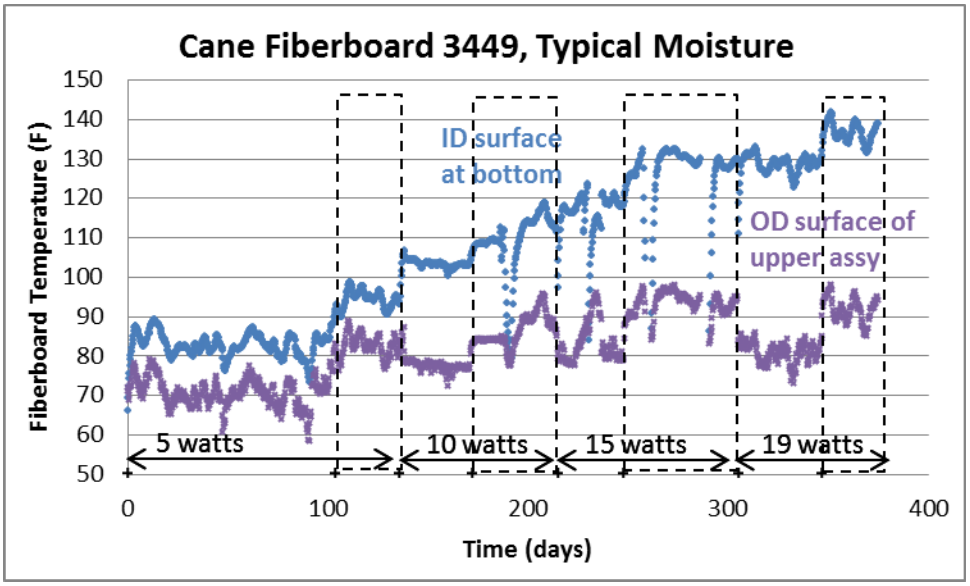


Figure 2. Temperature history for two locations within LE RH1 fiberboard assembly. An insulating blanket was placed around the package during the periods within the dashed boxes.

