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BACKGROUND REVIEW OF THE BRUSH BERYLLIUM AND DIAMOND MAGNESIUM PLANTS IN LUCKEY, OHIO

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INTRODUCTION

The Department of Energy (DOE) is conducting a program to identify and examine the radiological conditions at sites used in the early years of nuclear energy development by the U.S. Army Corps of Engineer's Manhattan Engineer District (MED) and the Atomic Energy Commission (AEC). This program, the Formerly Utilized Sites Remedial Action Program (FUSRAP), is administered by the Assistant Secretary for Nuclear Energy through the Office of Remedial Action and Waste Technology's Division of Facility and Site Decommissioning Projects. The sites of concern were owned by the Federal Government, private industry, and public and private institutions and were used primarily for research, development and production of uranium and thorium feed materials, and storage of radioactive ores and residues. Some investigations were also conducted for sites used in the plutonium development program.

The purpose of this report is to provide an historical perspective of the Luckey, Ohio, facility with respect to its owners and operators, and to investigate the activities conducted at the site in order to identify the types of materials and chemicals used. Preliminary information from an Oak Ridge National Laboratory (ORNL) survey indicated low levels of residual uranium and radium which appeared more widespread than would be expected if it were totally the result of contaminated scrap that was once used on the site. The information addressed in this report will help to determine if any alternative sources of radioactivity exist other than the contaminated scrap.

HISTORICAL INFORMATION

The facility was the site of the "M and B" Plant, which stood for Magnesium Reduction Plant and Beryllium Plant (see Figure 1). Diamond Magnesium and Brush Beryllium were both occupants of the site. Additional background information on the functions and operations performed at this site is included in this section.

Magnesium Production

There is limited information in the FUSRAP records on operation of the magnesium reduction plant by Diamond Magnesium at the Luckey site. The records indicate that National Lead Company (NL) of Luckey, Ohio, had a part in the design of the original magnesium production facility. This information was obtained from copies of blueprints, dated June and July, 1942. The FUSRAP records show that Melissa A. Laymen transferred the property to the Defense Plant Corporation (DPC) on June 1, 1942. On May 13, 1942, the Defense Plant Corporation contracted with NL, for their assignee,

