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Energimyndighetens titel på projektet – engelska Identification of quality tolerances for the recycling of glass and lead from glass land fills	
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Foreword

In this project we have investigated the quantities of foreign materials that can be tolerated in a lead/glass smelt separation process developed in another ongoing project. We have also studied the excavation of glass landfills as well as sorting and cleaning methods. It is within the strategic innovation programme Re:Source which is financed by Energimyndigheten, Vinnova and Formas. The project is a cooperation of RISE Research Institutes of Sweden AB, Linnaeus University and Ragn-Sells.

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Contents

Sammanfattning

I detta projekt har det undersökts hur mycket främmande material som kan tillåtas i en smältseparationsteknik för att skilja på bly och glas, som har utvecklats i ett annat pågående projekt. För att ta reda på detta har en inventering gjorts av vilka och hur mycket material som finns på glasdeponier och det har även undersökts vilka sorterings- och reningsmetoder som kan användas. Syftet med denna förstudie var att komplettera och koppla samman de pågående projekten kring en hållbar sanering av glasdeponier och återvinning av glas och metall för införande i den cirkulära ekonomin.
En geofysikalisk kartläggning är gjord för Alsterfors glasdeponi, för att lättare kunna avgöra vilka områden som är glasrika och lättare kunna avgöra hur djupt ner glaset ligger. De glasrika områdena är därefter sorterade i olika storlekar och vilken typ av material, för att kunna veta hur mycket material från byggnader, deglar samt jord och sten det följer med glaset.

Resultat visar att det deponerade glaset på glasdeponierna Madesjö och Alsterfors är tillräckligt rent för att separationen av bly och glas ska fungera i smältprocessen. När det gäller det oorganiska materialet, så bör man ta bort så mycket som möjligt innan, men en mindre mängd relativt små bitar av porslin, degel- och ugnsmaterial kan tolereras i smältprocessen.

Den här studien visar att det är fullt möjligt att faktiskt använda det deponerade glaset trots att det följer med del oönskat material i utgrävningsprocessen.

Summary

The current project investigates the quantities of foreign materials that can be tolerated in the melt separation process, to separate lead from glass, developed in another on-going project. The planned project relays on identification, characterization and excavation of glass landfills as well as sorting and cleaning methods. The purpose of this prestudy was to complement and combine the on-going projects that aims for a sustainable remediation of glass landfills and the recycling of glass as well as metals for introduction into the circular economy.

A geophysical mapping of Alsterfors glass landfill was done, to make it easier to decide where the glass rich areas are and also to know the depth of the glass. The glass rich areas were sorted, both in size and what kind of material, to know how much material from buildings, pots but also how much soil and stones following the glass.

Results show that the dumped glass on the glass landfills in Madesjö and Alsterfors is clean enough from soil and organics for the separation of lead and glass to work properly in the melt process. When it comes to the inorganic material, large pieces should be removed, but small once could be tolerated in the melt process.

This study show that it is possible to really use the dumped glass even if some unwanted material and soil will follow the glass in the excavation process.

Introduction/Background

The Kingdom of Crystal once flourished and the number of glassworks was large, but today there are few. Glass products have previously contained toxic substances like lead and arsenic, today they have been completely removed from the glass recipe, but large amounts of shards remain in old glass areas. Only in the Kingdom of Crystal there are about 100 glassy areas, as well as municipal landfills, of which about 40 were classified as a major risk at the County Administrative Board (Länsstyrelsen) in Kronoberg County's survey. The Swedish Environmental Protection Agency's (Naturvårdsverket), inventory at 22 glass mills in Kalmar and Kronoberg County showed that the total amount of deposited glass is approximately 130,000 m3, which includes arsenic and lead. Heavy metals leached from the glass waste are likely to enrich in fungi, plants and animals. Both lead and arsenic, in addition to being acute toxic by ingestion or inhalation, are highly toxic to aquatic organisms. People exposed to small (not acute toxic) amounts of lead and / or arsenic increase the risk of infertility and cancer, and this probably also applies to many animals. In summary, there is a great need to reduce the risk of exposure to glass waste in Sweden.

By recycling the large amount of glass waste, the leaching of toxic substances disappears to soil, water and air in the area. The process makes waste wasteful resources in a circular economy, and it is impossible to cover or deposit large amounts of hazardous waste. According to the 10th chapter of the Environmental Code, the person conducting or carrying out activities that cause environmental damage is responsible for restoring the environment and preventing human and environmental damage. If there is no activity, responsibility falls to the owner of the contaminated soil. This has become a major problem when companies do not dare to establish themselves in the region.