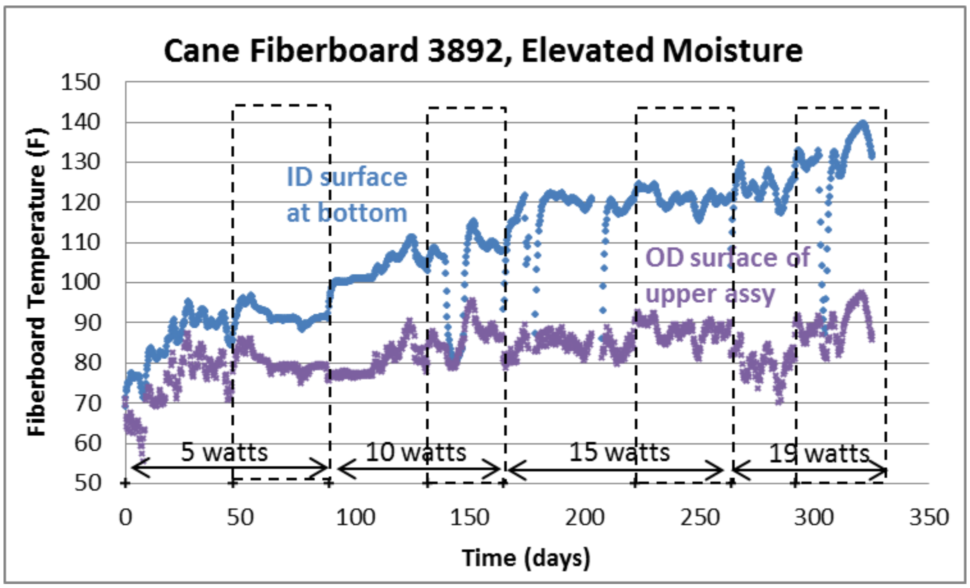
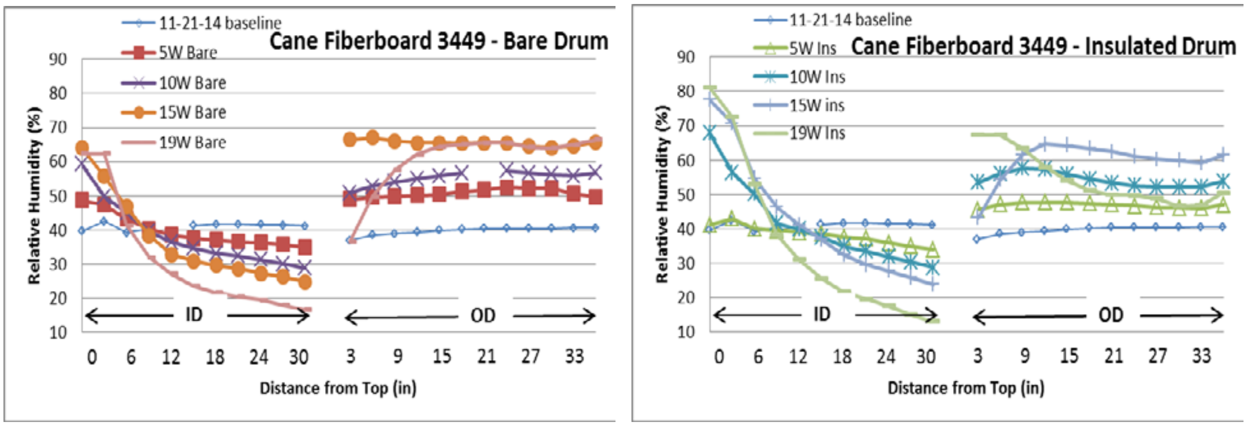
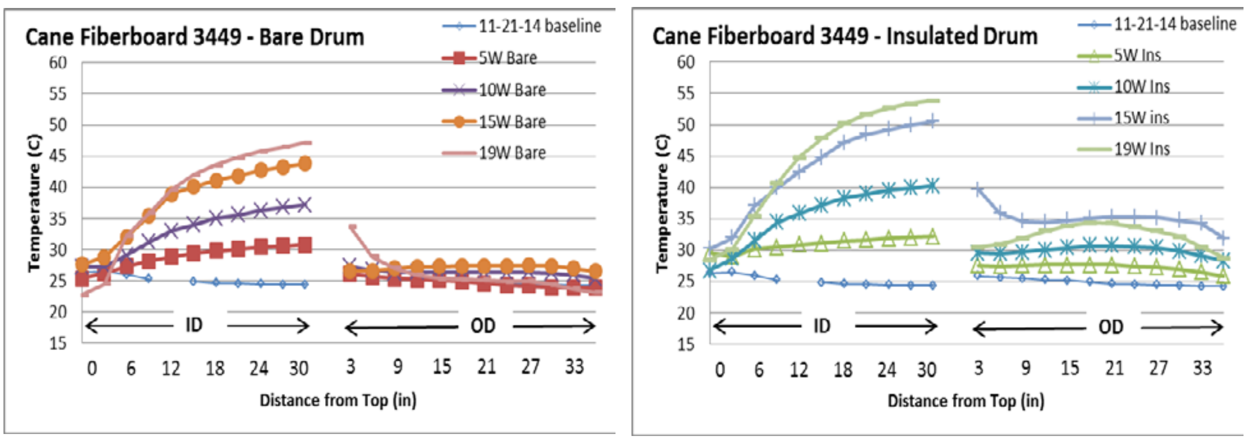