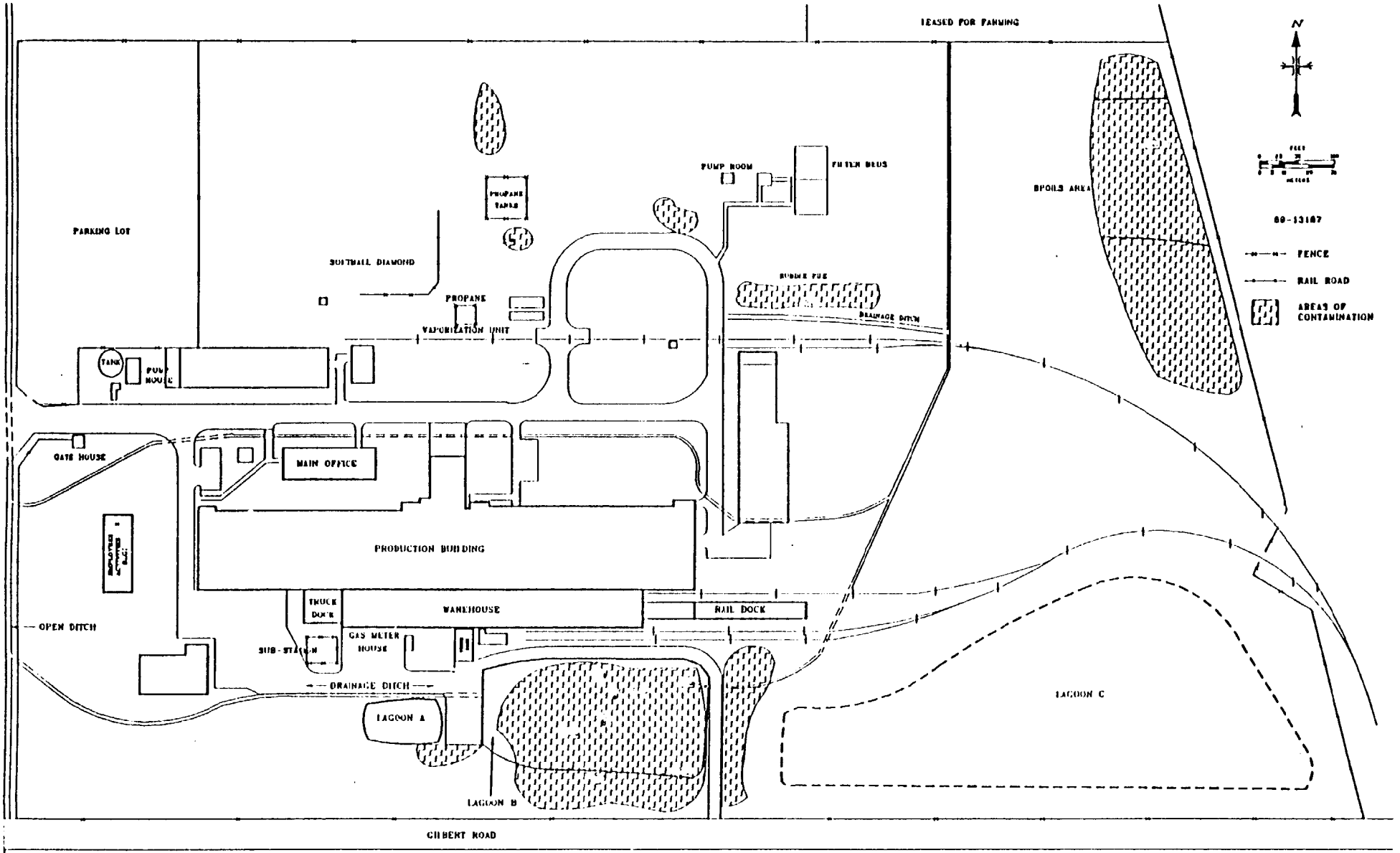

Fig. 1 the former Diamond Magnesium Company site, Luckey, Ohio

Magnesium Reduction Co., to operate the Luckey, OH, Magnesium Reduction Plant, with disbursements being made to NL. Then on May 18, 1942, the DPC entered into an agreement with Kelly Island Lime and Transport Co. to dispose of waste from the magnesium production facility into the quarry on property adjacent to the facility, in consideration of $.03/ton. No correspondence was found to indicate that the agreement was ever exercised. Both contracts are attached in Appendix A and B.

Following World War II, the magnesium plant was placed in a standby mode. The Brush Beryllium Company was contracted by the AEC for beryllium production at the site as described in the following section. According to its contract, Brush Beryllium was responsible only for the maintenance of the magnesium production facilities until they became occupied.

The magnesium production facility was reactivated in the early 1950's by Diamond Magnesium. It was during this period of operation that contaminated scrap was utilized. The first record of contaminated scrap being sent to the site was in 1951. A memo from Sam A. Rothenberg to Hanson Blatz, dated December 14, 1951, (Appendix A) described the quantities and types of materials transferred to Diamond Magnesium Company, Luckey, Ohio from the Lake Ontario Storage Area.

> "One thousand (1000) tons of scrap steel (drums and engine parts) were originally contaminated by fission products, yellow salt and concrete flooring. This scrap was measured and remelted last year (sample 600 lbs.) and the contaminant was removed during the remelting. Since the scrap was found to be usable it was sold to the Diamond Magnesium Company of Luckey, Ohio, to be used in the processing of magnesium. It will be shipped in boxcars at the rate of 10 tons (3 boxcars) per day. At the time of monitoring, loading had not begun and the measurements were made directly at the scrap pile. The following samples were used:
>
> DrumDinside covered with yellow crust.
> Beta plus gamma = 20 mrep/hr.
> Gamma = 1 mr/hr at contact inside
>
> Concrete inside drum
> Beta plus gamma = 1 mrep/hr
> gamma = .05 mr/hr at surface
>
> Steel sheet
> Beta plus gamma = 5 mrep/hr
> gamma = .05 mr/hr"

The yellow salt referenced in this memorandum was either uranyl nitrate hexahydrate or sodium diuranate, which are both intermediate compounds in the production of uranium. For the radiological units presented, the unit

"rep" is "Roentgen Equivalent Physical", which is a dosage term no longer in use. The term "mr" is "mRoentgen".

