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Review

Using Vitrification for Sewage Sludge Combustion Ash Disposal

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Abstract

The aim of this paper is to show possibilities for effective processing of hazardous waste using the vitrification method, which is recommended for disposing of ash from the combustion of sewage sludge and developing a process for the conversion of waste to a sustainable and environmentally safe product. The use of vitrification solves the problem of utilizing some hazardous waste by simultaneously reducing or eliminating their harmful effects and decreasing their volume. More and more research has been undertaken to determine specific properties of the resulting products (vitrified waste), in particular regarding the sustainability to ensure safe storage or in specific commercial applications. We describe the methods of processing of ash from combustion of sewage sludge to obtain "vitrified aggregates" as well "glass beads" that are useful substitutes for commonly used products.

Keywords: combustion, sewage sludge, ash, vitrification

Introduction

Combustion of sewage sludge is recommended as the best available technology for processing results in a considerable quantity of ash. Forecasts of the National Program assume that their total in 2015 will increase by approximately 181,000 Mg relative to the quantity produced in 2010 (43,700 Mg) [1]. The resulting ash from combustion belongs to the group of hazardous waste and should be used, as suggested by Białowiec et al. [2], in the environment after having been processed. The properties of the ash obtained mostly depends on the type and quality of incinerated sewage sludge [3, 4].

A popular solution in processing ash is its solidification in cement compositions and sintering to form granules [5]. The resulting aggregates (e.g., Luca and Pollytag) are practical applications, among others, as a filler in wetland sewage treatment systems. Their use increases the efficiency of removal of certain pollutants present in the wastewater [6].

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Another way of processing ash is vitrification – consisting of the formation of an impermeable and durable structure of glass. Active ingredients such as heavy metal compounds are closed tightly inside the structure, which prevents their migration to the environment. Hazardous components are disposed of by binding molecules in the crystal structure of the enamel and by hermetization [7]. Such elements as phosphorus, boron, and silicon are permanently bonded. During heating, they are melted into a liquid phase and, after cooling, form an integral part of the crystalline lattice of glass. Another issue is hermetization of waste components such as cobalt, lead, sodium, magnesium, lithium, and cesium. They represent intrusive additions trapped in the crystal lattice structure. As a result of diffusion melting the crystal lattice of the compounds, hermetization can take place either during the heating of waste or during cooling [8].

Vitrification is typically used to treat wastes of complex chemical composition and unfavorable physical properties, such as medical waste, asbestos, slag, etc., as well as the disposal of radioactive waste. Vitrification of ash is often

Table 1. A brief description of waste for vitrification.

No.	Type of waste	Description	References
1	Fine coal, rock dust	From mining and mineral processing	[30]
2	Contaminated soils	Agriculture and horticulture	[31]
3	Bio-waste	Forestry, fisheries, food preparation, and processing	[22]
4	Wood and paper	Manufacture of furniture, pulp, paper, and paperboard	[22]
5	Tannery sludge	Processing operations of leather and fur	[32]
6	Hydrocarbons	Petroleum refining, natural gas purification, and pyrolysis of coal	[22]
7	Chemicals	Inorganic chemical processes and photographic industry	[33]
8	Paints and varnishes	The production of paint, varnish, adhesives, and sealants	[34]
9	Fine-grained scraps	Mechanical surface treatment of metals	[35]
10	Construction aggregates	Construction and demolition of buildings	[36]
11	Medical waste	Sharps, organs and body parts, sanitary towels, bandages, reagents, drugs	[37]
12	Sewage sludge	Municipal and industrial water treatment	[38]
13	Ash and slag	From power plants and other energy facilities	[39]
14	Sludges and filter cakes	From machining of steel, aluminium, lead, zinc, copper and precious metals, and from electroplating and coating of metals	[40]
15	Glass and ceramics	From glassworks during glass processing and from households and industrial plants	[41]
16	Cement, lime, and plaster	From manufacture of mineral binders and from lime and cement industry	[42]
17	Radioactive waste	From production and processing of nuclear fuel and from the mining of uranium ore	[43]
18	Asbestos waste	From manufacture of textile articles, fibers, roof panels, water, and sewage pipes	[44]

used to neutralize the hazardous components, including heavy metals [9-11].