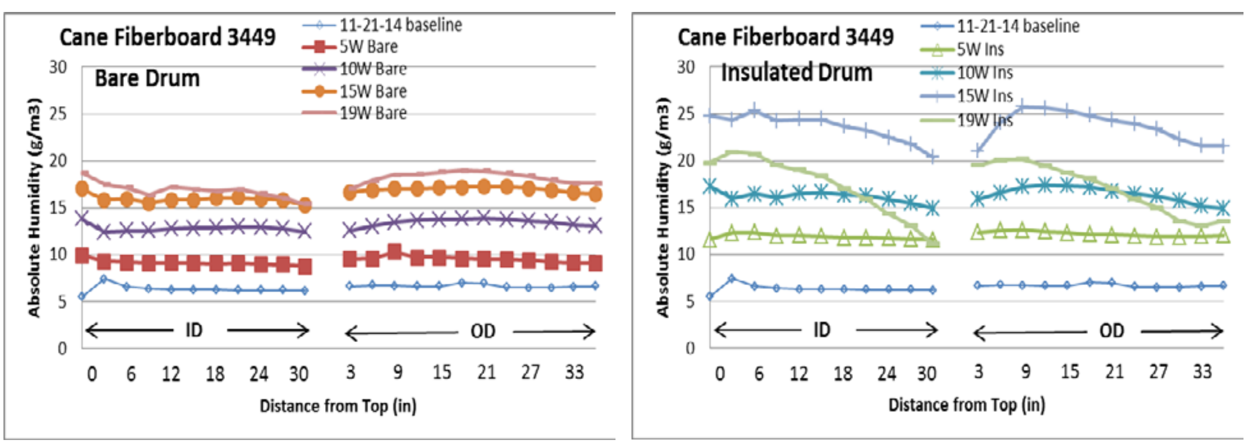
Figure 3. Temperature history for two locations within LE RH2 fiberboard assembly. An insulating blanket was placed around the package during the periods within the dashed boxes.