Beryllium Production

The former Brush Beryllium Company entered into Contract No. AT(30D1)D541 (Appendix B) with the Atomic Energy Commission on July 31, 1950, although the effective date of the contract was February 1, 1949. The period of performance was from 1949 until production was completed in 1958. The Department of Energy New York Operations Office administered this contract until it was transferred to Oak Ridge Operations in July, 1954. One month later the contract was transferred to the Chicago Operations Office. This office was responsible for the administration of this contract until production was completed in 1958 and the site was decommissioned in 1959 under Contract No. AT(11-1)-830.

In accordance with the terms of the contract, the AEC retained ownership of the following:

1. All property purchased by the Contractor under the contract and for which it is entitled to direct reimbursement;

2. All technical data of any kind or nature furnished or prepared by the Contractor, or developed in connection with the Contractor's undertakings under this contract;

3. All items of property including products, by-products, work-in-process, salvage, residues, wastage and scrap resulting there-from, except rejected materials and except retained duplicates of operating records.

Contract AT(30-1)-541 required Brush Beryllium to perform design, engineering, construction and management work for the establishment of a plant for the production of beryllium metal and other beryllium products. The beryllium production facility, referred to as the "M and B Plant", was constructed on land owned entirely by either the Atomic Energy Commission (AEC) or other Federal agencies. There was also a magnesium reduction facility, formerly owned and operated by the Defense Plant Corporation, located on the Luckey, OH, site. Under its contract, Brush Beryllium was required to maintain the magnesium production facilities of the M and B Plant in connection with possible future operation of the plant.

Title III of the contract outlined Brush Beryllium's responsibilities for "Maintenance of M and B Plant and Operation of Beryllium Plant". Brush Beryllium was responsible for (1) the production of beryllium metal powder from raw materials produced under this contract or furnished by the AEC; (2) fabrication by powder metallurgy of beryllium metal shapes in quantity, grades, and under specifications as directed by the AEC and from materials supplied by the AEC or produced under this contract;

(3) performance of such machining operations on beryllium metal as may be directed by the AEC; (4) fabrication of beryllium oxide crucibles as directed by the AEC; (5) performance of such other work with respect to beryllium materials, beryllium compositions, beryllium products, zirconium and zirconium composition as the AEC directed; and (6) performance of such fabrication operations on such quantities of thorium during the period commencing August 1, 1951, and ending February 28, 1953, as directed by the AEC. Appendix C provides a summary of operations at Brush Beryllium in 1950.

Site Ownership/Tenant History

Below is a chronology of the site ownership and tenant history.

1942 - Melissa A. Lehman transferred the property to the Defense Plant Corporation on June 1, 1942. Magnesium Reduction Co., assignee to NL, was the operator of the facility.

1948 - The property was quitclaimed to the United States of America by Reconstruction Finance Corporation, successor to Defense Plant Corporation by Quitclaim Deed, October 27, 1948.

1949 - The property was declared surplus and assigned to the Administrator of General Services for disposal pursuant to the provisions of the Federal Property and Administrative Services Act of 1949.

1949 - In 1949, Brush Beryllium contracted with the AEC to design, construct, operate, and maintain the "Beryllium Plant".

1961 - The "Beryllium Plant" Site was purchased by Aluminum and Magnesium, Inc. on May 19, 1961.

1968 - Goodyear Tire and Rubber Company purchased the site in 1968.

1987- The property was transferred to Motor Wheel Corporation in 1987.

ASSESSMENT OF THE SITE

In order to determine the source of contamination on the Luckey, OH, site, three different areas of information were assessed. First, the Preliminary Radiological Survey (Ref.1) performed by Oak Ridge National Laboratory (ORNL) was reviewed to characterize the site as it presently exists. Second, the characterization was compared with stereo aerial photographs obtained from the United States Geological Survey (USGS) that show the plant site in 1950, 1954, 1963, and 1969. Finally, the chemical processes used to manufacture magnesium and beryllium during the period of concern were analyzed to determine whether they could have contributed to the contamination.

