NATIONAL TOXICOLOGY PROGRAM STUDIES OF CELLULOSE INSULATION

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Abstract

Cellulose insulation (CI) was selected for study by the NTP based upon production volume, the potential for widespread human exposure, and a lack of toxicity data. Preliminary studies are in progress to determine the feasibility of conducting toxicity studies of CI in animals. Because there are insufficient data available to evaluate the potential hazards of occupational exposure to CI, a workplace exposure assessment is being conducted through an interagency agreement with the National Institute of Occupational Safety and Health (NIOSH). This study was designed to characterize the atmosphere to which workers are exposed during CI installation. The NTP is also conducting studies to characterize the size distribution of particles and fibers, and the chemical composition of bulk CI provided by four manufacturers. These results will help determine the feasibility of conducting inhalation studies of CI in animals.

Introduction

Cellulose insulation (CI) was nominated for study by the National Toxicology Program (NTP) based upon production volume, the potential for widespread human exposure, and the lack of toxicity data on this compound. The Interagency Committee for Chemical Evaluation and Coordination reviewed the nomination, and selected CI for chronic toxicity and carcinogenicity studies. The NTP Study Design Team evaluated all the available information on CI, and concluded that additional information was needed in order to design toxicity studies in animals that are relevant to occupational exposures.

Only limited data were available to evaluate the potential hazards of occupational exposure to CI. For this reason, a workplace exposure assessment is being conducted. The study was designed to characterize the atmosphere to which workers are exposed during the CI installation process. The hazard assessment study is being conducted through an interagency agreement with the National Institute of Occupational Safety and Health (NIOSH) and is the subject of a subsequent paper in this proceedings.

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In addition, insufficient data were available on the chemical composition, and on the amount of variability between CI products from different manufacturers. To fill these data gaps, studies were conducted to characterize the fiber and particle size distribution in CI samples from four major manufacturers. These samples were acquired with the assistance of the Cellulose Insulation Manufacturers Association. Studies were designed to provide information on the amounts of fibrous and non-fibrous particulate and on how much of the CI is potentially respirable. In addition, chemical analyses were performed on these four CI samples to determine the chemical identity and relative concentration of major inorganic additives, the relative concentrations of inorganic trace element impurities, and the relative concentrations of organic materials. These studies were designed to provide information on the presence of potentially toxic chemicals and to allow evaluation of the variability between products from different manufacturers.

After completion of the occupational hazard assessment and chemical characterization studies, the NTP Study Design Team will examine the new data and re-evaluate the necessity and feasibility of conducting toxicity/carcinogenicity studies of CI in animals. The recommendations of the Study Design Team will be reviewed by the NTP Protocol Review Committee, prior to initiating toxicity studies.

Methods

Fractionation of Particulates by Inertia in an Air Stream

A test system was built to fractionate the bulk CI (Figure 1). The test system included a commercial insulation blower, a rough separator, a cyclone separator, a sampling chamber, and a membrane filter bag. The bulk CI was placed in the hopper of the blower where agitator paddles broke the material into small pieces. Agitator vanes at the bottom of the hopper pushed the small pieces to an airlock chamber where about 40-cfm nitrogen was used to carry the test material through a static charge neutralizer into the rough separator.

The rough separator was designed to collect at least 50% of particulates of aerodynamic diameter $\geq$ 12 µm at a flow rate of 40 cfm. Particulates that passed through the rough separator entered the cyclone which was designed to allow $\geq$ 75% of particulates of aerodynamic diameter <10 µm to pass into the sampling chamber. For a fiber of 3 µm diameter to have an aerodynamic diameter of 12 µm, the length must be > 30 µm; for a 1 µm diameter fiber the length would be much greater than 100 µm, assuming the specific gravity of the fiber is 1 (Baron, 1993). Therefore the rough separator and the cyclone did not remove many of the fibers that were considered to have high potential risk to human health (Warheit, 1993).

Individual plastic collection bags at the end of both the rough separator and cyclone allowed weighing of collected material. The majority of particulates of aerodynamic diameter < 10 µm penetrate the cyclone to the sampling chamber where filter (0.2 µm pore size) samples were taken for various analyses. Particulates in the aerosol leaving the sampling chamber were collected downstream in a filter bag. All collection bags were weighed before and after each experiment to determine the fraction in each size range. After weighing the filter bags, the collected materials were transferred to tared Teflon coated filters and weighed again.

