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Inhalation exposure during spray application and subsequent sanding of a wood sealant containing zinc oxide nanoparticles

Michael R. Cooper[a], Gavin H. West[a], Leonard G. Burrelli[b], Daniel Dresser[b], Kelsey N. Griffin[b], Alan M. Segrave[c], Jon Perrenoud[c], and Bruce E. Lippy[a]

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ABSTRACT

Nano-enabled construction products have entered into commerce. There are concerns about the safety of manufactured nanomaterials, and exposure assessments are needed for a more complete understanding of risk. This study assessed potential inhalation exposure to ZnO nanoparticles during spray application and power sanding of a commercially available wood sealant and evaluated the effectiveness of local exhaust ventilation in reducing exposure. A tradesperson performed the spraying and sanding inside an environmentally-controlled chamber. Dust control methods during sanding were compared. Filter-based sampling, electron microscopy, and real-time particle counters provided measures of exposure. Airborne nanoparticles above background levels were detected by particle counters for all exposure scenarios. Nanoparticle number concentrations and particle size distributions were similar for sanding of treated versus untreated wood. Very few unbound nanoparticles were detected in aerosol samples via electron microscopy, rather nano-sized ZnO was contained within, or on the surface of larger airborne particles. Whether the presence of nanoscale ZnO in these aerosols affects toxicity merits further investigation. Mass-based exposure measurements were below the NIOSH Recommended Exposure Limit for Zn, although there are no established exposure limits for nanoscale ZnO. Local exhaust ventilation was effective, reducing airborne nanoparticle number concentrations by up to 92% and reducing personal exposure to total dust by at least 80% in terms of mass. Given the discrepancies between the particle count data and electron microscopy observations, the chemical identity of the airborne nanoparticles detected by the particle counters remains uncertain. Prior studies attributed the main source of nanoparticle emissions during sanding to copper nanoparticles generated from electric sander motors. Potentially contrary results are presented suggesting the sander motor may not have been the primary source of nanoparticle emissions in this study. Further research is needed to understand potential risks faced by construction workers exposed to mixed aerosols containing manufactured nanomaterials. Until these risks are better understood, this study demonstrates that engineering controls can reduce exposure to manufactured nanomaterials; doing so may be prudent for protecting worker health.

KEYWORDS
Coating; construction; local exhaust ventilation; nano-enabled; nanomaterial; nano-objects and their agglomerates and aggregates

Introduction

This study is part of The Center for Construction Research and Training's (CPWR) efforts to identify nano-enabled construction products, assess potential for worker exposure, and evaluate the efficacy of exposure controls. Manufacturers of over 500 construction materials in CPWR's inventory (http://nano.elcosh.org) state or suggest that their products contain manufactured nano-objects and their agglomerates and aggregates (NOAA)[1] and, as a result, have improved characteristics or performance. Claims for nano-enabled products are based on addition of NOAA or alterations to products' nanostructures. Properties, such as photocatalysis and strength, of materials in the nanoscale size range of

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approximately 1–100 nm can differ significantly from their larger scale counterparts.[2]

Smaller particle size allows nanoparticles to reach and impact parts of the body that larger particles cannot. For example, nanoparticles can pass through the blood-brain barrier.[3] Compared to larger particles of similar morphology, nanoparticles can be more bioreactive per unit of mass due to increased surface area.[4,5]

The U.S. demand for wood protection coatings and preservatives was estimated to be 4.5 billion dollars annually.[6] Coatings account for a substantial portion of nano-enabled construction products in Europe and over half of CPWR's inventory of nano-enabled construction products.[7] The application of wood protection has been described as "one of the world's largest end uses of nanoparticles."[8] Along with pre-market treatment of lumber, there is a wide range of coatings intended for use on interior surfaces, such as flooring and cabinets, and on exterior surfaces, such as decks or siding.

The current study sought to evaluate inhalation exposure during use of a commercially available wood sealant, which is featured in CPWR's online database of nano-enabled construction materials.[9] Marketing material provided by the sealant manufacturer indicated that it contained an unspecified concentration of ZnO nanoparticles.[10] The product was selected based on the potential for widespread use of similar products and exposure to NOAA during spray application and subsequent sanding.

Treating wood with ZnO nanoparticles protects it from water absorption, inhibits decay fungi, acts as a termite toxicant, and provides photostability of wood and coatings against ultraviolet degradation.[11–13] Use of nanoparticles aids the even distribution of ZnO and enhances antimicrobial effects.[11,14]

While consumers are likely to use these products infrequently, construction workers may apply wood protective coatings for extended periods on a regular basis. Unfortunately, information on the toxicity of NOAA and the likelihood of exposure is often limited, and manufacturers of construction materials often provide minimal information on NOAA used in their products.[7,15,16] Compared to other routes of exposure, inhalation of NOAA is generally considered to pose the greater risk to construction workers, though other routes of exposure should not be discounted.

Inhalation of welding fumes containing ultrafine ZnO can result in symptoms including metal fume fever, chills, muscle ache, nausea, fever, dry throat, cough, lassitude, metallic taste, headache, blurred vision, low back pain, vomiting, malaise, chest tightness, dyspnea, rales, and decreased pulmonary function.[17] Inhalation of ZnO nanoparticles has been shown to result in inflammation of lung and cardiac cells in animals.[18–21] Kuschner et al.[22] observed markers of pulmonary inflammation in humans following 10–30 min of exposure to 33 mg/m^3 of 170 nm ZnO aggregate composed of 8–40 nm particles. Beckett et al.[23] exposed humans to 0.5 mg/m^3 ultrafine ZnO for 2 hr per day for 3 days with no measurable effects. Fine et al.[24] exposed 13 subjects to ZnO fume through inhalation for 2 hr at 2.5 mg/m^3 and 5 mg/m^3. The subjects exposed to 5 mg/m^3 are described as experiencing fever and symptoms along with elevated plasma interleuken-6 levels. The subjects exposed to 2.5 mg/m^3 experienced fever and elevated interleuken-6 levels but did not report symptoms. Studies of chronic inhalation of ZnO are limited and contain incomplete information on exposures to mixtures containing ZnO.[25]

Study of exposure to NOAA during spraying has been limited to consumer products, such as sunscreens and disinfectants applied by pump spray dispensers or propellant gas dispensers,[26–29] but there are also concerns about products used in occupational settings.[30] Berger-Preiß et al.[31] examined spray application of wood preservatives without considering NOAA and found that inhalation exposures were higher when the solvent was organic instead of water, when application rates were higher, and when the aerosol generated by the sprayer was smaller.