(a) Relative humidity profiles

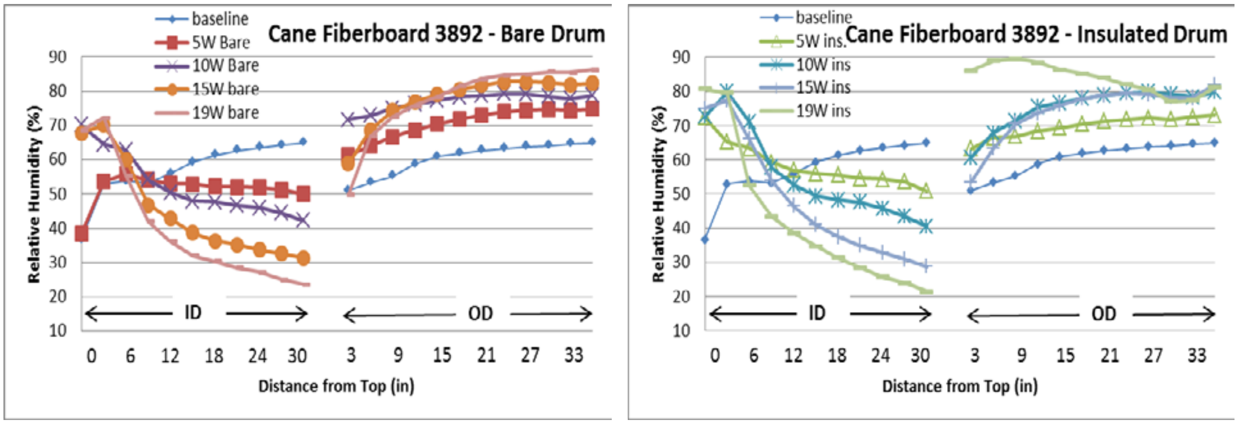


(b) Temperature profiles

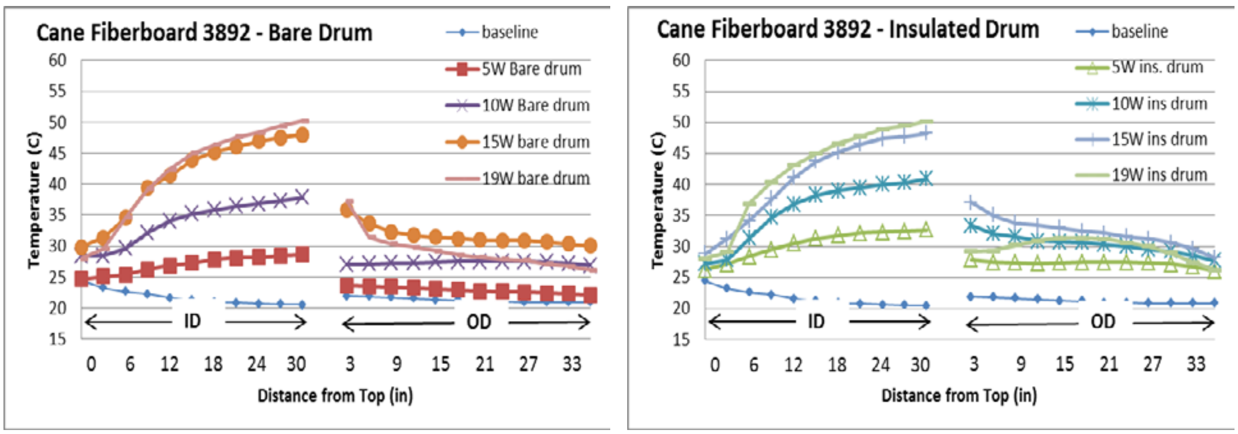


(c) Absolute humidity profiles

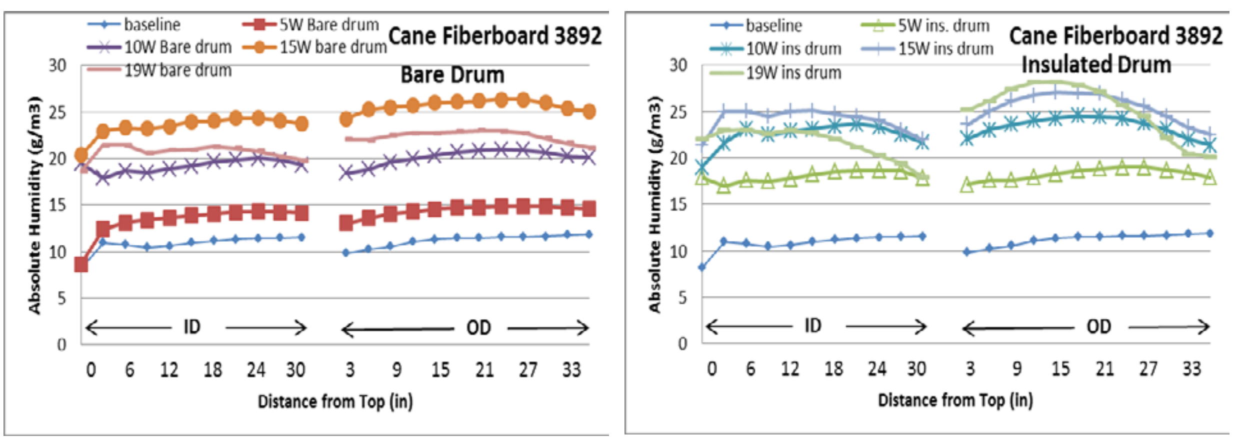
Figure 4. Profiles for package LE RH1 cane fiberboard (nominal moisture content). Each graph shows typical measurements along the ID and OD surfaces of the fiberboard. Profiles are graphed separately for measurements on the bare package and the insulated package for clarity/



(a) Relative humidity profiles



(b) Temperature profiles



(c) Absolute humidity profiles

Figure 5. Typical profiles for package LE RH2 cane 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/