To obtain vitrified forms of ash one should use a melting temperature of 1150-1450ºC, followed by intense cooling [12]. Vitrification of ash from the incineration of sewage sludge involves the addition of silica (SiO_2), whose share is too small in that waste. Effective vitrification can be achieved by the addition of fly ash from coal combustion, where the share of SiO_2 is twice as high [13]. Another solution may be the addition of shredded glass as powder. Additionally, its presence reduces the temperature of the synthesis reaction, thus reducing the energy requirement [14].

The method of vitrification of ash obtained from different types of boilers helps the formation of products with irregular or rounded shapes [15-17]. Their dimensions differ from a few millimetres to tens of centimetres or more.

Vitrification is often used to process ash from power plants or from incineration, land tannery sludge, and tailings, and waste containing sulphur compounds, lead, and other elements [18-20]. Using waste vitrification of different substances allows products to be reused, mainly in the ceramic and construction industries [21-23].

During heating, apart from chemical compounds, organic compounds contained in the waste are also disposed of. The waste may take the form of vitrified blocks in the shape of the dish in which it cools [24]. This shall also apply to the agglomeration of fine-grained fraction of the waste in closed body, and then heating (sintering) in rotary kilns [25, 26]. After completion of the transition phase it is cooled in water, causing it to form a coating and sealing the glass.

The use of vitrification solves the problem of the development of certain types of hazardous waste by simultaneously reducing or eliminating their harmful properties and reducing their volume [27, 28]. An additional advantage is that some of the products are used in commerce and do not need to be stored [29].

Vitrification of Waste

Polish industry generates different types of waste – including hazardous ones – that can be used for vitrification (Table 1).

Many problems are created by the inactivation of composites found in ash from the incineration of waste and sewage sludge, blast slag, fly ash from coal power stations, medical waste, asbestos, and radioactive waste.

Ash from incinerating municipal waste can contain very high concentrations of heavy metal chlorides and alkali chlorides. Prior to vitrification, waste will require further processing, usually by precipitation from a solution of hydroxides or phosphates. As for the vitrification process of ash from the municipal waste incinerator in Rotterdam, it does not use any additives [45]. This ash contains mainly silica, calcium oxides, carbonates, sulphates, chlorides, Na, K, Al, Fe, Ti, Mg, and Mn. The resultant glassy product has a much higher resistance against leaching toxins than unprocessed ash. A similar process of vitrification without any additives was carried out for the disposal of ash from the incineration of sewage sludge in Lisbon, Portugal [46]. The chemical composition of the ash included silica, calcium oxide, and metal oxides. Heating was carried out in crucibles for two h at 1400ºC, and then cast to form a liquid phase with a water-cooled brass.

Also in the Netherlands, ash from incineration of municipal waste is vitrified without additives [47]. The heating process was carried out at 1500ºC under an atmosphere of nitrogen. The obtained sample of homogeneous glass contained mainly SiO_2 (52.1%), CaO (16.2%), Fe_2O_3 (7.7%), and small amounts of Na_2O, MgO, K_2O, and TiO_2.

More often, however, the vitrification process is carried out – particularly with hazardous waste – after mixing with various additives to facilitate the melting process to form a more homogeneous product [48]. For example, ash from coal mixed with powdered glass, feldspars, and aluminosilicates, and heated at 1450ºC. The presence of aluminium oxide allows for a high chemical resistance of the final product. Ash also requires pre-treatment – grinding to a particle size <0.4 mm, and the removal of metallic impurities in a magnetic separator. The mixture prepared for vitrification was about 50% of bottom ash, 5% fly ash, and 45% cullet. The product was obtained as a clear, dark green glass characterized by excellent durability and negligible leaching of alkali [49].

