FINAL RESULTS FROM THE IN SITU VITRIFICATION TREATMENT AT MARALINGA

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ABSTRACT

Geosafe Australia Pty. Ltd. (Geosafe) and AMEC Engineering Pty. Ltd. (AMEC) are working together in Australia to use the GeoMelt vitrification process to treat hazardous wastes and remediate contaminated sites.

The GeoMelt process represents a group of vitrification technologies that can be arranged in various ways to meet a wide range of site remediation and waste treatment requirements. All of the GeoMelt technologies involve the electric melting of contaminated soils and wastes to result in the destruction, removal or permanent immobilisation of contaminants. The original GeoMelt technology is GeoMelt In Situ Vitrification (ISV) wherein contaminated soils and wastes are treated in place, in the ground. This base technology has been developed into other configurations that allow a wider range of treatment applications. Adaptations of the process include melting wastes above ground in refractory lined vessels.

Geosafe and AMEC recently completed a multi-year program involving the GeoMelt-ISV technology for the Australian Commonwealth Government at the Maralinga Site in South Australia. The Maralinga Site became contaminated as a result of above-ground nuclear weapons tests and safety trials conducted by the UK in the late 1950's and early 1960's. Geosafe's and AMEC's role in the clean-up project was to use the GeoMelt-ISV process to treat a group of burial pits containing mixed transuranic buried waste at the Taranaki area. A total of 11 pits were treated with the process. A lack of pit characterisation data and inaccurate historical records resulted in a number of challenges to the project. The total estimated volume of the 11 pits treated was on the order of 800% larger than that specified in the historical records. The GeoMelt-ISV process was successful in achieving its main role of converting the loose, friable, radioactive contamination in the pits into dense, hard, intrusion-resistant vitrified masses to eliminate the long-term hazards associated with subsidence or human intrusion. This paper summarises the final results from the project.

INTRODUCTION

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Geosafe and AMEC recently completed a multi-year remediation program involving the treatment of mixed transuranic buried waste pits at the Maralinga site in South Australia. This paper summarises the final results from the program.

GEOMELT PROCESS DESCRIPTION

The GeoMelt process represents a group of vitrification technologies that can be arranged in various ways to meet a wide range of treatment requirements. The original GeoMelt technology is GeoMelt In Situ Vitrification (ISV) wherein contaminated soils and wastes are treated in place in the ground. The base ISV technology has been developed into other configurations that allow a wider range of treatment applications. Some configurations involve the staging and/or above ground treatment of wastes contained in refractory-lined treatment vessels.

All of the GeoMelt technologies involve the electric melting of contaminated soils and debris to result in the destruction, removal or permanent immobilisation of contaminants. The melting process is initiated at the surface of a waste or soil mixture. Electrical power is directed to the treatment zone via graphite electrodes. Electrical power is regulated to maintain the desired melt rate. The melt temperature normally ranges from 1200-2000 degrees C depending on the materials being treated and the particular process being used. The melt gradually grows downward and outward until power is shut off. Melt rates for full-scale treatment plants can exceed 100 tonnes per day depending on the configuration of the equipment and the material being treated. The melt sizes depend on the particular configuration being used. Individual melts can exceed 1000 tonnes in size.

Organic contaminants such as dioxins, pesticides and polychlorinated biphenys are destroyed via pyrolysis and dechlorination reactions at elevated temperatures in reducing conditions in the hot soil adjacent to the melt. No organic contaminants remain in the melt due to the inability of organics to exist at the temperatures involved. A broad range of organic contaminant types has been successfully treated in commercial operations.

Heavy metals and radionuclides are predominantly retained in the melt and are permanently immobilised in the resulting vitrified product. The vitrified product typically consists of a mixture of silicate glass and silicate minerals. The product is typically ten times stronger than concrete and is extremely leach resistant.

Off-gases that evolve from the melt are collected in a steel containment hood and directed to an off-gas treatment system. The effectiveness of the melt in treating contaminants either through destruction for organic compounds or by retention for heavy metals and radionuclides minimises the contaminant loading to the off-gas treatment system.