During the period of Brush Beryllium operation, when the plant was converted to a beryllium extraction and production plant, ore containing beryllium was processed to obtain beryllium pebbles, which were shipped to other facilities for further processing and machining. The processing of beryllium ore to produce vacuum cast billets and beryllium oxide created waste solutions and precipitated sludges that were impounded in lagoons. All lagoons were of shallow construction, approximately 4 feet deep, and were formed by scraping the top layer of soil and construction dikes. Reportedly, the lagoon liner was compacted clay. Excess wastewater accumulated in the lagoons and, in accordance with Ohio Water Pollution Control Agency regulations, was discharged to the Toussaint Creek. As a lagoon section filled with sludge, additional ones were added. Lagoons A, B, and C (Fig. 1) were constructed and used for impoundment. A Lagoon D was constructed in 1956 but was never used (Reference 2).

Review of the Preliminary Radiological Survey

The ORNL Preliminary Radiological Survey included (1) a surface gamma scan of part of the property outdoors, (2) collection of surface and subsurface soil samples, and (3) collection of water samples. Laboratory analysis of soil samples showed concentrations of ^{226}Ra in excess of applicable DOE guidelines.

Gamma exposures of the major portion of the property ranged from 5 to 9 uR/h. Elevated readings were taken in Lagoons "A", "B" and "C" (Refer to Figure 1), with the highest measurement, 1500 uR/h, occurring in Lagoon B. Small, isolated hot spots, ranging to 800 uR/h, were found scattered over most of the property.

Six systematic and 31 biased soil samples were taken from two systematic and 19 biased locations at the site and were analyzed for radionuclide (^{137}Cs, ^{226}Ra, ^{232}Th, ^{238}U) concentrations and nonradioactive beryllium (Be) concentrations. Almost all of the biased soil samples were above DOE guidelines of 5 pCi/g for ^{226}Ra concentrations in surface and 15 pCi/g subsurface soil, with the highest concentration being 4000 pCi/g. Although specific guidelines are not given for Be, values from the ten samples analyzed appear high. The average Be concentration in soil ranges from 0.13 to 0.88 ug/g and the Be concentrations in the soil samples taken on the Luckey, Oh, site ranged from 6.8 to 6400 ug/g.

Three water samples were taken from three locations on the site: potable water taken from the plant's main office building, raw well water from the wellhead of the west well, and surface groundwater taken within 25 ft. of a known contaminated area. All samples showed permissible concentrations for radionuclides in water for unrestricted use.

The ORNL survey showed elevated gamma readings around the lagoons. However, the ORNL soil samples showed above background uranium concentrations

dispersed widely across the site and no direct correlation can be drawn between the process waste storage in the lagoons and the contamination.

Based on the results of the radiological assessment, a follow-up, detailed radiological survey was recommended by ORNL to be performed to more precisely define the extent of the contamination. This has not been performed to date.

Review of Stereo Photographs of the Site

Stereo aerial photographs obtained from the USGS taken on four different dates during the operation of the Beryllium production plant were examined. The dates the photographs were taken were August 23, 1950, September 3, 1954, May 2, 1963, and September 10, 1969. The photographs corroborate the historical information and drawings as to the existence and location of the lagoons. The photographs showed very clearly when a lagoon or landfill was active and when it had been closed and covered. In particular, the photographs show a lagoon that was constructed, but did not show up on the period drawings. However, according to a letter from Brush Wellman to the Ohio EPA in 1983 (ref. 2), Lagoon "D" was constructed in 1956 but was never used. A more detailed review of each set of photographs can be found in Appendix D.

Potential For Radiological Constituents In Magnesium Processing

Two possible sources of radiological contamination as a result of magnesium processing were investigated. The first was the possibility of radiological materials found existing naturally in combination with magnesium ores. The second possible source of contamination investigated was radiological contamination introduced as impurities during the processing of magnesium during the time period that the plant was in operation.

Dolomite was used as the magnesium-bearing ore in the Magnesium Reduction Plant. Dolomite is a double carbonate of calcium and magnesium ($CaCO_3$ $MgCO_3$), which has a theoretical magnesia (MgO) content of 22% This term is also applied to dolomite rock, a limestone that has been transformed to the mineral dolomite by hydrothermal alteration. It is mined using open pit methods throughout the United States. (Ref. 3)

The production process for magnesium metal used at the Luckey, OH, facility was the silica process, which requires calcined dolomite. The dolomite was delivered from the mine to a crushing plant where it was sized and crushed in a three stage process. From there it was fed to a rotary kiln plant where it was calcined by heating to 2,000 degrees F to drive off the water and most of the carbon dioxide.