Multiple studies have evaluated release of added NOAA from coatings during sanding. Compared to sanding conventional coatings or uncoated substrate, most have found no free nanoparticles, no change in particle size distribution, and no clear difference in count when sanding coatings containing added NOAA.[32–35] Few researchers have worked with products containing ZnO. Fransman et al.[36] compared release of NOAA during sanding wood coated with a water-based protective coating containing 0.8% ZnO to sanding untreated wood. An orbital sander with a 2 kg weight to provide consistent pressure was operated in a small chamber with minimal ventilation. They found no difference in NOAA number concentration when sanding ZnO-coated vs. untreated wood. Scanning electron microscopy (SEM) indicated ZnO nanoparticles were embedded in larger wood particles.

Objectives of this study were to assess inhalation exposure to ZnO NOAA during spray application and power sanding of a commercially available wood sealant with added ZnO NOAA, compare exposure profiles of the different work activities performed, and evaluate the effectiveness of local exhaust ventilation (LEV) in reducing exposure to NOAA.

Methods

Sampling was conducted inside an environmentally-controlled chamber to minimize confounding measurements of process-generated ultrafine particles, such as those found in vehicle exhaust. One tradesperson with decades of construction experience performed all tasks after receiving safety training and technical guidance from an apprenticeship instructor from the International Union of Painters and Allied Trades. A comprehensive three-pronged approach was used for exposure assessment. Filter-based sampling provided mass-based measures of exposure. Electron microscopy (EM), either SEM or transmission electron microscopy (TEM), allowed for morphological and elemental characterization of NOAA, and real-time particle counters provided particle size distributions and particle number concentrations. CPWR's Institutional Review Board (FWA 00000608, IRB 00001202) approved the sampling activities described below.

Bulk characterization

A 1-gal can of the wood sealant was stirred with a wooden paint stirring stick for approximately 10 min, and a 50 mL aliquot was submitted to the microscopy laboratory (Bureau Veritas NA, Inc., Kennesaw, GA). Particle size and morphology were determined via on-screen observation and micrograph analysis. A Tescan scanning electron microscope equipped with a Gresham light element detector and an IXRF digital imaging system capable of providing energy dispersive spectrometry (EDS) was used for elemental identification of particles.

Sampling protocol

Sampling was conducted inside an environmentally-controlled chamber in Columbia, Maryland (see Figure 1). Smoking was not allowed in or around the chamber. A HEPA-filtered ventilation unit was used to evacuate air from the 2,900 ft^3 chamber at a rate of approximately 2 air changes per hour and maintain negative pressure relative to outside. Make-up air was directed into the chamber through a HEPA filter, nominally 10 × 21.5 in., affixed to the wall of the chamber to reduce the particulate entering the chamber. Personal air samples were collected to assess exposure to the tradesperson spraying and sanding, and area samples were collected to assess bystander exposure. Temperature and relative humidity were measured and maintained throughout the study to assure consistent conditions during sampling.

Real-time measurements of particle diameter and count were collected once per minute with a Scanning Mobility Particle Sizer, a TSI NanoScan SMPS Nanoparticle Sizer Model 3910 (TSI Incorporated, Shoreview, MN) at the SEM sampling station during and between all spraying and sanding. The Model 3910 cyclonically removes large aerosol particles with a cutoff point of approximately 550 nm. The unit creates a charge on the remaining particles that are then sized with a Radial Differential Mobility Analyzer and counted with a Condensation Particle Counter. Together, the NanoScan SMPS and the Optical Particle Sizer (OPS) (TSI Model 3330, TSI, Shoreview, MN) measure size in three orders of magnitude from 10 nm to 10 µm. Data collected prior to spraying and sanding were used to establish background particle diameter and count.

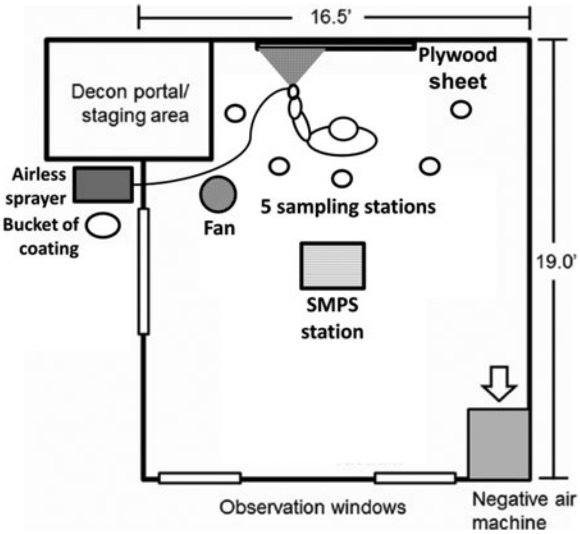

Figure 1. Diagram of the sampling chamber during spray application.

Entry into and exit from the study chamber were controlled through a segregated airlock attached to the testing chamber. All tools, sampling media, and personal protective equipment, i.e., respirator and Tyvek suit, were kept in the airlock between trials. The door from the chamber exterior to the airlock was sealed using duct tape during and between each trial.

Following each trial, the chamber air was purged at an accelerated rate using the HEPA-filtered negative air machine. Airborne particle counts were measured while purging to observe the decrease in airborne particle concentrations, returning the chamber to a baseline condition prior to the next trial.