During each test run, filter samples were taken from the sampling chamber for the purposes of determining the aerosol concentration and estimating the size distribution. The aerosol concentration was determined by collecting duplicate samples at known flow rates (8.9 L/min) for the duration of each test run. Duplicate samples for size distribution analysis using scanning electron microscopy (SEM) were taken at 0.5 L/min for 4 minutes.

Particulate Counting and Size Distribution Analysis

The size distribution of particulates collected in the cyclone collection bag was determined to evaluate the amount of respirable fibers that were collected in the cyclone separator. Samples were prepared for SEM by dispersing a small amount of this material onto a glass slide. Slides were turned upside down to remove large particles and clusters. Size distribution was analyzed by SEM from one slide for each test sample.

One filter sample taken from the sampling chamber for each test sample was also analyzed using SEM to determine number, concentration and size distribution of the collected particles. Micrographs of 20 randomly selected fields per sample were digitized and saved as image files. An image was randomly selected to determine number and size distribution of total particulate (both fiber and non-fiber particulates) for a sample using commercial software. A minimum of 100 particles per sample was measured.

Micrographs of filters were examined by electron microscopy to determine the number and size distribution of fibers in the sampling chamber. For Samples 1, 2, and 3, all 20 fields (2000x) of each filter from the sample chamber were examined for size distribution of fibrous particulates. Because Sample 4 had a very low particle population, six fields were examined at 500x (equivalent to 96 fields at 2000x). Micrographs of filters were examined using commercial software to determine the number and size distribution of fibers. This software requires manual identification of fibers and manual tracking of length and width. Only particulates of length >5 µm, width $\leq$ 3 µm, and aspect ratio (length to width) ratio $\geq$ 3 µm were counted as fibers. Additional counting rules stated in NIOSH Method 7400 were followed.

Inorganic Chemical Characterization

X-Ray Diffraction. X-ray diffraction (XRD) analysis was used to obtain qualitative and semi-quantitative identification of crystalline phases of the four CI samples.

Elemental Analysis. Triplicate samples of each CI sample were weighed into Teflon microwave digestion vessels and digested with nitric acid. The digests were diluted with water and centrifuged to remove an insoluble residue. The clear supernatants were analyzed for inorganic elements by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The insoluble residue from each sample was dried, weighed, and analyzed by XRD.

Organic Chemical Characterization

CI samples were solvent extracted with methylene chloride. The extracts were analyzed using gas chromatography with flame ionization detection (GC-FID) and gas chromatography / mass spectrometry (GC/MS). The chromatographic profiles and the relative response of total organic material from each of the test sample extracts were compared.

Results

Fractionation of Particulates by Inertia in an Air Stream

For all four samples ~ 99% of the total collected material was deposited in the rough separator collection bag and approximately 1% was deposited in the cyclone collection bag. Less than 0.1% of the collected material was found in the sampling chamber filter bag (as determined gravimetrically) and represents the potentially respirable, small particulate fraction. The amounts of materials transferred from the filter bags to filters were 0.2 mg (Sample 4), 292 mg (Sample 3), 395 mg (Sample 2), 1126 mg (Sample 1). These weights are shown as the relative fraction of the total weight of each processed sample in Table 1. Aerosol concentrations in the sampling chamber were derived from the total mass of particles passing through the sampling chamber and total flow rate.

Particulate Counting and Size Distribution Analysis

The size distributions of total particulates collected from the cyclone separator collection bag and the sampling chamber were determined. The mean equivalent diameter for particulates collected in the cyclone collection bag ranged from 3.5 to 11.4 µm (Table 2). Most of the material collected in the cyclone was in crumbs (several millimeters in diameter) and therefore not included in size determination. The mean equivalent diameter for particulates collected in the sampling chamber ranged from 0.6 to 0.7 µm (Table 3). These measurements demonstrate a difference in particle size distribution between samples collected from the cyclone and the sample chamber. Only one field (2000x) from each sample was examined for particulate size distribution because there were more than 100 particulates on each field. The filter from the sampling

chamber for Sample 4 had a very low particle population compared to other test samples.

Micrographs of filters were examined by electron microscopy to determine the number and size distribution of fibers in the sampling chamber. The total number of fibers identified in the examined fields ranged from 6 (Sample 4) to 172 (Sample 2). Based upon these counts the concentration of fibers in the sampling chamber air was estimated to be 5 (Sample 4), 146 (Sample 1), 538 (Sample 2), and 847 fibers/cc (Sample 3) (Table 4). The total number of fibers generated was estimated based on flow rate and sample times and these data were used to calculate the total number of fibers/gram of insulation. Sample 4 generated considerably fewer fibers than the other samples.