The GeoMelt process is distinguished by its ability to treat combinations of waste types and debris. The process can accommodate mixtures of contaminant types including organic and inorganic. The typical types of debris treated in prior commercial operations include concrete, asphalt, steel, wood and plastic.

MARALINGA PROJECT BACKGROUND

Maralinga Site

The Maralinga site is located in South Australia, approximately 1000 km northwest of Adelaide (the nearest large city). Smaller towns are located a few hundred km to the south along the southern coast of Australia. The site is semi-arid and is located in the Great Victoria Desert. Summer temperatures occasionally exceed 50 degrees C. The site consists of approximately 3,200 km^2.

Weapons Development Testing Background

The UK conducted a program of nuclear weapons development trials at Maralinga between 1955 and 1963, including seven atomic explosions. The smallest atomic explosion was about one kiloton in yield and the largest was about 27 kilotons. There were also several hundred minor trials conducted at Maralinga. These minor trials were essentially development experiments designed to investigate the performance of various components of nuclear devices, separately and in combination, and almost all trials involved radioactive materials with conventional high explosives and dispersed radioactivity to the local environment [1]. The minor trials were responsible for the majority of the contamination at the Maralinga site.

At Taranaki, Maralinga's most heavily contaminated site, the Vixen B series of minor trials was conducted between 1960 and 1963. These trials resulted in the explosive dispersal of approximately 22 kg of plutonium and similar amounts of uranium-235 and beryllium. In these trials, jets of molten plutonium were projected up to 1000 m into the atmosphere. The trials resulted in the contamination of large tracts of land. The trials were similar to those conducted in 1963 as a joint project between the UK and the USA in Operation Roller Coaster at the Nevada Test Site [1].

Contaminated wastes generated from the Vixen B trials were buried in a series of pits. The contaminated wastes included heavy steel debris, lead bricks, bartye bricks, concrete pads, electrical cable, instrumentation and contaminated soil. Each of the pits contained heavy structural steel including up to 10 large steel plates, each 50 mm thick, and structural beams associated with the test device. The total of the pits were expected to contain on the order of 2 kg of plutonium. The pits were subsequently covered with reinforced concrete caps. Figure 1 shows a photograph of one of the open pits in the early 1960's.

Radionuclide Composition

The plutonium contamination at the Taranaki site contains the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu. The minor trials involved negligible fission yield so the isotopic composition of the source material was unaffected by the explosion. Radioactive decay has resulted in the conversion of significant amounts of the ^{241}Pu to ^{241}Am [1]. Uranium-235 was also used at the Taranaki site during the minor trials for "calibration rounds".

Figure 1. Photograph of an open pit in the early 1960's.

Traditional Owners

The traditional owners of the Maralinga lands are the Maralinga Tjarutja aboriginal people. Between 60 and 200 of the Maralinga people have established a semi-traditional lifestyle at Oak Valley, which is approximately 100 km northwest of the Maralinga site. One of the principal reasons for the Maralinga clean up is so that the Maralinga Tjarutja can regain access to the area.

Primary Hazards

The inhalation of plutonium contaminated dust was determined to represent the most significant potential health hazard arising from the contamination at the Maralinga site. The aboriginals living a semi-traditional lifestyle were considered to be the critical group for radiation protection purposes [1, 2]. The burial pits at the Taranaki area were considered a radiological exposure threat over the long term due to the potential for human or other animal intrusion or through the failure of the concrete caps and subsidence exposing the contaminated pit material.

REHABILITATION PROGRAM

A technical assessment group was established by the Australian Government to evaluate a range of options for the rehabilitation of the site [2]. Through scientific studies and a program of consultation with the State of South Australia and the Maralinga Tjarutja, a preferred program of rehabilitation was agreed. This program primarily involved the collection and on-site burial of

contaminated surface soil from the areas most affected, and the GeoMelt-ISV treatment of burial pits in the Taranaki area.