Sampling during spraying

A tradesperson used a Graco Magnum X5 airless paint/coating sprayer (Graco, Minneapolis, MN), sprayed the product on the finished, sanded surfaces of vertically mounted 11/32" BC Sanded 4 × 8 foot Plytanium

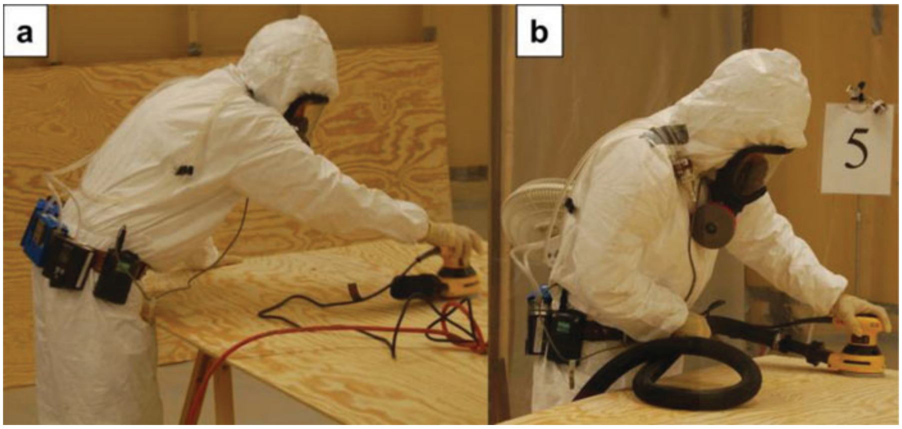

Figure 2. Sanding with the dust collection bag (a) and with LEV (b).

plywood sheathing. During each of six trials, the product was applied to four sheets of plywood. Spraying, at a rate of approximately 0.1 gal per minute, required 3–4 min of each 11-min trial. After drying, the plywood was sprayed in this manner again.

Four sampling stations, positioned in an arc around the work station, had identical sampling media: 37-mm diameter, closed-face cassettes containing 5-micron porosity, pre-weighed, polyvinyl chloride (PVC) filters (Bureau Veritas, Novi, MI) placed at breathing zone height on sampling stands approximately 4 ft from the work and attached by Tygon tubing to high volume samplers (Reliance Electric, Gallipolis, OH) calibrated to draw between 5.7 and 6.2 liters per minute (LPM). These cassettes were analyzed for total dust by National Institute for Occupational Safety and Health (NIOSH) Method 0500, respirable dust by NIOSH 0600 and for zinc by NIOSH Method 7300/7303. One additional station was established in the center of the sampling station arc, with a 37-mm diameter, open-face cassette containing a 5-micron porosity, pre-weighed, PVC filter (Bureau Veritas, Novi, MI) placed at breathing zone height on a sampling stand approximately 4 ft from the work and attached by Tygon tubing to a high volume sampler (Reliance Electric, Gallipolis, OH) calibrated to draw between 5.7 and 6.2 LPM for SEM/EDS analysis. SEM was completed to identify, photograph, and determine the elemental composition of NOAA encountered on the air sampling medium.

Sampling during sanding

A tradesperson used a DeWalt 5-inch random orbital sander (DeWalt, Baltimore, MD) to repeatedly sand the dry product on the horizontally-mounted Plytanium boards. Three dust capture methods were investigated: sanding with the DeWalt dust collection bag (DeWalt, Baltimore, MD) provided with the sander (Figure 2a), sanding with LEV supplied by a Hilti VC40-U (Hilti Corporation, Schaan, Liechtenstein) connected to the sander (FIGURE 2b), and sanding without any dust capture attachment. Three sanding trials were performed for each of these conditions for both coated plywood and uncoated plywood, which served as controls. Sanding was completed over the entire surface of the plywood in a period of between 8 and 10 min. A new 100-grit sanding pad was used for each of the sanding trials. The Hilti VC40-U was specified to provide an air flow rate of 129 cubic feet per minute.

The tradesperson who performed all of the work wore a MSA Ultra Elite full-face mask connected to an OptimAir powered air-purifying respirator (Mine Safety Appliances Company, Inc, Cranberry Township, Pennsylvania), a Tyvek suit with hood, and nitrile or latex gloves during the spraying operation. For the sanding operation, he wore hearing protection in addition to the previously mentioned personal protective equipment.

During sanding, identical sampling media were used. The work station was placed horizontally in the center of the room and the area sampling stations were placed in a circular fashion around the workstation, with each sampling station approximately 4 ft from the work, while the EM sampling station was placed in the northeast corner of the chamber.

Four personal sampling pumps worn by the test subject at waist level were connected by Tygon tubing to a sampling device affixed in his breathing zone. Two GilAir 5 personal air samplers (Sensidyne Inc., USA) pulled air at 4.2 LPM through an aluminum GK2.69 (BGI by Mesa Labs, Inc., Butler, NJ) respirable dust cyclone holding a 37 mm, 5-micron porosity, pre-weighed PVC filter. The other pumps worn by the test subject were two Escort Elf personal air samplers (Zefon International Inc., Ocala, FL) that pulled air at 3.0 LPM with 37-mm diameter, open-face cassettes containing 5-micron porosity, pre-weighed, PVC filters. One sample from each of the two types of personal air samplers, i.e., total particulate (open-faced) and respirable particulate (cyclone) underwent SEM/EDS or TEM/EDS analysis to identify, photograph, and determine the elemental composition of NOAA encountered on the air sampling medium. The remaining open-faced sample was analyzed for total dust by NIOSH Method 0500 and for zinc by NIOSH Method 7300/7303. Another sample was collected using the cyclone and analyzed by NIOSH 0600 for respirable dust and NIOSH Method 7300/7303 for zinc. All pumps were calibrated before and after sampling using an electronic dry piston primary flow meter (DryCal DC-Lite; Bios International Corp., Butler, NJ).

Analytical Methods

Air samples for determination of total dust, respirable dust and airborne zinc were sent under a chain of custody to Bureau Veritas, Novi, MI. Air samples collected for EM analysis were submitted to Bureau Veritas, Kennesaw, GA. Both labs are accredited by the American Industrial Hygiene Association and the National Institute of Standards and Technology under the National Laboratory Voluntary Laboratory Accreditation Program.