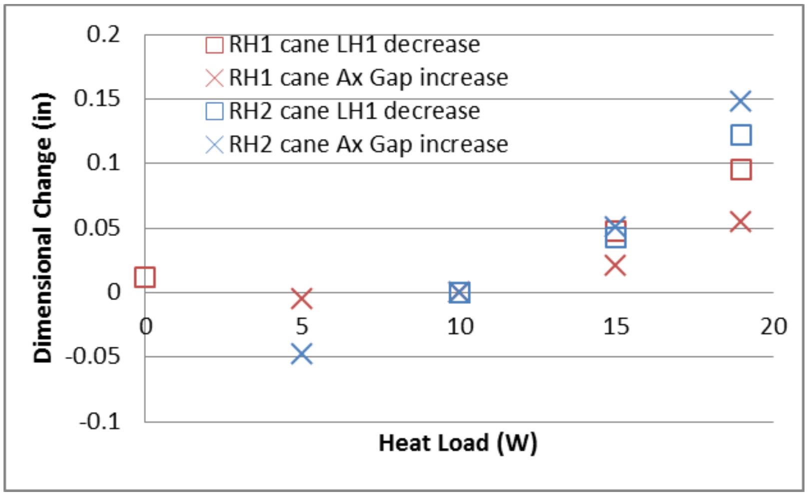
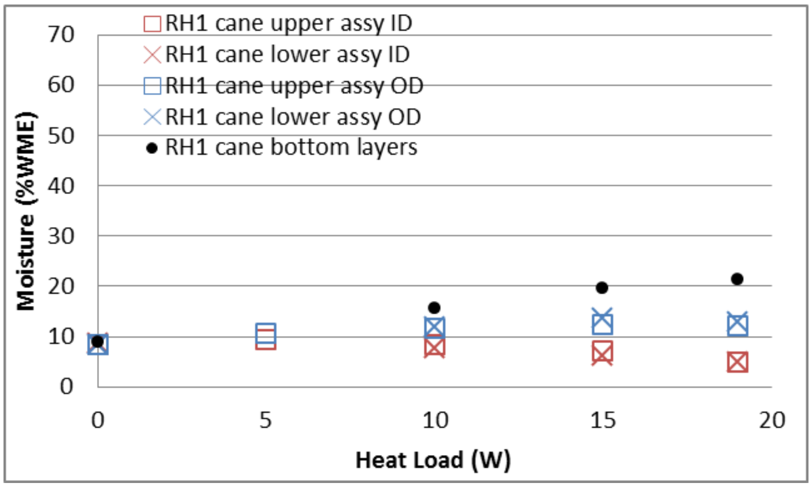
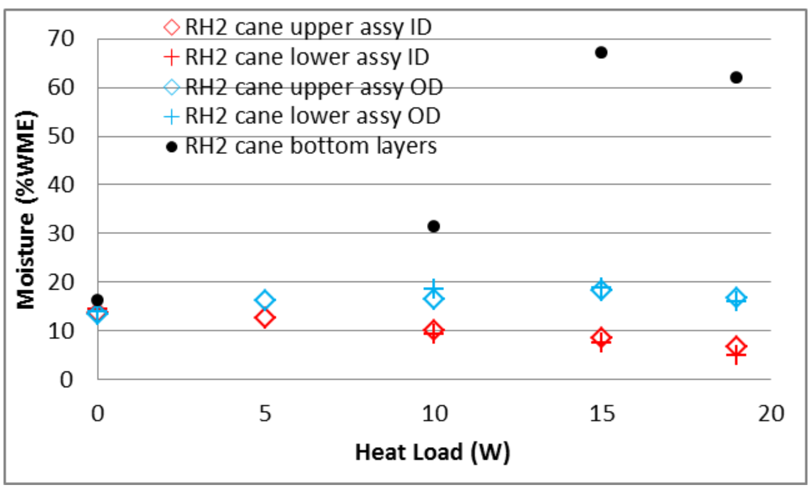


Figure 6. Decrease in lower fiberboard assembly height (LH1) compared to axial gap increase. Data are referenced to the 10W values since some data were not collected at lower heat loads.