The silicothermic process is a two-step operation. First, the calcined dolomite, ferrosilicon and alumina ore were ground, heated and briquetted. The ratios of reagents required to produce 1 pound of magnesium ore 13.15

dolomite (calcined to 6 lb) and 1.25 lb of 75% ferrosilicon. Next the briquets are charged into heated tubular retorts, which operate under vacuum. Magnesia in the calcined dolomite is reduced by the silicon, producing magnesium vapor which is crystallized in a condensing chamber. These crystals are melted into casting forms and ladled into casting forms. The capacity of the plant was 5,000 short tons of magnesium per year.

From the process description there does not appear to be any added reagents that present the likelihood of uranium contamination. The most probable source of uranium contamination in the process would be the dolomite. However, dolomite contains on the average 2 ppm uranium. (Ref. 4) The national average uranium concentration in soil is 3 ppm. (Ref. 5) Therefore, the amount of uranium contamination resulting from this process is no more than background concentration.

Potential For Radiological Constituents In Beryllium Processing

As with magnesium, two sources of radiological contamination as a result of beryllium processing are possible. The first is radiological materials found existing naturally in combination with beryllium ores. The second is radiological contamination introduced as impurities during the processing of the ore. Below is a description of the mining and processing of beryllium during the time period that the plant was in operation.

Although there are about thirty recognized minerals containing beryllium, the mineral beryl is the most widely used source in industry. (Ref. 6) Beryl is a pegmatite mineral widely distributed over the world, and generally occurs as hexagonal crystals. It is present as a minor constituent together with mica, feldspar, and other minerals including small quantities of uranium. Except in some locations where it occurs in quantities sufficiently concentrated to permit mining for its own sake, beryl is mostly produced as a by-product from mining other minerals. Only crystals large enough to permit hand sorting or cobbing are recovered.

Simple mining methods are used to excavate beryl-containing granite pegmatite (Ref.7). These pegmatite deposits are usually mined primarily for feldspar, mica, quartz crystals, lithium-bearing minerals, and gem stones. Beryl, a crystalline beryllium-aluminum-silicate ($3Be\ Al_2O_3\ 6SiO_2$), is present in very low concentrations in these pegmatites, or granitoid deposits; 100 tons of rock yields about 1 ton of beryl. (Ref. 8) Beryl-containing ore from pegmatites is crushed and hand-sorted, sometimes on belts, to reclaim beryl and other mineral values.

Upon receipt at the processing plant, the beryl was crushed to a two inch mesh size and washed with water to remove gangue, or extraneous rock material, that still remained with the crystals after hand sorting. Beryl ore is chemically inert material under normal conditions at atmospheric pressure but can be chemically attacked at elevated temperatures. Completely melting beryl, and quenching the melt in cold water destroys the

crystalline structure of the original complex beryllium aluminum silicate and results in glass which has a BeO crystalline structure that exhibits greatly increased reactivity with hot sulfuric acid. After dissolving the beryl in sulfuric acid, the solution was centrifuged to remove the silica, SiO_2. Ammonium sulfate was then added and the solution was again centrifuged to remove crystallized ammonium alum, $NH_4Al(SO_4)_2$. Finally, hydrogen sulfide was added to precipitate out the residual iron as iron sulfide. The resultant solution was crystallized and ignited in a furnace to produce high purity beryllium oxide. (Ref. 9).

The beryllium oxide was converted to beryllium metal by addition of ammonium biflouride, NH_4HF_2. The resultant beryllium fluoride was reduced with magnesium to beryllium metal and magnesium fluoride slag.