The observed fibers were irregular in shape (Figure 2). Typically fibers were curved or twisted, non-uniform in diameter and had several branches, making it difficult to identify the endpoints of some fibers. Many fibers lay across the boundary of the SEM field, and an additional image at different magnification (500x) was required to determine the length of those fibers. All of these complications made it virtually impossible to automatically measure the size of the cellulose fibers by computerized image analysis, therefore the reported fiber width is the average width visually estimated by the operator. The size ranges of fibers found in the sampling chamber varied considerably; however, fibers from Sample 4 were generally shorter and narrower than the other three CI samples (Table 5).

Inorganic Chemical Characterization

X-Ray Diffraction. XRD analyses indicate that the composition was similar for all 4 samples. Composition was primarily (60%-65% by volume) amorphous, with crystalline phases comprising the remaining fraction (35% to 40% by volume). The composition of the crystalline fraction was also very similar for all samples. The crystalline fraction was primarily native cellulose (75% to 85% by weight), with a smaller amount of cellulose nitrate (15% to 25% by weight).

Elemental Analysis. Elements that were consistently present in all CI samples in quantities greater than about 0.1% by weight include Al, B, Ca, Na, and S (Table 6). Samples 1 and 4 generally contained higher boron and sodium concentrations and Samples 2 and 3 contained higher concentrations of aluminum and calcium. All four samples contained sulfur concentrations greater than 1%. Mean concentrations of Be, Cd, Co, Cr, K, Mo, Ni, Pb, Sb, and Se were below quantifiable limits in all 4 samples.

The composition of the insoluble residue that remained in digested test material was determined for Samples 1 and 3. The relative amounts and composition of the residue were remarkably similar in the two samples analyzed. The insoluble residue comprised approximately 3% to 5% by weight of the original sample and was composed primarily of aluminum silicate hydroxide (kaolinite, ~85%), with smaller quantities (<5% each) of magnesium silicate hydroxide (talc), potassium aluminum silicate hydroxide (muscovite), and titanium oxide (rutile).

Organic Chemical Characterization

In general, the organic material found in the extracts was poorly resolved by the GC/FID under the experimental conditions. The summed area response for organic compounds is similar in all extracts with the exception of sample 4 which contained approximately 6 times the total FID response exhibited by the other 3 samples. Identification of individual compounds in the extracts by GC/MS has not been completed.

Discussion and Summary

Studies were conducted to characterize and compare the particle and fiber size distribution and the chemical composition of CI samples from four major manufacturers. These studies were designed to measure the amounts of fibrous and non-fibrous particulates, to determine how much of the CI is potentially respirable, and to evaluate the variability in CI produced by different manufacturers.

The aerosol generation system effectively separated the CI particles based upon aerodynamic size. The mean equivalent diameter of aerosol particulates ranged from 3.5 to 11.4 µm in the cyclone collection bag and 0.6 to 0.7 µm in the sampling chamber. For all four samples, less than 0.1% of the CI was collected as the small respirable particle fraction in the sampling chamber collection bag. These results indicate that very little of the bulk CI is of a respirable size. In order to provide sufficient respirable material for an inhalation study in animals, bulk CI would have to be mechanically processed; however, this may not provide an aerosol representative of occupational exposure.

The number of respirable fraction fibers generated from the four samples ranged from 9.7×10^3 to 1.4×10^6 fibers/gram test material. Aerosol concentrations in the sampling chamber were less than 60 mg/m^3, which is 20x the TLV of 3 mg/m^3 for respirable nuisance particles. Concentration limits for various highly regulated fibers set by regulatory agencies (OSHA, 1994; ACGIH, 1997). are:

OSHA: 0.1 asbestos fibers/cc 8-hour TWA;
1 asbestos fibers/cc /30 min excursion
ACGIH: 1 glass fiber/cc; 0.2 crocidolite, 0.5 amosite,
2 chrysotile and other asbestos/cc.

Under our experimental conditions, all four samples generated fiber concentrations in the sampling chamber that exceeded the stated limits for the cited glass or mineral fibers. Therefore in order to evaluate the potential toxicity

of fibers generated from cellulose insulation, further characterization of properties (i.e. solubility and durability in lung fluid) of these fibers may be warranted.

The chemical composition of the four CI samples was similar with only minor differences in inorganic additives. Boron, a potential reproductive toxin, was present in all samples at relatively high concentrations. Additional studies may be warranted to evaluate pulmonary absorption and distribution of boron resulting from inhalation exposure to CI. Similar trace amounts of organic compounds were present in all four samples; however, at this time these components have not been identified or quantified.