An indirect preparation for SEM analyses was used that involved cutting a representative portion from each filter, placing it into a crucible of isopropyl alcohol, sonicating the samples and then filtering the aliquot onto a polycarbonate filter. A portion of the filter was placed onto a carbon taped stub and carbon coated to provide electrical conductivity for analysis. Sizing and elemental identification of particles and structures on the sample filters were performed using a Tescan scanning electron microscope equipped with a Gresham light element detector and an IXRF digital imaging system capable of providing EDS.

A direct preparation for TEM analyses was used that involved cutting a filter into quarters, placing a portion of each filter on a glass slide, adding a dimethyl formamide/acetic acid clearing solution and then carbon coating in a vacuum evaporator. The filter was placed on a copper grid in a solution washer containing acetone. The sizing and elemental identification of particles and structures on the sample filters were performed with a Phillips TEM equipped with an IXRF digital imaging system capable of providing EDS.

Statistical methods

Univariate methods were used to generate summary statistics for the filter-based, gravimetric industrial hygiene measurements. Shapiro-Wilk tests and quantile-quantile plots were used to assess normality of the data. Spray application exposure data were normally distributed and are thus presented as mean concentrations with standard deviations and 95% confidence intervals. Sanding exposure data are presented as exposure ranges with median concentrations due to the high proportion of samples below detectable limits.

Real-time particle count data from the OPS and SMPS were merged and fitted using TSI Inc. software (NanoScan Manager, Aerosol Instrument Manager, and Multi Instrument Manager). Normalized particle number concentrations ($dN/dlogDp$) were used to account for differences in resolution between the two instruments, where $dN =$ particle concentration and $dlogDp =$ the difference in the log of the channel width. A representative particle size distribution is presented for each exposure scenario, each representing a single experimental trial. Concentrations were adjusted for background by subtracting background measurements immediately preceding each trial. The first three measurements from each experimental run were discarded; these measurements closely approximated background levels prior to the point in time when dust or aerosol was actually being generated or measured.

Normalized particle number concentrations in the nanoscale size range were summed for all experimental trials and exported for analysis. Analysis of Variance (ANOVA) was used to determine if airborne nanoparticle number concentrations differed for coated vs. uncoated wood and by method of dust capture used. Student Newman-Keuls multiple range test was used for post hoc comparison to examine where significant differences occurred (e.g., sanding treated wood with LEV versus no dust capture). Levene's Test for Homogeneity of Variance, quantile-quantile plots, and Shapiro-Wilk tests were used to verify that ANOVA assumptions were satisfied. All nanoparticle number concentrations while sanding were normally distributed, with the exception of sanding with LEV. Given the large effect size when sanding with LEV and the robustness of the F-test despite slight departures from normality, ANOVA was deemed an appropriate test. Hypothesis testing and univariate methods were performed using SAS 9.4 (SAS Institute Inc., Carey, NC).

Results

Electron microscopy observations

SEM confirmed the presence of ZnO NOAA in bulk samples of the product (Figure 3). ZnO nanoparticles remained bonded after sonication, suggesting the presence of more strongly bonded aggregates as opposed to agglomerates. SEM analysis indicated that 45% of particles in the sealant had diameters below 300 nm and that 8.3% of suspended particles were nanoscale. In aerosol samples collected during spray application, ZnO NOAA were observed on the surface or within larger globules of

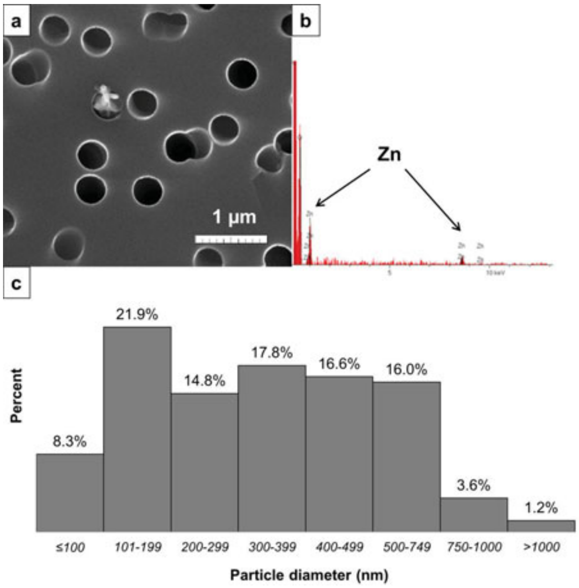

Figure 3. Bulk characterization of the sealant: SEM image of nanoparticles in a filter pore at x80,000 magnification (a), spectral analysis (b), and particle size distribution via SEM (n = 169) (c).

an organic component of the sealant, which ranged in size from approximately 0.5–12 μm (Figures 4a, 4b).

A range of particles with different morphologies, sizes, and chemical compositions were detected by EM in aerosol samples collected while sanding without dust capture. Agglomerated nanoparticles were observed in multiple samples, while free nanoparticles of unknown composition were only observed in one personal sample (Figure 5). Aside from aluminum oxide, agglomerated nanoparticles of silicon dioxide were observed (image not shown). Numerous metallic particles below 1 μm were observed by TEM (Figure 6). Finally, SEM with secondary electron back scatter revealed ZnO NOAA on the surface of, or embedded within larger micron-sized particles, presumed to be fragments of the plywood (Figure 7).

Particle number concentration and size distribution

Particle size distribution of the aerosol generated during spray application of the sealant was bimodal with a peak of 6.542×10^4 particles at 187 nm and a second peak of 9,340 particles at 2.16 μm (Figure 8). The left tail of the curve illustrates the proportion of particles below 100 nm detected by SMPS.

In contrast to spray application, the highest differential particle number concentrations were observed in the size range below 100 nm during sanding. With use of the dust collection bag, particle size distributions were strikingly similar for sanding of treated versus untreated wood (Figures 9a, 9b). Peaks were observed at 14 nm, 28 nm, and 154 nm. Sanding treated wood without any dust capture mechanism resulted in a similar particle size distribution but lower particle number concentrations (Figure 9c). Sanding treated wood with LEV produced a markedly different bimodal particle size distribution with peaks at 34 nm and 332 nm, and much lower particle number concentrations were observed (Figure 9d).