Glass containing lead has been used mainly for tableware and artifacts. Arsenic, on the other hand, was used in many more glass types (such as tableware, art and packaging glass but also various specialty glasses). Much of the old deposited glass waste has high content of lead and significant arsenic levels. Glafo (now RISE – glass) has developed lead and arsenic-free glass recipes for the various applications of the Nordic glass industry since the 1970s, and today it is the standard for the Swedish glass works. Some parts of Europe are still melting a lot of lead-holding glass, and similar problems are found in many countries, in other words; in the future there is potential for exporting the technology.

Figure 1. Madesjö glass landfill. Photo: Elisabeth Flygt RISE Glass.

Figure 2. Pictures from Målerås glass landfill. Photo: Elisabeth Flygt RISE Glass.

The cleaning costs for each glass mill have been estimated at approximately SEK 10-50 million per item if carried out using traditional landfill technology. The options used today to take care of old glass wastes are to transport the excavated materials to another landfill or to cover the waste in place so the leaching of heavy metals decreases. The project "Glass deposits - from waste to resource" have shown that on a small scale it is possible to separate metal and glass so that the remaining glass contains less than 0.1% by weight of lead. Recycling of metals and glass also contributes to the reduction of new resources. Handling the glass waste is necessary for the built environment around glass dumps to be good and facilitates the preservation of the cultural and industrial history buildings in the glass industry. Recontamination is required for the re-establishment of activities in disused glass works buildings and -environments.

As a part of gradually scaling up and commercializing the technology of the separating of metal and glass; sorting, purification and tolerance requirements become an important issue. Today there are large amounts of material other than cutting on glass slabs because they are used by both the glass works and the village. Therefore, it is possible to find all kinds of pulp, porcelain and brick but of course soil and stone, and possibly a subset of municipal waste and also other industrial waste. The amount of foreign matter entering the metal and glass separation process should be minimized but the question is to what extent. It is a matter of how the glass quality will be after the separation, i.e. dissolution of inorganic materials such as porcelain, stone and crucible material, but also how the melting technique is influenced by organic matter. The melting technique of metal and glass is based on an additive of reducing agent and the metal oxides will be reduced to metals for removal. Adding organic matter will cause oxidation and thus deteriorate metal degradation.

Procedure

The project was divided into four different work packages that investigate the chain from materials on glass landfills, sorting and purification processes as well as different materials impact on the separation process of lead and glass. The first two work packages were led by Linnaeus University (LNU), and the work was carried out primarily by the doctoral student financed by the foundation of Ragnar Sellberg. In these, we looked at sustainable methods of inventing, sorting and cleaning materials from glass dumps. The other two work packages were led by Glafo (RISE glass) and were focused on the melt separation process and foreign materials impact on the glass quality and the reduction conditions in the melt. Within the project, one workshop was carried out focusing on utilizing the knowledge of other organizations and information of the project was reported on in one conference and in one seminar in the end of the project.

WP1 - Inventory of glass deposits

Inventory of glass deposits involved literature search as a first step, in which it was established that the Småland region had about 90 glassworks between the 16th and mid-19th Century, leading to different dumpsites where crystal glass waste especially from the production process was dumped. Over 40 dumpsites have been classified as high-risk sites, 22 of which are reported to have a total of over 420000m3 of contaminated soil and glass, comprising cadmium (Cd) arsenic (As) and lead (Pb) at 30 tones, 420 tones and 3100 tons respectively. 1,000,000m3 of contaminated soil and glass are estimated in 50 sites. Thus, WP1 focused on landfill mapping and materials characterization.

Figure 1 and 2 show how Målerås and Madesjö landfills look like. In Madesjö it is mostly glass in a quite focused area, in Målerås the glass is spread in a large area with a lot of other things dumped in the same area.

Geophysical Mapping

Two different Surface Geophysical methods were employed in the project, and these were Electrical and Electromagnetic methods, performed as follows:

Electrical method: It was achieved by use of the ABEM Terrameter LS instrument, which simultaneously measured Induced Polarization and Electrical Resistivity. It involved spreading out two 40-meter cables on either side of the Terrameter LS instrument and over and across the landfill, after which electrodes were nailed into the ground at 2m intervals. The measurement protocols employed were Gradient and Wenner.

Electromagnetic method: It was achieved by use of the ‘Slingram’ instrument otherwise known as CMD Explorer (Multi-depth Electromagnetic Conductivity Meter). It involved walking with the instrument over and across the landfill.