In general, CI from the different manufacturers was similar in composition, although Sample 4 had considerably fewer fibrous and nonfibrous particles than the other samples. It would be of interest to determine whether the low number of respirable particles in sample 4 was achieved by a difference in the manufacturing method.

Data from these chemical characterization studies will be evaluated and compared with analogous data obtained from the NIOSH occupational exposure assessment. This information on CI is needed in order to design inhalation studies in animals that are relevant to occupational exposures.

References

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Table 1. Distribution of Cellulose Insulation in the Test System

Sample No.	Mass (kg) Processed	Amount Deposited in Collectors: Rough Separator	Cyclone	Chamber Filter Bag[b]	Chamber Aerosol[a] (mg/m^3)
1	13.6	98.7 %	1.3 %	8.3×10^{-5}	48.3 ± 9.9
2	14.1	99.0 %	1.0 %	2.8×10^{-5}	22.9 ± 0.2
3	14.2	99.1 %	0.9 %	2.1×10^{-5}	15.4 ± 0.2
4	8.6	99.5 %	0.5 %	2.3×10^{-8}	$0.3 + 0.08$

[a] Estimated from air flow rate through chamber and amount of particulate collected on filters.
[b] mg

Table 2. Number and Size Distribution of Total Cellulose Insulation Particulates in the Cyclone Collector

Sample Number	Particle Count[a]	Equivalent Diameter[b] (µm) Mean	SD	Min	Max
1	136	6.8	3.9	0.7	23.0
2	434	3.5	4.0	0.4	27.5
3	286	7.5	6.5	0.9	52.6
4	196	11.4	9.9	0.7	64.4

[a] Total Particulate counts in one field at 2000x magnification.
[b] Equivalent Diameter: the diameter of a circle having the same surface area as the target item.

Table 3. Number and Size Distribution of Total Cellulose Insulation Particulates in the Sampling Chamber

Sample Number	Particle Count[a]	Equivalent Diameter[b] (µm) Mean	SD	Min	Max
1	2658	0.6	0.49	0.1	6.9
2	6930	0.7	0.57	0.1	10.8
3	4933	0.6	0.59	0.1	5.5
4	18	0.6	0.62	0.2	2.8

[a] Total Particulate counts in one field at 2000x magnification.
[b] Equivalent Diameter: the diameter of a circle having the same surface area as the target item.

Table 4. Number of Fibers in Samples from the Sampling Chamber

Sample Number	# Fibers[a]	Fibers /Filter	Fibers /cc Air	Total Fibers[b]	Fibers/g Insulation
1	30	2.4×10^5	146	5.6×10^9	4.1×10^5
2	172	1.4×10^6	847	1.9×10^{10}	1.4×10^6
3	109	9.0×10^5	538	1.2×10^{10}	8.6×10^5
4	6[c]	1.1×10^4	5	8.4×10^7	9.7×10^3

[a] Total fiber counts in 20 fields @2000x.
[b] Total number of fibers generated from CI; estimated based on flow rate and sample time.
[c] Total fiber counts in 6 fields @ 500x (equivalent to 96 fields @ 2000x).

Table 5. Size Ranges of Fibers Found in the Sampling Chamber

Sample Number	Length (µm) Max	Length (µm) Min	Width (µm) Max	Width (µm) Min	Aspect Ratio[a] Max	Aspect Ratio[a] Min
1	33.3	5.2	2.4	0.2	35.2	3.9
2	53.5	5.0	2.9	0.2	56.3	3.3
3	29.1	5.0	2.9	0.2	91.1	4.3
4	18.5	7.7	1.2	0.6	18.2	11.7

[a] Ratio of length : width

Table 6. Major Chemical Elements in Cellulose Insulation

Element	Sample 1	Sample 2	Sample 3	Sample 4
Al	0.40	0.49	0.80	0.52
B	1.37	2.00	0.53	0.52
Ca	0.11	0.23	0.24	0.33
Fe	0.017	0.033	0.031	0.027
K	<0.07	<0.1	<0.1	<0.1
Mg	0.024	0.057	0.022	0.027
Na	1.47	2.12	0.12	0.082
P	<0.01	<0.01	0.04	0.17
Pb	<0.005	<0.005	<0.005	<0.005
S	3.60	1.43	2.81	2.47
Si	0.030	0.030	0.032	0.026
Ti	0.007	0.008	0.009	0.006

Less than values (<) are below the quantifiable limit for the ICP-AES method.