Real-time particle counts showed that LEV was effective in reducing levels of airborne nanoparticles during sanding. Nanoparticle concentrations were significantly reduced when using LEV ($p \leq 0.05$) compared to any other sanding protocol (Figure 10). For example, the mean concentration while sanding treated wood with the dust collection bag was 241,779 nanoparticles/cm^3 compared to 20,592 nanoparticles/cm^3 when using LEV, a reduction of 91.5%. Sanding with the dust collection bag provided with the sander produced the highest nanoparticle concentrations, exceeding nanoparticle concentrations when no dust capture mechanism was being used ($p \leq 0.05$). There was no statistically significant difference, however, between sanding of treated and untreated wood using the manufacturer-provided dust collection bag.

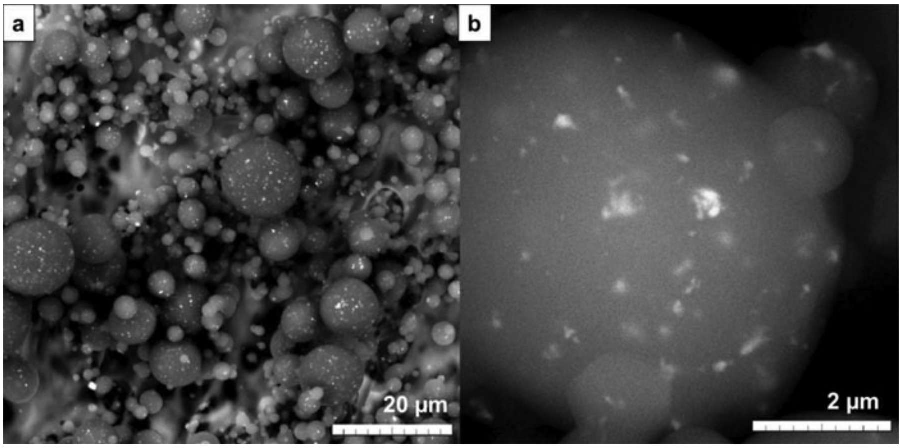

Figure 4. SEM images of aerosol captured during spray application showing ZnO nanoparticles or agglomerates positioned on the surface or within larger globules the sealant at x5,000 magnification (a) and x60,000 magnification (b).

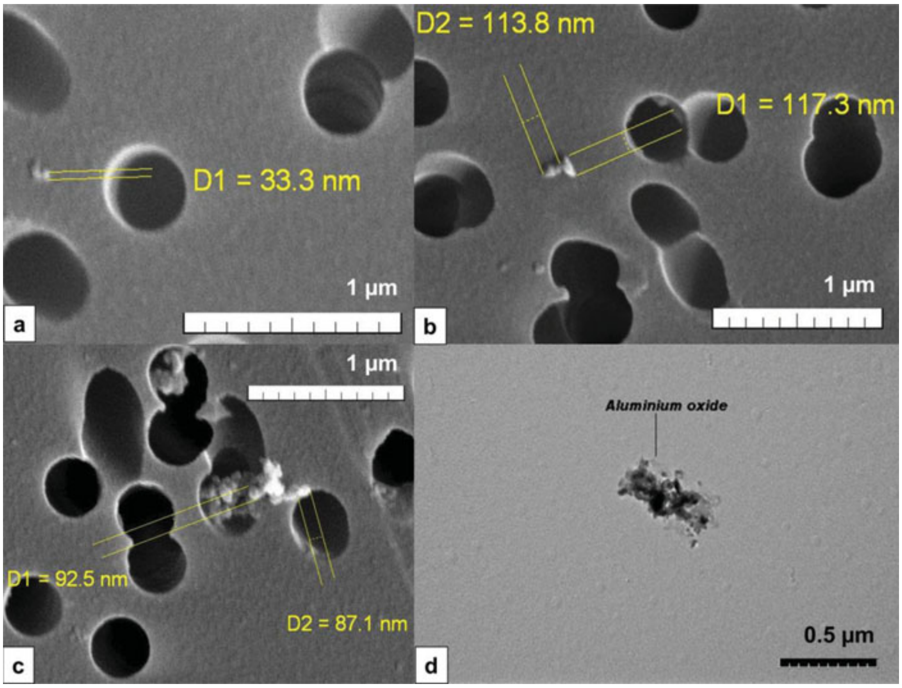

Figure 5. Nanostructured components of the airborne sanding dust: individual nanoparticles of undetermined chemical composition (a, b), agglomerated nanoparticles of undetermined chemical composition (c), and agglomerated aluminum oxide (d).

Mass concentrations of filter-based samples

Spray application of the sealant generated total aerosol concentrations of 96 mg/m^3 for area samples and 84 mg/m^3 for personal samples (Table 1). Approximately 2.4% of the total aerosol mass concentration was zinc. The 95% confidence interval for personal exposure to respirable zinc was 0.39–0.59 mg/m^3. All area and personal exposure measurements during spray application were below the NIOSH Recommended Exposure Limit (REL) for total zinc (5 mg/m^3 time-weighted average (TWA)); the maximum observed concentration was 3 mg/m^3.