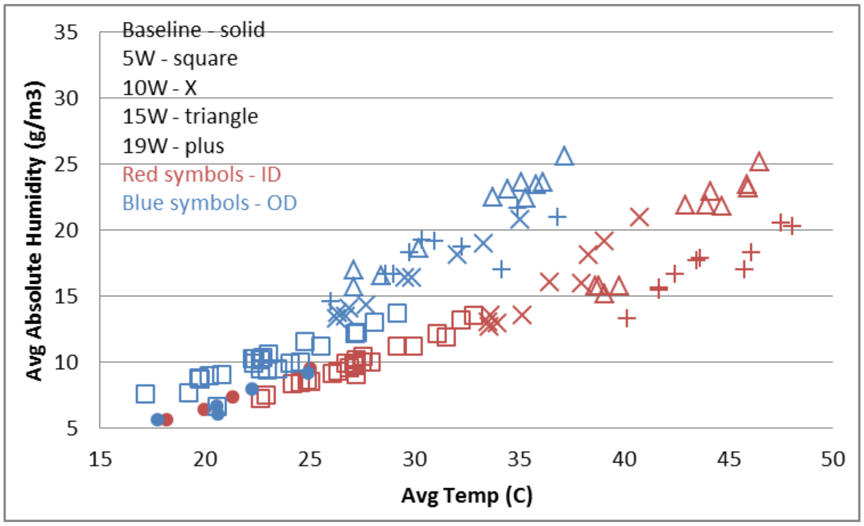
(a) LE RH1



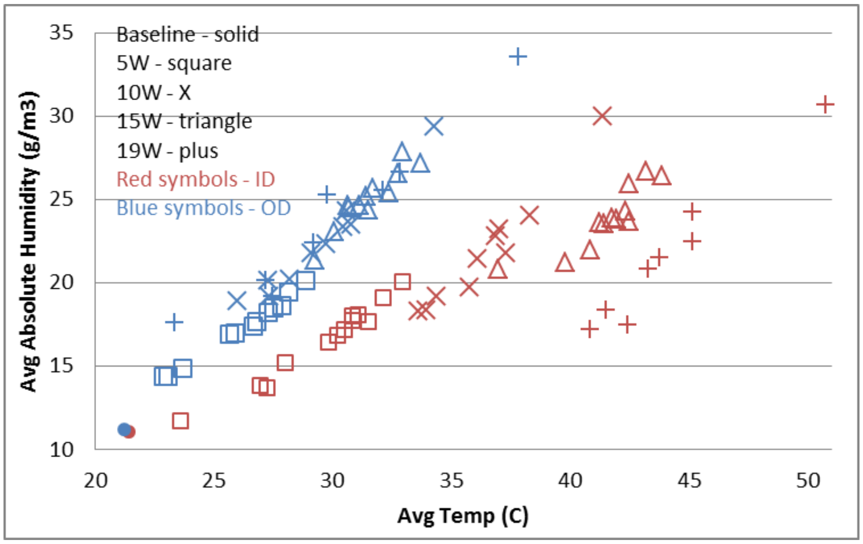
(b) LE RH2

Figure 7. Average moisture values for each fiberboard region

Figure 8. Mold (pink, gray and white) observed on the bottom fiberboard layers of package LE RH2 after 15 watts.



(a) LE RH1



(b) LE RH2

Figure 9. Average absolute humidity vs temperature for all LE RH1 data (a) and all LE RH2 data (b). Results are shown separately in each graph for OD and ID, and for each heat load.

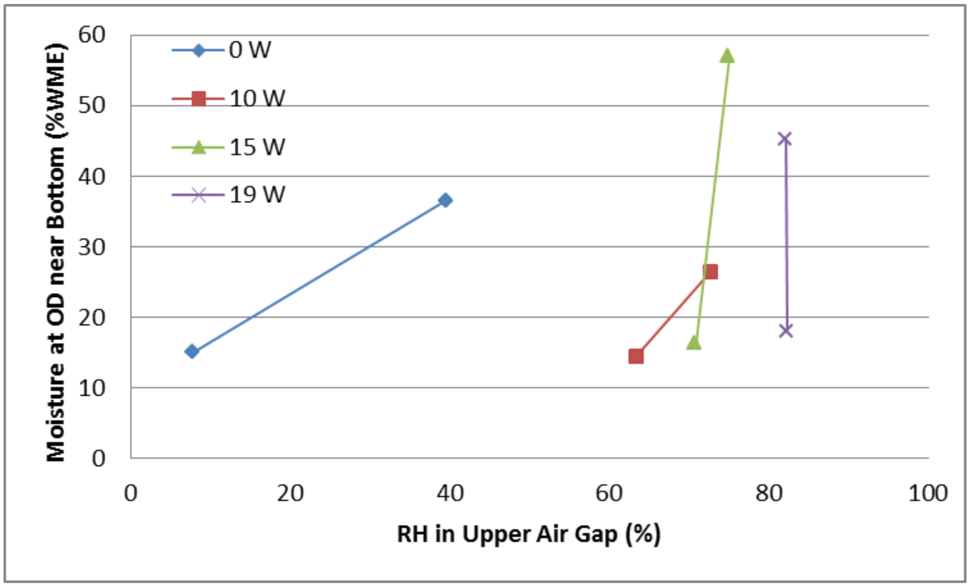


Figure 10. Correlation between relative humidity in the upper air gap and the elevated moisture content in the lower fiberboard layers.

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