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The Feasibility of Studying the Health Implications of Surface Beryllium Contamination: A Review of Eight Industries

ABSTRACT: It has been suggested that beryllium can enter through either intact skin or breaks in the skin to initiate sensitization. Therefore, it is essential that all possible pathways of exposure be considered when assessing a work site for potential exposure to beryllium. A meta-analysis was done on the data from eight different industries that use beryllium or beryllium alloys, which were surveyed by Occupational Safety and Health Administration contractors, with reports being issued. During those visits, measurements were made to characterize worker exposure to skin and surface contamination levels of beryllium. Surface contamination evaluation followed the well-established protocol for lead surface contamination measurements using NIOSH Method 9100 and hand contamination using NIOSH Method 9102. It was found that both the arithmetic and geometric means of the surface samples were significantly higher in work areas as compared to administrative areas. Skin samples were also higher for individuals in production areas than for those in non-production areas. If skin contamination is a route of sensitization, these results would mean that past studies of beryllium exposure and subsequent disease might have been confounded by the lack of skin exposure data.

KEYWORDS: beryllium, skin contamination, industrial processes

Introduction

The measurement of one's exposure to total airborne beryllium dust alone might not be the best predictor of chronic beryllium disease (CBD) [1]. The particle size, surface area, number of particles, solubility, and chemical form of beryllium involved might all be relevant to the development of disease. It has also been suggested that beryllium can enter through either intact skin or breaks in the skin to initiate sensitization [2]. Recent studies have shown that particles less than 1 μm in diameter can penetrate intact skin that has been flexed [3]. Past epidemiology studies have not addressed this issue. Therefore, the case should be made that future studies should consider all possible pathways of exposure when assessing a work site for potential exposure to beryllium. One question that can be addressed now, which is pertinent to this issue, is whether, in a previous sample of industries using beryllium, the surface contamination level is sufficient to result in measurable skin exposure.

Background

Eight sites using beryllium or beryllium alloys were visited by Occupational Safety and Health Administration (OSHA) contractors who provided information on beryllium exposure and its control [4]. As part of the evaluation, both skin and surface sampling were done in each of the facilities. In summary, there were three machining plants, a metals recovery plant, a dental laboratory, two smelters, and a ceramics facility. A brief description of each operation follows.

Plant 1: Aluminum Beryllium Metal Machining Plant

The company was located in a 5000 ft^2 manufacturing facility and specialized in machining high tolerance aluminum, beryllium, aluminum-beryllium alloy, and other metals. The company operated three eight-hour shifts per day, five to six days per week. The facility had 11 full-time employees. Eighty percent of the machining was performed on aluminum, 19.9 % on aluminum beryllium, and 0.1 % on beryllium metal.

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Plant 2: Beryllium Recovery Plant

This recycling facility is recognized for its metals recovery capability in the automotive, electronic, jewelry, and metal coating and fabrication industries. Copper beryllium's unique properties make it a material of choice for certain specialized electronics applications. Due to their high strength, high conductivity, and resistance to elevated temperatures, electronic component manufacturers make extensive use of copper beryllium alloys in connectors for automotive, computer, telecommunications, and information transmission equipment. Copper beryllium alloys are often present in the input products and specialized electronics systems that are introduced into metal recovery operations at this facility.

The company was located on a 160-year-old site that originally operated as a sawmill and textile mill in 1841. The site, approximately 48 acres with a total size including all buildings of approximately 110 000 ft^2, has operated as a precious metals recycling facility since 1973. The company specializes in the business of buying and processing materials that contain precious metals (gold, silver, platinum, palladium, ruthenium, rhodium, and iridium) and selling the metals recovered from these materials. Secondary materials containing precious metals are received from a variety of industries and are assayed and prepared for shipment to an appropriate off-site smelter. The facility process capabilities are characterized as follows: material receipt and handling, mechanical preparation, granulation/shredding, thermal reduction, ball milling, screening, blending, melting, drying/grinding, and electrowinning.