Prior to geophysical mapping, the site (Alsterfors) had to be geocoded and maps for topographic wetness index obtained to facilitate the mapping.

Test-pit Excavations and Sampling

Points for test pit excavations were set differently depending on whether a site had been initially mapped with geophysics or not. For the mapped ones, test pits were set according to geophysical results obtained and along geophysical mapping lines, as these showed areas of interest for excavations. For sites without geophysical mapping, they were divided as much as possible to ensure representative site coverage. Excavation of test pits was then achieved using a 4.5 tone excavator. After excavation and heaping, the heaped materials would be sampled systematically according to the Nordtest Method NT ENVIR 004-1996/05 under sampling procedures from stockpiles. At Madesjö, 19 samples were taken and analyzed in triplicates to yield a total of 57 samples.

WP2 - Sorting material from glass deposits

Excavated material sorting was done in order to ascertain particle size ranges as per feedback from Glafo (RISE Glass) (WP 3 and 4). It was done on materials from Madesjö and Alsterfors landfills. Excavated material heaps were sampled into 14l buckets, after which they were subjected to sieving using 63mm, 31.5mm, 20mm, 16mm, 11.3mm, 8mm, 4mm and 2mm sieves. The sieving was achieved on laboratory scale, and involved hand sorting of the fractions to fully understand their compositions. The fraction smaller than 4mm (>2mm) was further hand-sorted with the aid of a magnifying glass.

WP3 - Impact of inorganic material on glass quality after melt separation

RISE-Glass (former Glafo) examined the influence of various inorganic materials, such as materials from crucibles (pots), rocks, insulating materials, porcelain and refractive material, on the lead/glass melting separation process. Controlled pieces, about 1 cm^3, of different inorganic materials were put in the glass melt for different amount of time. The time it takes for the different materials to be solved are investigated and also how the separation process are affected by the inorganic contaminations. The lead, antimony, arsenic and cadmium content were measured in the glass phase before and after the melt separation process, a portable XRF Bruker S1 Titan.

WP4 - Impact of organic material on melt separation

RISE Glass investigated this through adding known amount of organic matter (soil, twigs, leaves, sand etc.) to the glass cullet before the melting process. Figure 3 shows when the soil and glass cullet are put into the crucibles in the furnace. As organic matter can affect the reduction ratio in the glass melt, which the glass/lead separation is very dependent on, the quality tolerance in weight % organic matter as soil was determined. The lead, antimony, arsenic and cadmium content were measured in the glass phase before and after the melt separation process. A portable XRF Bruker S1 Titan was used and some of the samples were also measured with an Atomic Absorption Spectrophotometer, Perkin Elmer AAnalyst 400, after acidification. The additions of different materials are modeled in a trial plan (DoE) in the computer program MODDE.

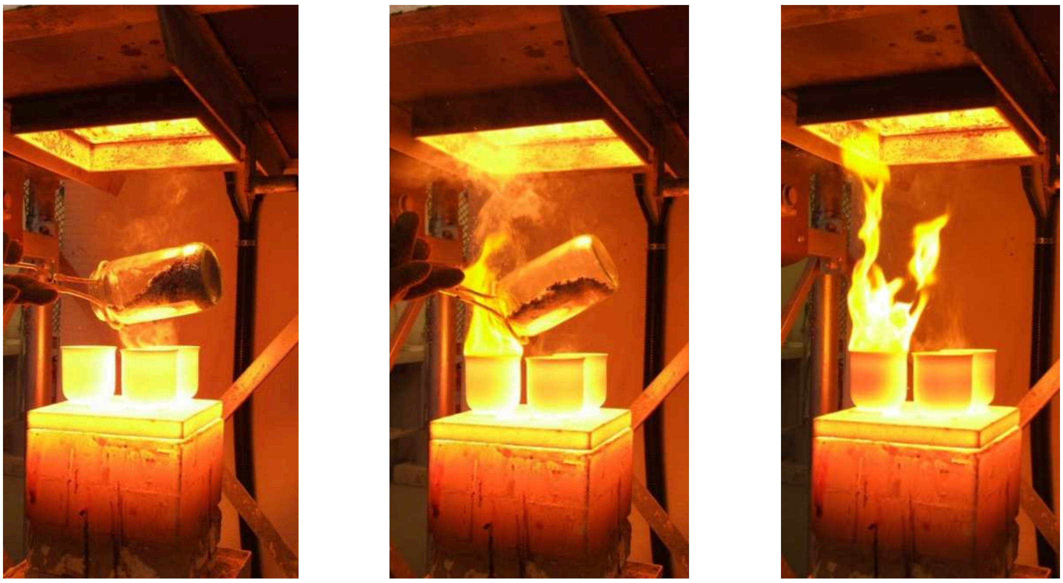

Figure 3. Dried soil blended with the glass shards poured into the crucibles in furnace keeping the 1400 °C.