To provide technical advice concerning the program definition and its implementation, the Commonwealth established a committee of technical experts. The committee, the Maralinga Technical Advisory Group (MARTAC), provided advice to the Commonwealth concerning the implementation of the program from its inception to its completion. In evaluating the GeoMelt-ISV process for its potential use on site, the MARTAC determined that the main role of the treatment should be to convert the loose, friable contamination in the pits into dense, hard, intrusion resistant masses to effectively eliminate the potential for intrusion or subsidence over the long term. Issues such as the leach resistance of the vitrified product were considered secondary.

At the commencement of the program, the burial pits at Taranaki were covered with soil and what was known about the pits was largely based on information contained in historical records. Some trenches and bore holes were installed in the area to investigate the soil types present and non-intrusive characterisation activities of the pits were conducted in an effort to learn more about the pits. However, the results from the non-intrusive characterisation activities were considered to be inconclusive and further pit characterisation efforts were not conducted. The Commonwealth determined that the risks and complexities associated with the intrusive characterisation of the pits or the exhumation of the pits was not warranted.

In 1996, following a period of smaller-scale trials and related studies, the Commonwealth selected the GeoMelt-ISV process to treat the Taranaki pits. The primary project planning basis used was the historical records. The conversion of the loose, friable contamination in the pits into dense, hard, intrusion resistant masses was considered the most important role for the treatment of the pits by the GeoMelt-ISV process.

ISV PROJECT

Four Phase Project

The GeoMelt-ISV project to treat the Taranaki pits involved four phases.

- The first two phases were conducted from 1993-1995 and involved site assessments and small-scale testing to evaluate the performance of the GeoMelt-ISV process on soils and wastes from the Taranaki site. The testing included on-site tests with uranium oxide and actual plutonium-contaminated debris that originated from the minor trials. The results of these phases were used to develop the overall technical approach to the project.

- Phase 3 involved the design and construction of the full-scale GeoMelt-ISV treatment plant. The plant was designed and constructed in Adelaide by AMEC in 1996 and 1997. This phase also included three operational acceptance tests of the full-scale treatment plant and site establishment work such as installing the diesel fuel tank farm and delivering and assembling the treatment plant at the site.

- Phase 4 involved the treatment of the pits. The treatment involved pit preparation, vitrification operations and sampling and analysis. Pit preparations commenced in November 1997.

Vitrification operations commenced in May 1998. The treatment plant was operated by AMEC under the guidance of Geosafe.

PIT PREPARATIONS

The pit preparations included locating the pits, removing the concrete caps, investigating the pit area to determine the pit boundaries, installing trenches around the pits to position instrumentation to monitor melt progress and probing of the pits.

The concrete caps were either removed in one piece or, for the larger caps, cut into sections and removed with a crane. Then the surface dimensions of the pits were estimated through physical investigations. This process involved intrusive investigations and radiological surveys by removing the soil covering each pit to locate the edges of the pits. Essentially, the layer of soil covering the pits was scraped away until the edges of the pits were located.

Pit probing involved the use of a heavy steel rod affixed to a hydraulic rock hammer mounted on a tracked excavator. The rod was driven vertically down into each pit over a grid pattern in an effort to collapse and fill voids. This technique was also used to try to locate the lateral boundaries of the pits and later in an attempt to determine the pit depths. A photograph of pit probing activities is provided as Figure 2.

Figure 2. Health Physics check of probe surface during probing of Taranaki Pit 3. Due to the nature of the contamination in the pits and the presence of clean cover soil above the pits, the retracted probe did not normally have significant surface contamination.

Finally, a mound of sand was constructed over the top of each pit and the equipment positioned for the treatment. The mound of sand served two purposes. First, it provided a level and

uncontaminated base upon which workers prepared the system for processing. Second, the sand above the pit melted and was incorporated into the melt. The silica sand augmented the chemistry of the melt, providing more glass-forming ions, which improved the chemical and physical characteristics of the vitrified product. This was required since the soil at the Taranaki site consists predominantly of limestone, which does not make a good vitrified product without the addition of glass-forming materials. The calcium from the limestone also effectively reduces the melt temperature.