The company operated primarily with one eight-hour shift per day, five days per week, 50 weeks per year. The company employs approximately 110 personnel, of which 75 are employed at the location visited, with an estimated 18 full-time workers reportedly having a potential for beryllium exposure. The facility processes approximately 15 000 000 pounds of electronics scrap per year and was reportedly at 50 % of capacity at the time of the visit.

Plant 3: Beryllium Ceramics Plant

The company was originally founded in 1956 as a beryllium oxide manufacturer. The company was a pioneer in the research and development of high purity oxide ceramics. In the early 1960s it developed the technology to press, extrude, and fire BeO in both standard and custom shapes to suit a wide variety of applications. In the late 1960s and early 1970s, precision ceramic grinding machinery was installed, along with the technology and equipment needed for drilling, metallizing, lapping, polishing, and dicing operations. During the period of peak production, the company operated with a schedule involing three eight-hour shifts and employed over 500 workers.

Plant 4: Beryllium and Aluminum Beryllium Metal Alloy Machining and Fabrication Plant

The company was located in a 100 000 ft^2 manufacturing facility and specialized in high tolerance beryllium metal and aluminum beryllium machining. The company operated two eight-hour shifts per day, five to six days per week. The facility had 214 employees, of whom 110 worked in the production area of the machine shop, with 71 on the day shift and 39 on the evening shift. Approximately half of the production area workers machined beryllium at least part of the time. However, the office workers also, on occasion, visited the beryllium work area. Because all employees had access to the production area, all employees were considered as beryllium exposed.

The company had over a dozen lathes and two dozen computer controlled mills, all of which could be used to process beryllium-aluminum alloy, beryllium, or aluminum. Some of the milling machines were enclosed, but all had ventilation supplied. Lapping, grinding, deburring, plating, heat treating, and manual milling were also performed as needed, along with a submerged electrical wire discharge cutting process called electric discharge machining. The pieces worked on ranged in size from a centimeter to several meters in length. The components produced were largely for defense and aerospace applications. Multiple pieces might be produced hourly, but some pieces require months or even years for completion, depending on their complexity and size. There is no standard production flow, with work going from one machine to another. Some pieces could be produced using a single machine, whereas others might require several processing steps on multiple machines. In addition to the machining, there was an optics department that specialized in making high tolerance mirrors, primarily with nickel, but on a beryllium substrate, although beryllium parts were also lapped there. There was a gas bearings department that also used beryllium.

Plant 5: Dental Laboratory

The laboratory was located in a 300 ft^2 suite in the basement of a 35-year-old building in a health sciences school. The lab operated one eight-hour shift per day, five days per week. The facility had one employee on the day shift.

There were three rooms joined together in this laboratory. The primary work location was the grinding lab. A bench with a spinning wheel was located there for the technician to operate. Most of the technician's time was spent in this room. Next to this room was the casting room where the metal was melted and the casting was done. This room could be accessed only through the common lab area, in which no beryllium work was done. Several other technicians worked in this common room from time to time. There was no physical separation between the rooms, and they shared the same ventilation system.

Plant 6: Copper Beryllium Alloy Stamping Plant

The company was located in an approximately 20 000 ft^2 manufacturing facility and specialized in the precision stamping, forming, and plating of copper beryllium parts. The company operated one nine-hour shift per day, four and a half days per week. The facility had 22 employees. Copper beryllium was not used during every shift for any of the machines, and might be used for only part of any given shift.

Materials were generally received as plates or strips of beryllium copper. The alloys used were numbers 25, 10, 17410, and 7717. Material was stored on-site until required. The operators retrieved the raw material from storage and loaded the machines with the appropriate dimensions and material specifications. "Stamping" is a term used to refer to various press forming operations including coining, embossing, blanking, and pressing. The operations most commonly done were blanking, piercing, forming, and drawing. These operations were done with dedicated tooling, also known as hard tooling. Hard tooling is used to make high volume parts of one configuration. In the production process, a die is selected depending on the pattern required, and the material can be placed into either the stamping machine or the forming machine, or sometimes into both machines. The stamping machine is used to cut out patterns, much as a cookie cutter would. The forming machine is used to put bends and depressions into stamped parts. Workers sat at the machines, manually controlling the process and handling parts as they were produced. Manufactured parts coming from the machines were then placed into containers. These parts could be heat-treated, cleaned, dried, and plated. The assembly of parts could be done if required, and quality control was performed. Finished parts were then packaged and shipped to the customer.