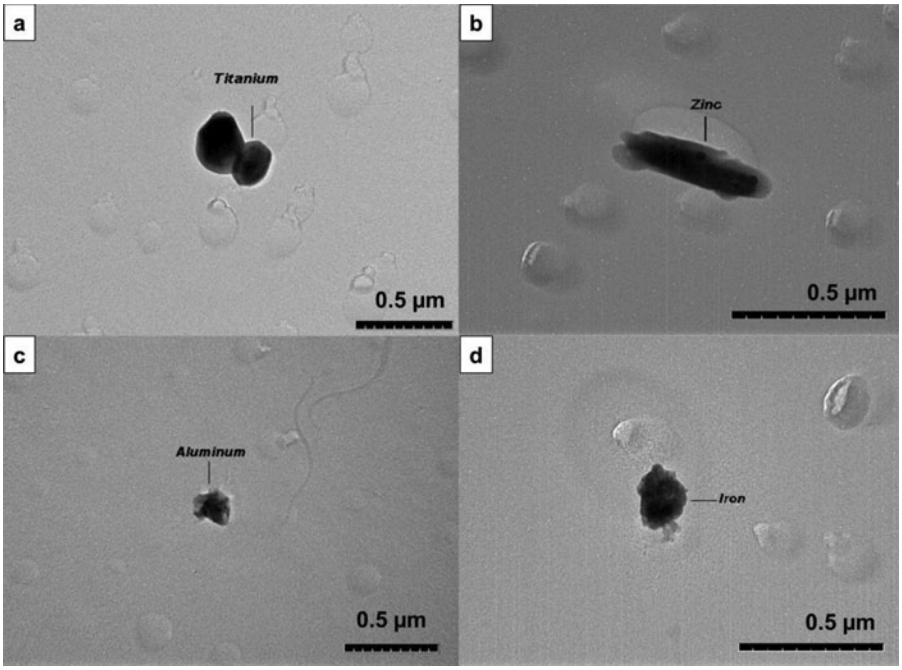

Figure 6. Submicron components of the airborne sanding dust composed of titanium (a), zinc (b), aluminum (c), and iron (d).

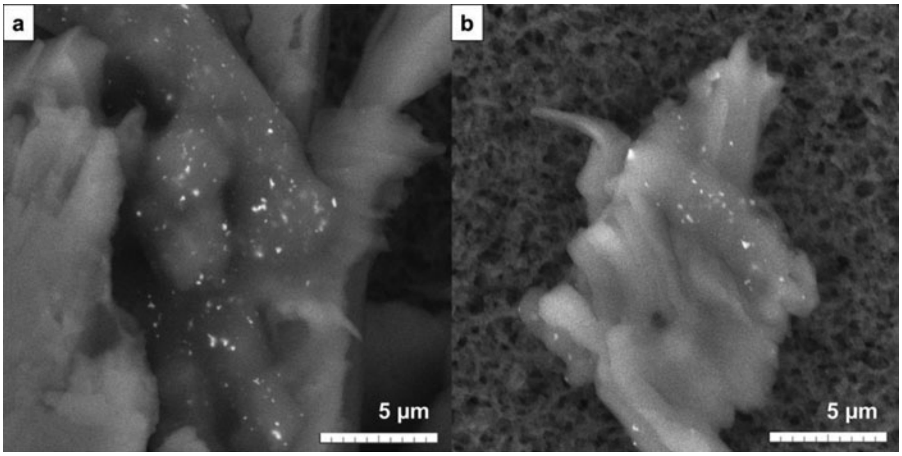

Figure 7. Micron-sized components of the airborne sanding dust with ZnO nanoparticles or agglomerates appearing as bright white specs from a personal sample (a) and area sample (b).

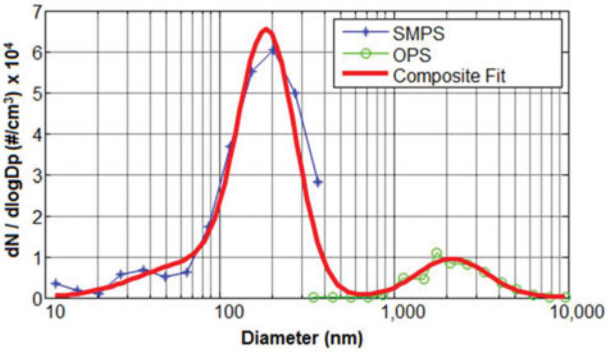

Figure 8. Background adjusted particle size distribution during spray application.

Gravimetric results for filter-based samples collected during sanding are presented in Tables 2 and 3. Total dust concentrations were relatively low compared to the Occupational Safety and Health Administration's (OSHA) Permissible Exposure Limit (PEL) (15 mg/m^3 TWA). The highest measurement (25 mg/m^3) was a personal sample collected while sanding treated wood with no controls (Table 3). This was the only sample of sixty-two analyzed that exceeded the PEL; the second highest measurement of total dust was 11 mg/m^3 (Table 2). Low concentrations of airborne zinc were detected when sanding the treated wood, whereas no zinc was detected when sanding the untreated wood. Zinc concentrations during sanding were below the NIOSH REL (5 mg/m^3 TWA). The highest concentration of airborne zinc during sanding (0.19 mg/m^3) coincided with a personal exposure measurement obtained while sanding treated wood without any dust controls. All samples analyzed for respirable dust and respirable zinc from sanding were below sample-specific limits of detection, with the highest

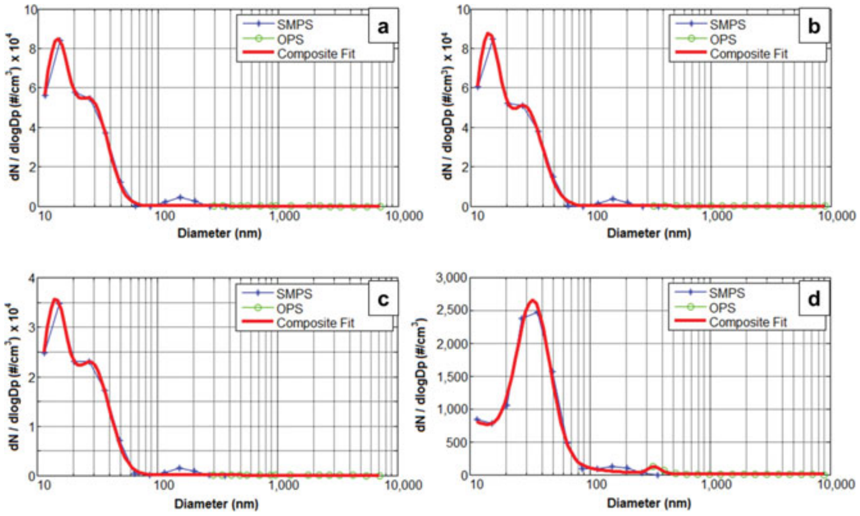

Figure 9. Background adjusted particle size distribution during sanding of: untreated wood with dust collection bag (a), treated wood with dust collection bag (b), treated wood with no dust capture (c), and treated wood with LEV (d). Multiple particle size distributions were generated for each experimental condition (a–d), each derived from a separate trial. Disparate time series data could not be merged using the Multi Instrument Manager software, thus a single representative particle size distribution is depicted for each experimental condition.