From the process description, there does not appear to be any added reagents that present the likelihood for introduction of uranium contamination. However, as cited in one reference, small quantities of uranium are found to coexist with the beryl ore in the pegmatite formations, predominantly in Brazil, which was a major source of beryl during the 1940's (Ref. 7). This is also supported by the fact that Brush Wellman, Inc., the former Brush Beryllium Company, presently recovers uranium oxide as a by-product from its beryllium extraction process at its plant in Delta, Utah (Refs. 10, 11). Records for the Brush Beryllium Plant in Luckey, OH, show that beryl ore was obtained from Columbia. Archived records for the Defense Plant Corporation indicated that the Brush Beryllium Plant in Lorain, OH, obtained their beryl ore from Brazil and Argentina. This leaves open the possibility that the Luckey, OH, facility also received ore from these other South American countries.

At the mining site an attempt was made to remove the beryl crystals from any gangue material through hand cobbing. The efficiency of this operation is not known. However, it appears likely that some extraneous materials remained with the ore, and this material had a high probability of containing uranium. There are two possibilities for where the uranium could have been removed from the process and disposed on the site once the beryl reached the plant. The first and most likely would be during the crushing and water washing step as the material entered the process. The uranium-containing fines would have been separated from the beryl crystals and then discarded with the washwater or filtered/settled out of the recycle wash water and discarded. The second possibility would arise if some of the uranium containing material remained with the beryl and was dissolved in the sulfuric acid. If the processes remained acidic, uranium would have stayed in solution and would have been discarded with the liquid effluent from the crystallizer in the final stages of the process.

Finally, from the analysis of the soil samples discussed earlier in the Preliminary Radiological Survey by ORNL, a correlation appears to exist between the samples with high beryllium concentration and the samples with high uranium concentration. A graphical representation of the

concentrations in the 10 samples analyzed for both beryllium and uranium is shown in Figure 2. In order to draw a definite conclusion, a more rigorous statistical analysis must be performed on a greater number of samples. However, the preliminary results seem to indicate that the uranium contamination and the beryllium contamination came from the same source, which would be the beryl ore brought to the plant site for processing.

SUMMARY

The potential sources of radioactive contamination at the site that were identified are the uranium contaminated scrap metal purchased by Diamond Magnesium from the Lake Ontario Ordinance Works and waste from the beryl ore processing by Brush Beryllium that may have contained varying amounts of uranium. A correlation appears to exist between samples taken by ORNL with high beryllium concentration and samples with high uranium concentration, although further sampling and analysis would be required to confirm this.

Other general results are as follows:

1. The AEC did maintain a contract with Brush Beryllium to construct and operate a beryllium plant from 1949 to 1958. In addition, Brush Beryllium received a contract to decommission the facility in 1959.

2. No contract information was identified to indicate that AEC maintained a contract with Diamond Magnesium to manufacture magnesium at the Luckey, Ohio, site. However, additional archived records need to be reviewed to determine the connection between the AEC and Diamond Magnesium.

3. From the stereo area photographs, it appears that the landfill located in the northeast section of the site became operational in 1957. If this can be proven, then the amount of contamination in this landfill resulting from AEC sponsored operations at the site may be minimized. Also, the 1957 photographs identify a small landfill north of the sewage disposal plant which was apparently operational. This landfill has not been identified on the site descriptions provided by Motor Wheel Corporation.

4. It is likely that the waste material from the beryl ore contained some concentration of uranium, with the concentration of uranium dependent upon the area from which the beryl ore was obtained. Beryl ore obtained from Brazil and South Africa has been documented to contain uranium in the gangue. In addition, Brush Wellman is currently recovering uranium oxide as a byproduct of