Plant 7: Copper Beryllium Casting Plant

The company's core expertise was melting and casting beryllium copper and other beryllium containing alloys; over 1×10^6 pounds were melted and casted annually. Inert gas cover and degassing technology were utilized for melting and casting operations. Automatic furnace controls and ingot mould conveyers produced standard five-pound and two-pound ingot configurations. All beryllium alloys were manufactured utilizing either pure metallic beryllium or certified beryllium copper master alloy. No recovered, recycled, or purchased scrap was utilized in the standard production processes.

The company produced BeCu, BeNi, and BeAl casting and master alloy ingot; BeCu semi-continuous as-cast 21 in. diameter input billets; BeCu forged and turned precision input billets; BeCu forged rod and plate products; and BeCu large diameter hollows and custom forged products. The fractional content of beryllium in the products varied from 0.35 % to 10.5 % in the master alloy.

Administrative work was done on the day shift. Casting was done during the night shift, with only one casting done on an average night. Three people worked the night shift. Copper ingots and copper beryllium master alloy ingots were loaded into the pot before melting began. The ingots were mostly unloaded by hand into the pot from crates hoisted into place above the melting pot by the forklift. All three workers were involved in removing the approximately 12 in. long ingots from the crates and filling up the furnace.

Once the furnace was loaded, heat was applied and the melt began. Dross floats to the surface during the melt and needs to be skimmed off. Also, the melt needs to be sparged in order to ensure complete mixing (sparging involves the introduction of gas into the furnace to stir the melt). The majority of the shift, however, was spent waiting for the melting to be completed.

When the melt was complete, the pour began. The furnace was tilted, in-place, to pour the molten metal into moulds. The type of mould selected depended on the customers' specifications and could change from pour to pour. A number of moulds were available, as noted above in the list of products. The cast materials were allowed to cool and were then placed into containers for shipment, by either hand or forklift. Larger moulds could be trimmed using a band saw before being shipped. The band saw could have a lubricant stream that was used during cutting.

Plant 8: Non-ferrous Forging and Machining Plant

The company was a manufacturer and distributor of forged copper, including beryllium copper, chrome copper, and aluminum bronze in plates, blocks, bars, or rings, and other copper alloy forgings for plastic mould tooling, resistance welding applications, metal melting liners, end caps, and bearing components. Metal conversion services were also provided to customers who supplied their own metal materials. Services included the recommendation of alloys, properties, and designs for tooling applications; concurrent design; fabrication; heat treating to meet various temper and grain size requirements; near net shape processing; rough or finish machining; testing; and technical certifications.

The company had over 50 000 ft^2 of manufacturing space, and the operating schedule involved a single ten-hour shift, four days per week, with a total of 40 workers employed. There were 25 employees in the production shop, with 5 having direct involvement with CuBe alloys.

Methods

Surface Samples

Surface contamination evaluation followed the well-established protocol for lead surface contamination measurements, as did skin sampling. The exact surface locations were randomly selected to reflect all types of areas with which a worker might come in contact. These survey data were meant to answer the question of what the range of concentrations might be, rather than what specific processes and operations contributed to each sample. Surface sampling was done by marking off a 100 cm^2 area, using a plastic template with a square hole 10 cm on each side that was placed on the surface, with the corners marked with a moistened towelette. The template was then removed and the perimeter lined out with the same moistened towelette. The inside of the square was then wiped according to NIOSH Method 9100 [5], and the towelette was placed in a screw-top glass vial for analysis. The template was cleaned before reuse. Workers' gloves were also wiped, but at a separate time from the hand wiping so as not to disrupt the ordinary wear of the glove immediately before skin sampling.