PIT VARIABILITY

A major challenge for the project was the lack of characterisation data for the pits. As noted previously, the Commonwealth decided at the commencement of the program to rely on historical records rather than characterising the pits. The historical records generated by the UK concerning the pit sizes, locations and contents were used as the planning basis for the project but the records were found to contain significant inaccuracies. The total estimated volume of the 11 pits treated was on the order of 800% more than expected based on the historical records. Some pits were determined to be up to 25 times larger than reported in the records. Additional pits not listed in the records were found and some pits were found in different locations than specified in the records. The concrete caps that were put down to cover the pits were typically undersized and some caps were not covering any part of their corresponding pits.

A particular challenge was the lack of data concerning pit depth. Because the obvious inaccuracies regarding the pit sizes, pit locations and the positioning and size of the concrete caps, the accuracy of the historical data concerning the pit depths was brought into question. Thus, to account for the uncertainty regarding pit depths and to account for any variability in the depth of each melt, a strategy of melting deeper than the suspected pit depths was adopted to provide a reasonable degree of assurance that the pits were being fully treated.

Originally, 21 pits were going to be treated by GeoMelt-ISV. However, because the pits were found to be significantly larger than expected and because of the variability in the shapes of some of the pits, a different approach was adopted by the Commonwealth. It was decided that the GeoMelt-ISV process would be used to treat a group of pits referred to as the inner pits. The inner pits were more clearly defined and were expected to contain more plutonium than the other, outer pits. The outer pits were more variable in size and shape and were expected to contain less plutonium. It was determined that the outer pits would be excavated and the contents buried on site in a deep trench with five metres of clean cover.

TREATMENT RESULTS

A total of 11 pits were treated with the GeoMelt-ISV process. The resulting vitrified monoliths associated with each pit were all intrusively sampled and examined to characterise the vitrified product and to confirm the completeness of treatment. To confirm whether or not the process had treated the whole of each pit the vitrified monoliths were all exhumed. A photograph of one of the exhumed monoliths is shown in Figure 3.

Determination of Plutonium Inventory

A known quantity of rare earth tracer, either lanthanum or cerium oxide, was added to the cover sand above each pit prior to treatment. Once the pit was treated and samples of the vitrified product collected and analysed, the distribution and concentration of the rare earth tracer was evaluated.

Figure 3. 500 tonne vitrified monolith resulting from the treatment of Taranaki Pit 19A.

The concentration of the rare earth tracer in the vitrified product was used to estimate the mass of each vitrified monolith. The masses of the monoliths were also estimated based on an energy to mass relationship or, alternatively, based on physical dimensions of the vitrified monoliths combined with density values for the vitrified product. Based on the activity of plutonium in the resulting samples and knowing the approximate mass of each monolith, the original inventory of plutonium in each pit could be estimated. The mass of each vitrified monolith ranged from about 300 to 600 tonnes. Using this methodology, each pit was determined to have contained on the order of 80-175 GBq of plutonium contamination.

Plutonium Distribution and Behaviour in the Melts

The vitrified monoliths resulting from the treatment were extensively sampled. The product composition was found to be variable within the first 500 mm in from the top, bottom and sides but the main core of the monoliths were usually relatively uniform in composition. The plutonium was found to be distributed fairly uniformly throughout the core of the monoliths. This distribution is due to the convective mixing that occurs in joule heated melts. Such mixing within ISV melts has been established by a number of numerical and experimental studies including experimental studies that evaluated the compositional uniformity resulting from starting conditions involving point sources of plutonium and uranium [3]. Plutonium was typically found at lower concentrations at the boundaries of the vitrified monoliths where surrounding, uncontaminated soils were being

incorporated into the melts at the time each melt was terminated. There was no indication of any partitioning or elevated concentrations of plutonium in any of the monoliths. In addition, there was no indication that plutonium was present in the melted steel phase, when such a phase was present at the base of the vitrified monoliths. This result is consistent with results from earlier trials of the process at the site [3].

Retention of Plutonium in the Melt

The retention of radionuclides and certain heavy metals in a melt was determined from characterisation studies by estimating the amount of plutonium in a completed melt as described above and by determining the amount removed from the off-gas stream by the off-gas treatment system. The retention of plutonium in the melt was determined to be in excess of 99.99%. The excellent retention of plutonium in the melts minimised the degree of contamination of the equipment and the radiological hazard to workers. The hoods and off-gas piping were not required to be decontaminated after any one melt due to the level of contamination.