For ash from the incineration of sewage sludge, shredded glass cullet and limestone were also added, and then heated at 1450ºC. The resulting glass products were commercially used in the manufacture of ceramic tiles and as insulation material [50].

There also have been successful attempts of vitrification of furnace slag mixed with sand. A mixture comprising 50-75% of slag is preheated to 1100ºC, which becomes molten at 1450ºC. The products were obtained with good mechanical properties and resistance to external factors. It was proposed to use them in the production of wall and floor tiles [51].

Other reports show the possibility of slag and dust vitrification from an electric arc furnace with the addition of kaolin clay [52]. It has been found that the process of melting occurs at a temperature of about 1200ºC, and then after cooling the products were vitrified. Pelino [53] describes vitrification of zinc dust electric-arc furnace with the addition of cullet and sand. On the other hand, Kim et al. [54] added steelmaking waste to 30-50% calcium borosilicate, which melts at 1350ºC. Attempts at vitrification of slag with the addition of sand and dolomite were also conducted by Duca et al. [55], who found that the product can be used as a replacement for pottery.

Refining slags with yellow phosphorus were successfully vitrified when mixed with quartz sand, feldspar, calcium carbonate, and sodium nitrate while heating to 1400-1420ºC for 2-3 h in a corundum crucible [56]. The resulting glass contained up to 55% phosphoric slag and can be used to produce glass-ceramic materials. Similarly vitrified metallurgical dust from the flotation of copper with the addition of silica, sodium, and calcium carbonates was successful with heating to 1200ºC [57]. In tests very low levels of toxic metals in water extracts were found.

Vitrification goes for the flotation waste mixed with blast furnace slag and glass cullet, sand, and limestone. The resulting products can be used to produce ceramic materials with the appearance of granite. Similar were vitrified metallurgical dusts of a nickel production plant in Slovakia using the method of melt microwave radiation [24] and the products of glass-ceramics.

Barbieri et al. [27] vitrified ash from a coal power plant mixed with up to 50% by weight of glass cullet and dolomite, yielding coloured glass and glass-ceramic products. They found that the higher the silica content in the ash, the more readily the process forms crystalline structures of glass, and for lower silica the content of the structural changes are slower and require a suitable heat treatment. The coal ash melting process was carried out at 1500ºC for five hours. Researchers have found that the addition of the flux allows the melting temperature of the charge to be reduced to 900-1100ºC, which is sufficient to form glass material [29].

Similarly, Francis et al. [33] mixed 10% to 60% of ash of the El Maghara coal power plant in Egypt with fine-grained cullet of soda-lime glass, and then melted them at 1500ºC. Glass-ceramic products were obtained, which also fired at 800-1200ºC to compensate for the stress in the crystal structure. Research presented a comprehensive analysis of the thermal process of heating and melting the setting of the activation energy of crystallization using a modified Kissinger method.

Asbestos waste vitrification is the only presently known way to allow their use as a construction material [58, 59]. Such waste can be converted into a product by heating with a glass cullet in an electric furnace at a temperature above the decomposition temperature of asbestos [60]. The waste asbestos and fine glass can be delivered to the furnace by a screw mixer, wherein the furnace pressure must be maintained in order to avoid the possibility of contamination from the outside.

Asbestos vitrifying blocks can also be obtained by using plasma arc heating [61] and melting the liquid phase by microwave radiation [59]. A pilot trial vitrification of asbestos waste and asbestos-cement waste was also carried out using resistive Joule's method [62]. In this experiment, asbestos material encapsulated in soil was placed in a stainless steel vessel and graphite electrodes were inserted. The applied high current melted at 1500ºC and cooled to give a glassy 100 kg slab.

The vitrification method was also applied successfully to the disposal of medical waste using a plasma torch under

Table 2. A brief description of melting methods.