Results

WP1 - Inventory of glass deposits Geophysical Mapping

Figure 1 shows results of Electrical Resistivity measurements on one gradient.

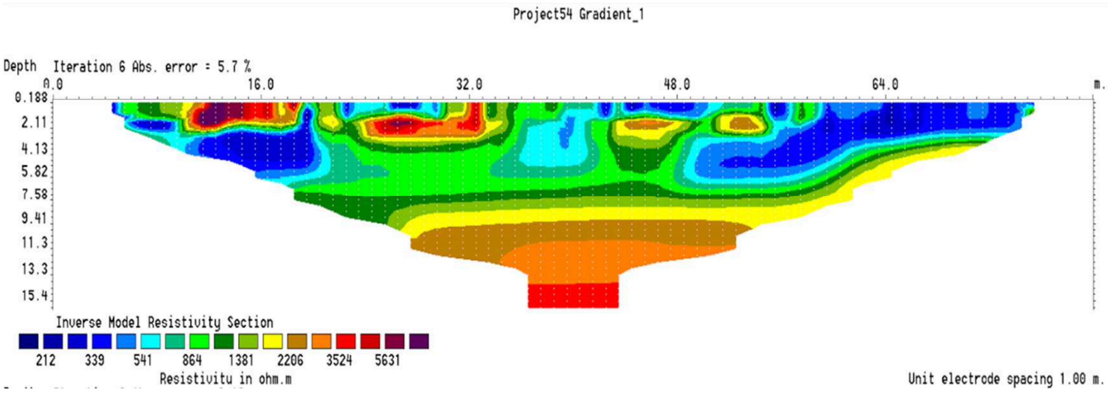

Figure 4: Gradient 1 profile at Alsterfors

Using the image as a guide for setting test pits, test pits were dug at 10m intervals from the 0m position. As shown in the figure, the region between 10m to 20m was filled with glass all the way to 3m depths. The region around 30m (±2m) had glass in the top half-meter depth, after which big stones were found, along with a bigger stone at around 2m depth. Other points (40m, 50m and 60m) excavated contained soil and some traceable organics with the water table within reach. Results indicate that geophysics can be relied on to show areas to concentrate on during excavations, with areas of glass concentrations indicating higher resistivity than those of other materials concentrations (stones inclusive).

WP2 - Sorting material from glass deposits

The results for particle size distribution for both Alsterfors and Madesjö show that bigger particles (>63mm) accounted for between 30 and 40% of the overall mass whereas the fine fraction (<8mm) accounted for about 15-20% of the sampled mass. The compositions of the individual size fractions from the 2 samples and 4 trials at Alsterfors and Madesjö are within the quality tolerances for inorganic and organic materials investigated in WP3 and WP4. The Madesjö glass landfill contains over 95 weight% glass for the fractions. In Alsterfors landfill there are some percentage stones, construction materials and soil, but still good enough to use for the melt separation process.

In both landfills and among all sieve sizes, glass was highly abundant. In the Alsterfors case, stones, soil and organics are notable since the waste was buried in soil, as opposed to the Madesjö case where glass lies in an open dump. The notable fractions from the Madesjö case were construction materials (from furnace walls) and organics.

Basing on the laboratory sorting experiments, different sorting techniques have been suggested for trials on pilot (and eventually large) scale. The techniques will take advantage of particle size, density, magnetism and material composition, and will be as follows:

i. Source Separation: the excavated materials will be source-separated, which will involve manual sorting onsite to remove bigger and easily noticeable materials.

ii. Vibrating/Drum Screening: This is to separate course from fine materials based on particle size.

iii. Magnetic Separation: This is to remove ferrous and metallic objects (based on magnetism).

iv. Air/Wind Screening: This is to further separate glass from other materials based on differences in their densities.

v. XRF-based Sorting: The final step would be to concentrate glass of desired metals based on their metal compositions. This would employ an XRF-based glass-sorting machine produced by Redwave in Austria. While equipment for all other stages is available and accessible, the XRF-based sorting technique is yet to be tried and verified with our materials and melting process requirements.

WP3 - Impact of inorganic material on glass quality after melt separation

The insulating material and stone was dissolved after about 20 minutes .Materials from porcelain and refractives (Alu 60) were not dissolved after 24 hours. The material from crucible was gone after about 18 hours.