Hand Wipe Samples

Hand wipe samples taken according to NIOSH Method 9102 [5] were obtained by asking study participants to wipe their hands before the end of their shift or during the shift, at least two hours after their last hand washing. They were instructed to lift a fresh wet wipe from an open container and to thoroughly wipe both hands (including the front and the back, up to the wrists, and each finger), removing as much visible dirt as possible. The wipe was then placed in a labeled screw-top glass vial. The hand wiping exercise was supervised and timed for 30 s by the investigators in order to ensure consistency from subject to subject. A pen tracing on graph paper with a millimeter scale as far as the wrist, where the wiping stopped, was taken of the subject's right hand in order to estimate the total surface area of the participants' hands. The number of 1 mm blocks on the graph paper within the tracing was counted and multiplied by four to give the total surface area of both hands. The concentration of beryllium on the workers' hands is reported in micrograms of beryllium per 100 cm^2 of estimated hand surface area.

Laboratory Analysis

OSHA's Salt Lake City Laboratory performed the laboratory analysis. Gravimetric results were reported to the nearest microgram. The beryllium mass was determined for surface wipes and hand wipes using NIOSH Method 7102 [5], graphite furnace atomic absorption spectrophotometry. Field blanks made up 10 % of the samples analyzed, and another 5 % were media blanks.

Results

It appears from Fig. 1 that hand contamination follows a general trend with surface contamination in each plant, although not with quite the same proportion in each plant. With an approximately 300 cm^2 area on

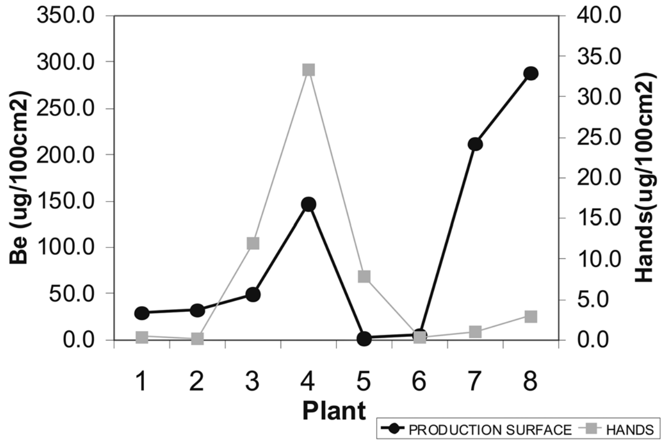

FIG. 1—*Average beryllium concentration on surfaces in production areas and on workers' hands.*

each worker's hand, there could be as much as 100 μg of beryllium, on average, on each of the hands of the workers in Plant 4. This figure would seem to indicate that surface contamination might generally be an indicator of the risk of dermal exposure but only a rough approximation of dermal exposure, and that direct measures of dermal exposure might be necessary for epidemiologic studies. Table 1 demonstrates that the surface contamination is usually higher in production areas than in non-production areas, which should not be surprising. The difference between many of the plants is by orders of magnitude. The geometric mean was found by taking the average of the log of the concentration values, averaging the log values, and then taking the antilog of that mean. The geometric mean tends to lessen the effect of a small number of extreme values. In Table 2 the large difference between the geometric and arithmetic means for the surface samples indicates that a broad range of contaminated surfaces were seen in each of the plants.

Discussion

In plant 1 there were five major machining centers, which together contained four computer numerically controlled milling machines and one lathe, all of which were enclosed, automated machining equipment. All equipment was operated by machinists, three of whom were evaluated during this site visit. Additionally, after machining, the workers performed deburring on certain products as a manual process for which specialized controls were used. Of all the plants in this study, only in plant 1 was there a glove policy in place requiring the strict use of gloves. As can be seen from the data, this might be part of the reason that exposures and the potential for exposures might have been lower here than for most other plants. Enclosures around the equipment almost certainly contributed to the lower levels of surface contaminations as well. However, this is still somewhat speculative, because the objective of the reports cited was to simply document what existed, not what caused it to exist.