No.	Melting methods	Description	References
1	Melting in a crucible furnace	A cyclical process waste mixed with fine glass is melted in a crucible that also serves as a tank for the vitreous phase	[56]
2	Melting in an induction furnace	A continuous process waste mixed with fine glass is melted by induction in steel containers limited durability of the furnace due to corrosion of the tank	[68]
3	Resistance melting by Joule	A continuous process waste and fine glass are melted in a ceramic crucible using an electric current of high intensity the liquid phase collects on the walls of the crucible, and then is discharged to a separate tank	[62]
4	Induction melting in a crucible	A continuous process used a water-cooled induction furnace the liquid phase collects on the walls of the ceramic crucible replacing the steel crucible ceramic refractory materials significantly extends the life of the induction furnace	[60]
5	Plasma melting	The use of very high temperatures disadvantage is the limited lifetime of the plasma-generating torch used in the United States to vitrify contaminated soils	[65]
6	Microwave melting	A cyclical process melting and collecting the liquid phase takes place in the same container the method is energy-saving as a microwave furnace is small and can be installed at the waste location	[69]
7	Volume melting	A cyclical process it is used in a large steel vessel lined with refractory material, in which the melting process occurs along with the collection of the liquid phase	[70]
8	Induction electrodeless melting	Alternative method single melting system in the basic convertor furnace	[71]

heating to a temperature of 1550ºC [63]. The waste consisted of combustible parts such as bandages, gauze, swabs, etc., and non-combustible parts such as fine glass, scalpels, needles, and syringes. As a result of the heating of the plasma two fractions were obtained: slag and glassy form. Analysis confirmed that the composition of the vitrified waste is mainly composed of silica and oxides of Ca, Cr, Al, P, and Zn. An attempt to wash showed good resistance to leaching of heavy metal ions.

Sobiecka [64] presented a method for vitrification when ash from the incineration of medical waste is mixed with chemical preparations containing the compounds SiO_2, $Na_2B_4O_7$, $CaCO_3$, KNO_3, CaF_2, and $BaSO_4$. The melting was carried out in the plasma arc and, depending on the type of additive, occurred at approx. 1000ºC to 1600ºC. In order to produce a stable vitrified product the effects of the addition of compounds SiO_2 and $Na_2B_4O_7$ – in a proportion of 2 to 5% – was found to be beneficial. These additives allowed lowering of the heating temperature, obtaining a low leaching of heavy metals and high hardness of the vitrified product.

Vitrification technology of high-level waste generated in industrial processing of exploited nuclear fuel from light water reactors (pressurized and boiling) has been implemented in France (La Hague), the UK (Sellafield), Japan (Tokai, Rokkashomura), and Russia (RT-1 and RT-2). It is the delivery of a significant amount of thermal energy required to form the amorphous structure, and then rapidly cooling the material [65].

To vitrify these wastes, one-step and two-step systems are used. The one-step system heats the substance in a crucible to 1300-1450ºC, wherein the partial liquefaction of the waste occurs, and then melting continues until the end of constituent migration processes. In this installation, it is necessary to add a fine glass to act as flux and easily bring the mixture to form a semi-solid, often in a form of paste [66]. In the two-step process various heating steps of heating and melting material take place in separate devices. During heating, the addition of chemical compounds also disposes of organic compounds found in waste. The final form of the vitrified waste takes a form of blocks in the shape of a vessel in which cooling takes place. Vitrified radioactive waste is characterized by high resistance to mechanical damage, low chemical reactivity, and minimal release of harmful substances into the environment [67].

Therefore, the methods for vitrification of waste are already well known and used on an industrial scale worldwide. In the existing plants in France and Great Britain the furnace charge is continuously used to introduce melting (Table 2). In the United States, Japan, Russia, and China, melting the batch is done in ceramic crucibles by Joule. The advantage of the melting crucible is the reduction and secretion of waste gases and small amounts of impurities remaining after the process.