Treatment of Contaminated Steel Debris

The heavy steel debris in the pits sometimes fully melted and sometimes did not depending on the melt temperatures achieved during the treatment of each pit. As noted previously, the calcium from the limestone influences the overall composition of the melt and can lower the melt temperature. The silica sand was added via the mounds above each pit to counteract the influence of the limestone and to enhance the physical and chemical properties of the vitrified product.

For those cases where the steel fully melted and pooled at the base of the melt, there was an apparent absence of plutonium in the melted steel ingot. For those cases where the steel did not melt, there was also an apparent absence of plutonium contamination on the surfaces of the steel exposed to the melt. Samples of unmelted steel in contact with vitrified product (the melt) were collected and studied. Electron photomicrograph analyses revealed that the corrosiveness of the melt was sufficient to promote active dissolution of the steel surfaces, which resulted in transferring plutonium from the steel into the melt (refer Figure 4). Iron rich minerals and particles of essentially pure iron were observed being incorporated into the vitrified product at the transition zone between the steel and the melt. Thus, the contaminated steel debris in contact with the melt was decontaminated by the GeoMelt-ISV process either by the complete melting of the steel or through the dissolution of the steel surfaces exposed to the corrosive melt.

Vitrified Product Characteristics

The resulting product was a mixture of glass and crystalline materials as predicted. The primary crystalline phases were wollastonite and diopside, both calcium silicate minerals. Previous studies have shown that the crystallisation process removes calcium from the glass phase of the melt into the crystal structure. This effectively increases the localised silica content in the glass phase [4]. The plutonium remains in the glass phase, which is effectively enriched in silica (70-80 wt%). The higher silica content in the glass phase results in greater durability and leach resistance.

A variety of leach tests of the vitrified product resulting from earlier phases of the project were conducted including long term (>3 years) leach tests that are still under way. The leach tests include various durability tests including the Product Consistency Test [5]. Leach data from these tests

indicates that the normalised leach rates for the vitrified products are extremely low (<0.1 g/m^2day) for all oxide species and in most cases the leach rates approach 0.01 g/m^2 day [3, 6].