CONCENTRATIONS OF BERYLLIUM AND URANIUM IN SOIL SAMPLES AT THE LUCKEY, OH, SITE

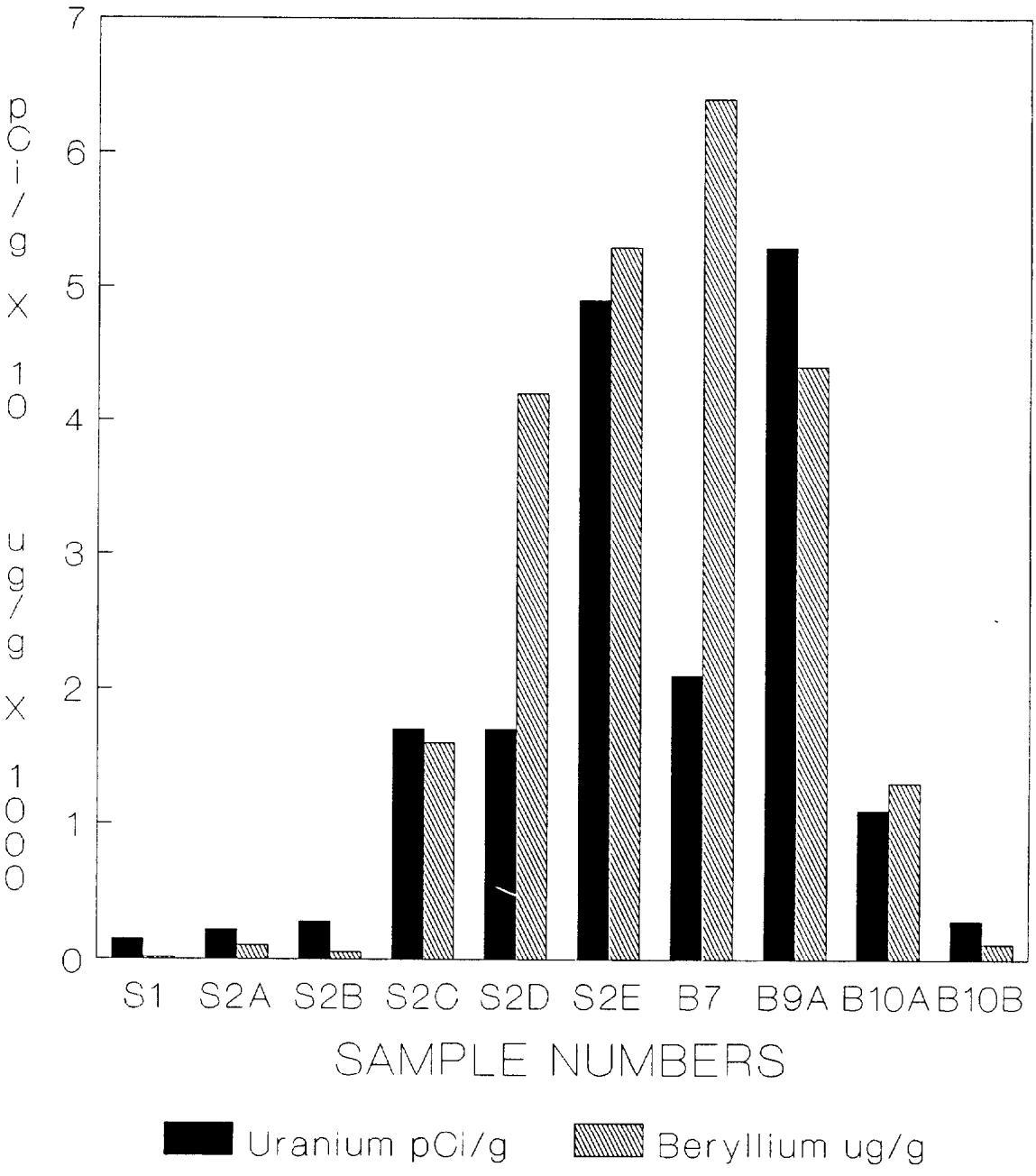

FIGURE 2.

its beryllium extraction process at its plant in Delta, Utah. Although records indicate that the Brush Beryllium, Cleveland, Ohio, office purchased beryl ore from Columbia, it is possible that the waste from this ore contained uranium and that this and other waste from ore processed at the Luckey, Ohio, site contained uranium.

ADDITIONAL AREAS OF RESEARCH

Additional areas of research to determine conclusively whether the residual contamination found at the Luckey, OH, site is solely from the contaminated scrap or resulted from processing beryl ore are listed below:

- Survey similar beryllium processing facilities that used beryl ore from South America, and are known not to have handled any uranium contaminated scrap.

- Perform a chemical analysis on the contaminated soil and sludge from the lagoons to determine the uranium compound present. If the uranium is naturally occurring, then U_3O_8 would be found, but if the uranium is from yellow salt, $UO_2(NO_3)_2$ or $Na_2(UO_3)_2$ would be found.

- Perform a more rigorous statistical test of the hypothesis that beryllium and uranium are found together in the soil samples taken at the Lucky, OH, site.

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