In plant 2 there were four operations at the site having a potential for beryllium exposure: mill/blending, thermal reduction, melting, and shredding mill. It is important to note that workers in the aforementioned processes and operations have potential exposures to lead and cadmium. The facility has characterized exposures to lead and cadmium and has implemented controls, personal protective equipment, and work practice control requirements in accordance with applicable regulatory standards. The lower levels of beryllium surface contamination and dermal exposure might be due to the already heightened awareness of the potential for toxic exposures to other materials.

At the time of the survey, plant 3 was operating at an estimated 10 % of capacity but had over 35 000 ft^2 of manufacturing space. It operated with a single eight-hour shift and had 14 workers employed. The low production levels, more than any exceptional controls, were probably responsible for the less than maximal surface contamination relative to the other plants.

Plant 4 had a history of diagnosed beryllium disease in its workers. There was no distinct separation between production and office areas, allowing employees to come and go between the areas, possibly spreading contamination. Although some control measures had been taken, other places where controls should have been in place showed a lack of adequate control. This might be why the plant had one of the highest levels of contamination.

TABLE 1—*Individual plant results for beryllium contamination.*

	Arithmetic Mean, μg/100 cm^2	Standard Deviation, μg/100 cm^2	Number of Samples
Plant 1			
Surface production area	29.5	75.1	16
Surface non-production areas	1.5	2.0	9
Under gloves production area[a]	0.5	0.4	7
Plant 2			
Surface production area	32.7	128.0	29
Surface non-production areas	0.1	0.1	6
Hands production area	0.2	0.3	25
Hands non-production areas	<0.1	<0.1	3
Plant 3			
Surface production area	49.6	84.8	22
Surface non-production areas	0.5	0.6	7
Hands production area	11.9	20.5	12
Hands non-production areas	2.3	2.4	2
Plant 4			
Surface production area	147.2	564.8	88
Surface non-production areas	2.4	3.4	18
Hands production area	33.5	44.3	6
Plant 5			
Surface production area	2.0	4.1	18
Hands production area	7.8	-	1
Plant 6			
Surface production area	5.4	14.3	34
Surface non-production areas	<0.1	-	3
Hands production area	0.3	0.5	11
Plant 7			
Surface production area	212.0	185.3	13
Surface non-production areas	110.2	244.5	12
Hands production area	1.0	1.5	6
Plant 8			
Surface production area	288.7	1022.1	21
Surface non-production areas	0.4	0.5	4
Hands production area	3.0	4.7	19

[a]These samples were taken from hands after gloves were removed, in an area where workers wore gloves.

Plant 5 had only one employee working on a low number of very small objects containing beryllium. Controls were virtually absent. The low production was the probable reason that this plant had lower contamination levels.

The alloy stamping plant (plant 6) had the lowest contamination levels and was the least energetic of those observed (it had the lowest air contamination level of any of the production areas, as well). Controls directed at reducing beryllium exposure were absent. However, copper beryllium was not used on every shift for any of the machines and was usually used for only a part of any given shift. The lower surface and dermal contamination is probably due to those latter effects rather than to the engineering design of the process.

Plants 7 and 8 were operated as metal refineries without reference to the toxicity of beryllium. Controls were minimal. It is presumed that the higher surface contamination was reflective of this.

Epidemiological studies to date have neglected or dismissed the possible effects of skin exposure, either drawing conclusions without measuring [6] or not including the analysis [7] of the skin exposure when measured (3.8 versus 1.1 μg/100 cm^2 production versus nonproduction areas) in plants with known disease. The current American Conference of Governmental Industrial Hygienists threshold limit value for beryllium addressed only air exposure because there are no studies with data on skin exposure to

TABLE 2—*Comparison of arithmetic and geometric mean surface level beryllium contamination.*