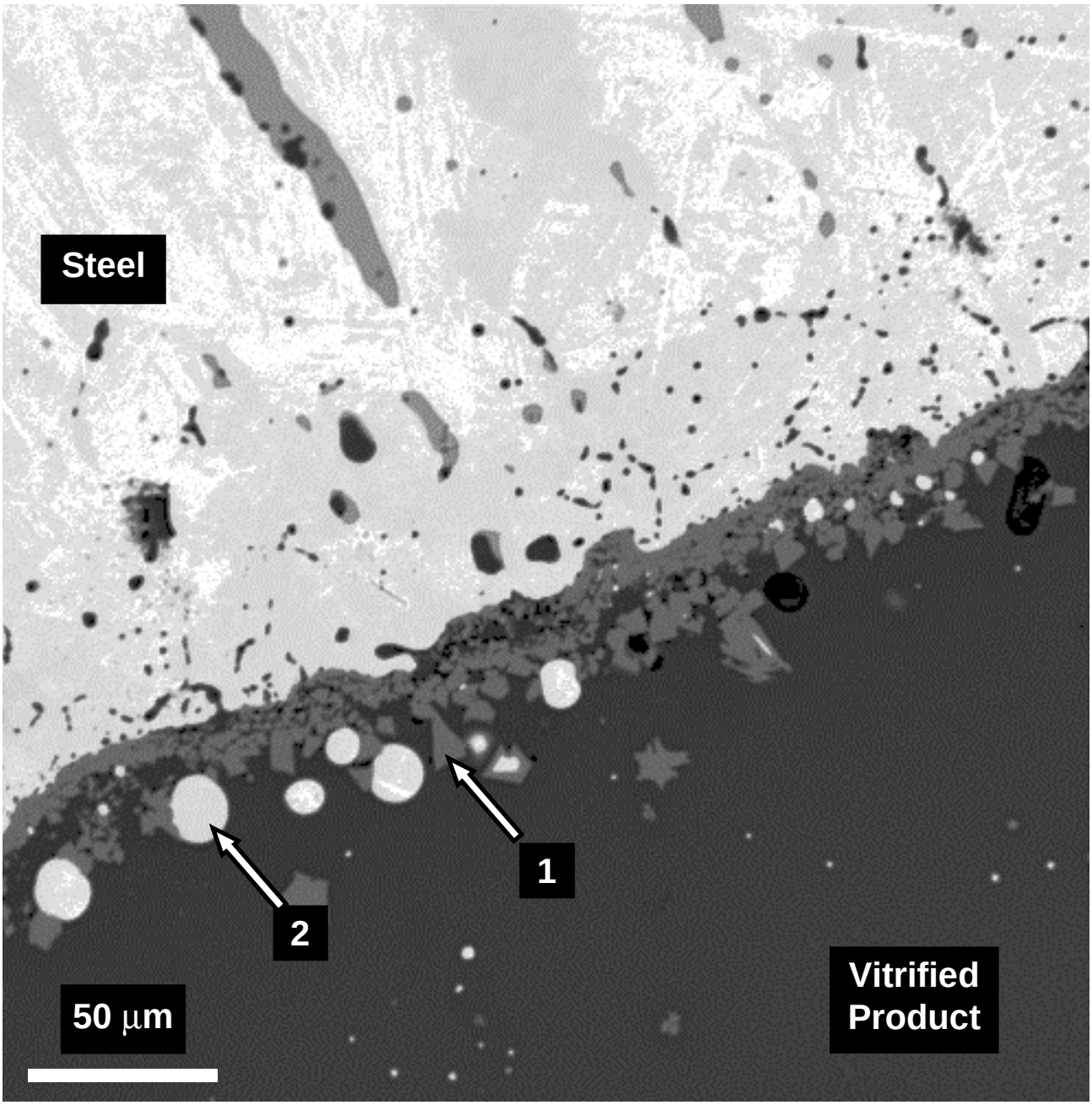

Figure 4. This image is a high-magnification electron photomicrograph of the interface between a piece of stainless steel and vitrified product. The image shows the dissolved surface of the steel. The crystals (point 1) next to the heavily corroded steel interface are Mg-Al-chromite and contain nearly 60% chromium in the trivalent state. Iron droplets (point 2) are almost pure iron. The dissolution process shown in the image results in the removal of radioactive contamination from the surfaces of the steel to the melt where it is immobilised in the vitrified product.

More recently, samples of vitrified product from two of the full-scale melts at Taranaki were subjected to PCT testing. The samples were selected from monoliths that represented the two extremes of composition. The normalised release rates for the major oxides from the 28-day tests were substantially less than 1 g/m^2 day with most release rates less than 0.1 g/m^2 day. The results demonstrate that the vitrified products produced at Taranaki have outstanding leach resistance properties.

Challenges

The treatment of the pits was challenging because of the unknown and heterogeneous nature of the pit contents. The pits were ultimately determined to have contained sealed drums and reactive chemical compounds such as fuels, and probably explosive materials, in spite of the fact that the historical records did not indicate the presence of such materials. The heterogenous materials did cause small processing transients from time to time but the equipment was designed to accommodate such transients. However, once significant transient occurred during the treatment of one of the pits. The transient did not result in any injuries and there was no detectable release of airborne contamination but the event did result in equipment damage. An investigation team consisting of representatives of several organisations was assembled to determine the cause of the transient. Chemical and physical evidence was discovered during the investigation process including the finding of traces of reactive chemicals below the melt and in the off-gas treatment system. The investigation resulted in the finding that the transient event was caused by a sub-surface explosion at the base of the melt, which was most likely caused by explosive materials in the pit. Because Geosafe could not be assured that the remaining pits did not contain similar materials, it recommended to the Commonwealth that it should not proceed with further treatment at the site unless the pits could be exhumed, unacceptable materials removed, and the materials restaged for treatment. Thus, the Commonwealth decided to excavate and bury the contents of the remaining pits.