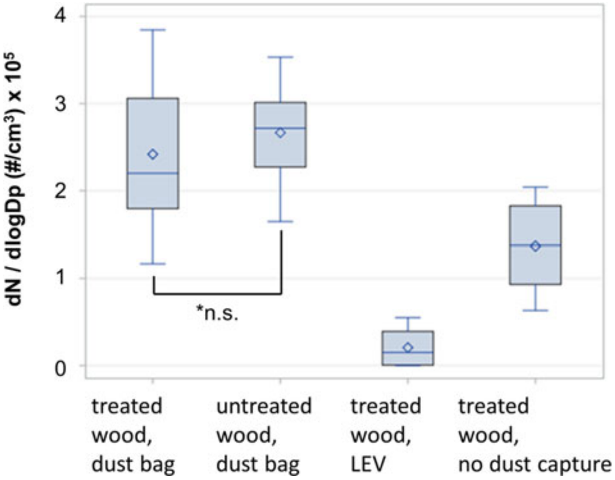

Figure 10. Background adjusted airborne nanoparticle concentrations during sanding. This figure accounts for all sanding trials, including those not depicted in Figure 9. The box plots represent the summation of all nanoparticle number concentrations for each condition, corresponding with the areas below the curve and left of the 100 nm cutoff in the prior figure. The peaks in Figure 9 are for a single particle size (e.g., 28 nm), whereas the peaks shown here represent the maximum observed concentrations for all nanosized particles. * n.s. = not significant, all other differences in mean nanoparticle exposures shown in the figure were statistically significant ($p \leq 0.05$).

limits of detection for each test condition presented in Tables 2 and 3. In terms of mass, zinc exposures were in some cases two orders of magnitude higher during spray application of the coating vs. sanding.

Measureable exposures to dust and zinc occurred while using the dust collection bag, whereas all samples collected with LEV were below detectable limits shown in Tables 2 and 3. Personal exposure to total dust while sanding treated wood with LEV was below 1.7 mg/m^3, a reduction of at least 80% relative to the median dust level under the same conditions but without any dust capture (8.5 mg/m^3).

Discussion

The highest mass-based exposures to total particulate and zinc occurred during spraying. The mean respirable zinc concentration during spraying approximated the concentration of ultrafine ZnO that humans were exposed to for 2 hr per day for 3 days without measurable effects previously.[23] SEM showed that the aerosols collected during spraying consisted of ZnO NOAA positioned on the surface of, or contained within much larger organic globules of the sealant.

Table 1. Exposure during spray application.

N	Sample Type	Analyte	Mean Conc. (mg/m^3)	Std. Dev.	95% CI
24	Area	Aerosol	96	14	90 – 102
		Zinc	2.3	.34	2.1 – 2.4
6	Personal	Aerosol	84	9.2	75 – 94
		Zinc	2.0	.19	1.8 – 2.2
		Respirable aerosol	20	3.5	16 – 24
		Respirable zinc	.49	.090	.39 – .59

Compared to spraying, a greater proportion of the aerosols counted during sanding were nanoparticles. In addition, a greater number of nanoparticles were counted during sanding even though mass concentration was almost two orders of magnitude less than during spraying and total particle counts were similar. Others have found electric vacuum and sander motors to be a major source of nanoparticles.[33,34,37] In this study, the sprayer was located outside the test chamber where particles produced by the motor would not contribute to aerosols measured. The fan, air sampling pumps, sander, and the vacuum used for LEV were inside the test chamber and could have contributed to particles counted, but conditions were constant except LEV use.

In agreement with prior findings,[36] sanding treated wood did not release excess nanoparticles compared to untreated wood, even though ZnO was only detected when sanding the treated wood. These results suggest that the nanoparticles in the sealant did not contribute significantly to free nanoparticles exposure during sanding, although the composition of nanoparticles detected by SMPS during sanding remains uncertain.

The source of the fine metallic particles present during sanding could not be determined, although the zinc is likely to have originated from the coating, the titanium from the plywood, the aluminum from the sand paper grit, and the iron from the sand paper or the sander itself. Prior studies have shown that incidental ultrafine copper particles generated from electric vacuum and sander motors contribute substantially to nanoparticle emissions.[33,34,36,37] The same was expected for this study but copper nanoparticles were not detected. Nanoparticle number concentrations detected by SMPS were orders of magnitude lower when using LEV vs. the dust collection bag and no dust capture. Given that LEV was directly connected and sealed via a hose to the portion of the sander where dust was emitted, LEV would not be expected to capture the majority of incidental ultrafine particles generated by the electric motor. This suggests that the observed reduction in nanoparticle number concentration can be attributed to effective collection of the sanding dust and that the nanoparticles detected during sanding were not primarily composed of ultrafine particles generated by the electric motor.

Although it was difficult to quantify nanoparticle release in relation to elemental composition, the EM

Table 2. Area exposures during sanding.

		Total Dust			Total Zinc		
Wood	Dust Capture	<LOD (n[a])	Median (mg/m^3)	Range (mg/m^3)	<LOD (n[a])	Median (mg/m^3)	Range (mg/m^3)
Untreated	Bag	0/12	1.7	.96–3.4	12/12	—	<.019
Treated	None	0/12	5.1	1.4–11	0/12	.050	.019 – .085
	Bag	1/12	1.5	<.88–4.3	5/12	.019	<.017 – .031
	LEV	12/12	—	< .84	12/12	—	<.017

[a] (# of non-detects / # of samples) Limits of detection varied by volume of air sampled for each measurement. The highest detection limits for a given condition are denoted by "<" in the table, where applicable.

results provide valuable insight into the transformation of ZnO nanoparticles at different stages of the product life cycle. ZnO NOAA were first observed as agglomerates in the bulk sample, then embedded within or on the surface of larger particles during spraying and sanding. These results demonstrate the different forms of ZnO NOAA construction workers would be exposed to during handling of the initial product, spray application, and subsequent sanding of a coated substrate. Whether the presence of nanoscale ZnO in mixed aerosols affects toxicity merits further investigation. As discussed in the supplemental online material, currently there are no established occupational exposure limits (OELs) for nano-ZnO.