	Arithmetic Mean, μg/100 cm^2	Geometric Mean, μg/100 cm^2
Plant 1		
Surface production area	29.5	3.9
Surface non-production areas	1.5	0.6
Plant 2		
Surface production area	32.7	2.3
Surface non-production areas	0.1	(<0.1)
Plant 3		
Surface production area	49.6	21.8
Surface non-production areas	0.5	0.3
Plant 4		
Surface production area	147.2	3.3
Surface non-production areas	2.4	0.2
Plant 5		
Surface production area	2.0	0.9
Plant 6		
Surface production area	5.4	0.7
Surface non-production areas	<0.1	<0.1
Plant 7		
Surface production area	212.0	94.4
Surface non-production areas	110.2	22.3
Plant 8		
Surface production area	288.7	17.0
Surface non-production areas	0.4	0.2

reference. What needs to be addressed is the potential confounding effect of skin exposure, which could lead to sensitization and, with subsequent lung burden (from air exposure), then lead to lung disease.

In order to consider the possibility of establishing an epidemiologic investigation of the relationship between skin exposure and sensitization, it is necessary to have certain conditions. It is proposed that these conditions are as follows:

1. A source of exposure, which results in.
2. A quantifiable level of exposure across.
3. A range of exposures.
4. The range of exposures must include a putative level thought to cause an effect—in this case, sensitization.

It is obvious in this cross-section of plants performing varying kinds of work on beryllium and its alloys that there is a measurable source of exposure to the skin on the hands throughout the production areas of the plant, and often in non-production areas, thereby meeting criterion 1 above. Figure 1, which shows a similar trend between hand contamination levels and surface contamination levels, indicates that surface contamination is the likely source. The levels found by OSHA were reported with a limit of quantification of 0.1 μg. This is in agreement with recent work [8] that shows a limit of detection for surface samples of 0.05 μg. The sample of plants for the OSHA reports therefore also meets criterion 2 above.

There is a distinct difference, for the most part, for both surface and hand concentrations between the production and non-production areas, with the production areas showing significantly higher contamination. This, then, achieves the first three proposed criteria for considering an epidemiologic study of skin exposure. The fourth criterion is the most problematic, in that no one has yet proposed a level that is either proven safe or hypothesized to cause sensitization. Unfortunately, most of the plants in this report did not screen for disease, and so disease rates are not available. One recent study [9], however, notes that there is a significant difference between sensitization rates in areas of a beryllium alloy plant between production and office areas, and the levels of airborne beryllium were generally lower than any published estimate of the exposure capable of causing sensitization or disease (geometric mean = 0.003 μg/m^3 and maximum = 0.02 μg/m^3). The same paper conducted a surface sampling analysis and found that surface contamination levels (also significantly different) were 0.95 and 0.05 μg/100 cm^2 for production and office areas, respectively. If it is assumed that the production area contamination level of 0.95 μg/100 cm^2 is the cause of the sensitization, then all of the plants in the OSHA survey have sufficient levels of contamination to cause sensitization. The fourth criterion is therefore met.

The plants in the OSHA reports were informed that the measurements would be given to OSHA, but with their names removed. One might assume that these plants, in choosing to cooperate, probably represent the best of the controlled plants. Becaus plants that did not wish to cooperate were not sampled, it is impossible to say for sure what relative level of surface cleanliness and control the plants studied actually represent. However, as the levels of surface contamination and dermal exposure that did occur in the plants studied are sufficient to warrant inclusion in an epidemiological study, and given that the assumption that these plants are the best controlled is not unreasonable, an epidemiological study of the relationship between dermal exposure and beryllium sensitization seems to be clearly warranted.

Conclusion

A biologically plausible hypothesis has previously been proposed relating skin exposure to beryllium sensitization [3]. Examining the data collected for OSHA in order to evaluate the current levels of exposure in various industries using beryllium, it is clear that dermal exposures are sufficient ($>$0.95 μg/100 cm^2) to warrant a study to prove or disprove the skin hypothesis, and that sampling and analytical methods are available with sufficient sensitivity to support that study. Because sensitization seems to be an important step in the development of CBD and because the skin is a plausible means of exposure [10], unless the skin hypothesis can be dismissed, an airborne exposure limit might be difficult to support on its own.

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