Completeness of Treatment

Each of the vitrified monoliths was extensively evaluated to characterise the product and to confirm the completeness of treatment. For this evaluation process, trenches were excavated around the vitrified monoliths to increase the rate of cooling.

In all cases, the melt proceeded out laterally far enough to incorporate all of the pit material. In no case was there any pit material found around the perimeter of the monoliths that was not fully incorporated into the melts.

As noted previously, the depths of the pits were not known. Consequently an approach of melting deeper than the expected pit bottom was adopted to provide the best chance of ensuring that the whole of each pit was treated. The project regulator had determined that leaving up to 10 g of plutonium oxide within untreated material beneath a vitrified monolith would be acceptable since the main goal for the GeoMelt-ISV process was to minimise the potential for intrusion and subsidence. A vitrified monolith with a mass of several hundred tonnes was considered adequate in that regard. All of the monoliths were destructively evaluated to determine the completeness of the treatment. The process treatment depths achieved were adequate for all but two pits. In those two pits, and even though the final melt depths were well below the historical pit depths, trace amounts of unincorporated plutonium were found at the base of the two vitrified monoliths. The amounts of

plutonium found in these two cases were estimated to be milligram quantities in the first case and microgram quantities in the second case. Had the treatment process been continued and deeper melting achieved, the contamination would have been incorporated into the melt. However, since the depths of the pits were not known, it left the possibility that the treatment depth may not be sufficient.

CONCLUSIONS

In spite of the lack of characterisation data for the pits and the inaccurate historical records, the GeoMelt-ISV process was successful in achieving its main role; to convert the loose, friable contamination in the pits into dense, hard, intrusion resistant masses to effectively eliminate the potential for intrusion or subsidence over the long term. The vitrified product associated with each pit represented a dense, hard, intrusion resistant mass. The geochemistry of the product was consistent with expectations.

The vitrified product compositions were found to be variable within the first 500 mm in from the top, bottom and sides but the main cores of each monolith were usually relatively uniform in composition. The plutonium was found to be distributed fairly uniformly throughout the central cores of the vitrified monoliths. There was no indication of any partitioning of plutonium in any of the monoliths. Plutonium was typically found at lower concentrations at the boundaries of the vitrified monoliths where surrounding uncontaminated soils were being incorporated into the melts at the time each melt was terminated.

Examinations of steel debris samples in contact with vitrified product established that the corrosiveness of the melt was sufficient to dissolve the steel surfaces in those cases where the steel did not appear to melt. This resulted in the transfer of the plutonium contamination from the exposed surfaces of the steel to the melt. There was no indication that plutonium was present in the melted steel phase when such a phase was present at the base of the vitrified monoliths.

The lateral extent of treatment was adequate in all cases. In no case was there debris or other contaminated pit material found untreated around the lateral perimeter of the monoliths. In spite of the lack of accurate pit depth information, the depth of treatment was adequate in all but two cases and in those two cases, only milligram and microgram quantities respectively of plutonium were found at the bases of the two melts.

Although leach resistance of the vitrified product was not of primary importance in this application, the vitrified product exhibited outstanding leach resistance based on PCT testing. The normalised leach rates for the vitrified products are extremely low with most oxide species having release rates of less than 0.1 g/m^2 day.

The retention of the plutonium in the melts was determined to exceed 99.99%. This retention minimised the amount that was evolved to the off-gas containment hood and off-gas treatment system minimising the potential for exposure to personnel and the need for equipment decontamination.

There were no injuries to operating personnel during the project and no measured up take of contamination as a result of the operation.

The lack of characterisation data for the buried waste pits combined with inaccurate historical records resulted in the need to make several adjustments to the project approach. Operational adjustments were required for the process including making much larger melts than expected. However, the primary result is that the GeoMelt-ISV process was able to convert the loose, friable contamination in the pits into dense, hard, intrusion resistant monoliths thus satisfying the main goal that was established at the start of the project.

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