The inorganic materials did not affect the lead/glass separation process.

WP4 - Impact of organic material on melt separation

The additions of different materials were modeled in a trial plan (DoE) in the computer program MODDE and they showed that the only component that affected the separation process in a negative way was when quite large amount sand was added to the glass cullet. All other component did not affect the lead/glass separation process in a negative way when the amount of organic materials were in the same level as the contamination as were discovered in WP2.

Figure 7 displays a divided crucible after the lead/glass melt separation process and also the glass phase after crushed with a mortar and pestle.

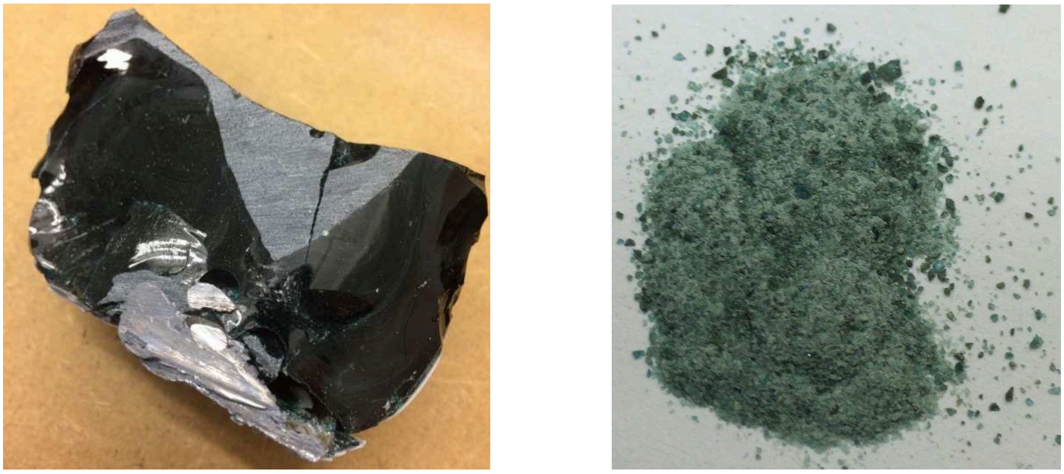

Figure 7. To the left: crucible divided with diamond saw, the glass phase and the lead phase can be seen. To the right: glass phase crushed with a mortar and pestle.

Discussion

The overall results are positive; i.e. the geophysical mapping of Alsterfors shows that the method is very promising to locate the most glass rich areas. In another ongoing project there will be geophysical mappings of more landfills [1], this project will be finished in august 2017. The sorting of the materials with different sieves in lab scale show both the size distribution of the glass pieces and the different materials and soil following the glass pieces. This sorting made it very easy to compare the amount of inorganic and organic material following the glass from the landfills with the controlled amount of soil, sand, peat, stones and materials from furnaces and buildings. As the results show that the levels of contamination of the glass from the landfills is within the quality tolerances determined in WP3 and WP4 this is one step closer to reuse the large amounts of glass waste on landfills in Småland and at the same time get rid of the heavy metals which pollutes the soil and groundwater close to the landfills.

In an ongoing project[2], there have been two workshops about how the separated glass can be used for. As the glass is quite dark, the use might be limited to some kind of building material, but several other proposals are also discussed.

All of the glass in the landfills is not containing lead, and at the moment Ragn-Sells, Lnu and RISE Glass discussing how to separate lead and arsenic containing glass from lead free. If this is possible, the lead free glass can be used directly as a resource. This will save a lot of energy for the glass works, because when making glass from virgin material the energy needed are significantly more than from sherds, about twice amounts of kWh/kg ([3]).

Publicationlist

Conferences:

- Glass mining workshop at Orrefors (6-13 April 2017)
- Linnaeus Ecotech Conference (21-23 November 2016)

Papers and Manuscripts:

- Characterization of waste from glassworks towards resource recovery – the case of Madesjö dumpsite (Conference Paper)
- Potential for Industrial Symbiosis in the region of old crystal glassworks with focus on landfilled waste (manuscript)

References

1. *Glasdeponi Materialkarakterisering i Glasriket*. 2017.
2. *Glasdeponi - Från usch till Resurs*. 2017, RISE Glass
3. Falk, T., et al., *Boken om glas*. 2nd ed, ed. E. Flygt. 2005, Växjö: Glafo AB.

Appendix

- Administrative appendix
- Sekretessbelagda resultat