Effectiveness of controls

LEV was an effective means of minimizing exposure to airborne particulate during sanding. Estimated reductions in airborne nanoparticles (∼90%) and personal dust exposure (≥80%) were commensurate, and LEV reduced exposures to a greater extent than the dust collection bag provided with the sander.

It was unexpected that sanding with the dust collection bag generated significantly higher nanoparticle concentrations than sanding without any dust capture. The reason for this finding is unclear. Specifications for the dust collection bag were unavailable, despite a request to the manufacturer. Further investigation may be warranted because these bags are in wide use, representing the sole control device packaged with many power sanders.

The results presented on LEV performance support prior research demonstrating that engineering controls can effectively reduce exposure to nanomaterials.[38–41] The hierarchy of controls is essential to nanotechnology risk management in the workplace and recommended internationally.[42,43] Respirators have been shown to be effective in this context.[44–46] Although inhalation was the focus of this study, dermal exposure, particularly during spray application without engineering controls, should not be discounted. Research suggests that there is little to no penetration of nano-ZnO through healthy skin,[47–49] yet damaged skin is common among construction workers. In addition, the product tested contained propylene glycol,[50] which can damage the skin and is used to enhance dermal penetration of pharmaceuticals.[51,52]

Strengths and limitations

This study is the first to evaluate operator exposure to ZnO NOAA during spray application of a wood sealant. In addition, a multifaceted exposure assessment approach was employed, following recommendations from NIOSH and an international workshop on the harmonization of measurement strategies.[53,54] A third strength of the study was the use of a containment chamber, enabling

Table 3. Personal exposures during sanding.

		Total Dust			Respirable Dust	
Wood	Dust Capture	<LOD (n[a])	Median (mg/m^3)	Range (mg/m^3)	<LOD (n[a])	Range (mg/m^3)
Untreated	Bag	0/3	2.0	2.0–2.6	3/3	< 1.3
Treated	None	0/5	8.5	2.8–25	3/3	< 1.1
	Bag	1/3	1.9	<1.9–4.4	3/3	< 1.2
	LEV	3/3	—	< 1.7	3/3	< 1.2
		Total Zinc			Respirable Zinc	
Wood	Dust Capture	<LOD (n[a])	Median (mg/m^3)	Range (mg/m^3)	<LOD (n[a])	Range (mg/m^3)
Untreated	Bag	6/6	—	< .037	3/3	< .027
Treated	None	1/6	.048	<.022–.19	3/3	< .022
	Bag	6/6	—	< .037	3/3	< .026
	LEV	6/6	—	< .033	3/3	< .024

[a] (# of non-detects / # of samples) Limits of detection varied by volume of air sampled for each measurement. The highest detection limits for a given condition are denoted by "<" in the table, where applicable.

isolation of the tasks being performed from background and other sources.

Conversely, sampling in the chamber may have overestimated exposures compared to performing the same tasks outdoors or in a larger enclosed space. Other limitations relate to apparent discrepancies in the results, stemming from difficulties in the integration and interpretation of data derived from multiple instruments. For example, despite high number concentrations of nanoparticles detected by SMPS, few individual nanoparticles were observed in EM samples. For this reason, it was not possible to make definitive conclusions about the morphology and composition of nanoparticles detected by SMPS. This could be due to nanoparticles agglomerating with each other or larger particles during sample preparation or collection on filter media. Similarly, no respirable mass was detected using gravimetric methods, despite high numbers of respirable particles detected with the real-time instruments. This might be explained by "phantom particles" contributing to SMPS counts,[55] however the fact that airborne nanoparticles number concentration decreased substantially with LEV suggests that airborne nanoparticles were in fact generated while sanding. The difference between SMPS and gravimetric results may also be explained by the relatively high limits of detection for respirable mass due to short sampling times and the fact that nanoparticles have negligible mass. While the location of equipment inside the chamber is a limitation of the study design, the use of equipment, with the exception of LEV, was held constant for all trials. The LEV was unlikely to capture particles emitted from the sander motor or other equipment and the lowest particle number concentrations were measured when sanding with LEV, suggesting the equipment did not contribute significantly to measured particle emissions. Finally, in consideration of prior studies, it was unexpected that higher numbers of larger particles were not detected by the OPS during sanding. It is possible that the OPS was compromised by the liquid aerosol generated during prior spray application, leading to an undercount of particles >420 nm not detected by the SMPS during sanding.

Conclusions

Mass-based OELs for ZnO were not exceeded during spraying or sanding, although there are currently no OELs for nano-sized ZnO. Airborne nanoparticles above background concentrations were detected during both spraying and sanding activities. Spraying generated higher mass-based concentrations, but higher number concentrations of nanoparticles were observed during sanding. Particle size distributions and number concentrations were similar for treated and untreated wood. The elemental composition of the nanoparticles detected remains uncertain, though the EM results illustrate differences in ZnO exposure across the product life cycle. Contrary to prior studies, our observations suggest that the sander motor might not have been the primary source of airborne nanoparticles generated during sanding. LEV effectively reduced exposure to airborne nanoparticles during sanding.

Recommendations

Occupational health professionals and workers would benefit from the development of health-based OELs for comparison with ZnO nanoparticles exposure measurements. Until OELs and additional information on the toxicology of ZnO nanoparticles become available, it may be prudent to reduce exposure to the extent feasible. Additionally, research on the toxicology of ZnO nanoparticles in mixtures and an agglomerated state would enhance the ability to perform risk assessments since NOAA are often used in mixtures or added to materials. Additional work is needed to identify effective controls that are practical in construction, and to assess potential release of free ZnO nanoparticles, and other nanoparticles, during all phases of product life cycles. Manufacturers should include information on NOAA in safety data sheets and product documentation so that employers and users can perform a risk assessment. The hierarchy of controls can be effective in reducing exposure to engineered nanomaterials and should be followed.

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