There is more research to determine specific properties of the resulting products (vitrified waste), in particular regarding sustainability to ensure safe storage or in specific commercial applications. For example, processing technology of fly ash developed by Rocktron from the UK [www.rktron.com] into small glass spheres using select ash rich in silicates to facilitate vitrification without the use of silica additives. The resulting products are named MinTron – glass microspheres with a full diameter of 8 to 14 microns and balls with a diameter of 5 to 9 microns. They are used as mineral fillers, functional plastics and rubbers, and as additives for processing by extrusion, injection molding, and compounding. Rocktron together with the Ford Motor Company plan to create new types of hybrid composites for applications in car body structures. Glass microspheres have major advantages in comparison with conventional fillers such as composites of polypropylene and polyamide, including improved resistance to scratches and abrasion. In the near future, empty glass microspheres will also be produced for thermal insulation and weight reduction of plastic components.

Vitrification of Ashes from Combustion of Sewage Sludge

Vitrification to Make Construction Aggregates

Vitrification of ashes from fluidized combustion of sewage sludge collected in the municipal sewage treatment plant in Lublin, Poland, was examined. The concentration of heavy metals in the waste is on average four or five times higher than the concentration of metals in the sludge, due to a reduction in the weight of the incinerated waste. For this reason, dumping the ash in landfills is cumbersome due to the need to isolate groundwater from contamination and the need for protection against dusting [74].

With this in mind, it was proposed to merge the ash into briquettes by adding a portion of powdered glass. It was found that adding from 40% to 50% of powdered waste glass (lamps, domestic glass) in particles up to 0.2 mm facilitates the formation of the liquid phase and allows for the reduction of the melting point of the surface of the body, thereby reducing the energy expenditure. Lots of ash pellets were placed in a laboratory 1400 W furnace, and were heated at from 1050ºC to 1200ºC. Warm-up time ranged from 30 to 90 minutes. The better results were obtained for samples containing ashes, glass dust, and cement (5% by weight). After intensive water cooling, it was found that the surface of these samples formed a layer of glass, wherein the shape of the body was taken [73].

The basic physical properties of the vitrified pellets (bulk density, surface area, compressive strength, and resistance to gravity dump) were tested. The measured values were compared with the limit values, which met the minimum requirements for mineral and artificial aggregates used in road construction [73].

Harmful substances that leach into the environment after they are vitrified also were tested. The results showed a much smaller concentration of ions in the leachate, as compared to the limit values for the leaching of waste after treatment in landfills. It was found that almost no heavy metals were contained after vitrification. This means that environmentally safe product was obtained [73].

Vitrification to Make Glass Beads

There is a method of vitrification of ash from combustion of sewage sludge to achieve a shape of regular balls, referred to as "glass beads." The technology for producing glass beads includes [74]:

- Preparation of the waste material to melt
- Heating and melting in a high-temperature furnace
- Intensive air-cooling, whereas the liquid material goes to form the glassy phase

The technological line for the production of glass beads with a screw mixer can operate on ash from bed combustion of sewage sludge, as well as from coal power plants. Modifiers are added (e.g. boric acid) to facilitate liquefaction in the furnace. The blend is molten in an induction furnace crucible, and then the liquid phase (spout) is fed into the dryer with an intensive blow of air, which disintegrates the material into fine particles that cool into glass beads. Small fractions of beads are carried with the airflow of the upper part of the dryer and fall onto a conveyor, and then to the tank. Larger and heavier parts are re-melted in an induction furnace. For heating and melting of ash rotary kilns, microwave ovens and plasma furnaces can be used [74].

Glass beads with a diameter of about 1-2 mm are applicable, inter alia, as a substitute for the so-called proppants in hydraulic fracturing of shale gas and oil. Hydraulic fracturing involves pumping fracturing fluid into the wellbore at high-pressure (600 atmospheres). The fluid enters the wellbore and then runs through horizontal holes, causing cracking shale formation and an extensive network of narrow (max. 2 mm) slots through which hydrocarbons emerge. To prevent closing of these cracks upon the withdrawal of the blood pressure and the subsequent impact of the rock pressure, the fracturing fluid is injected into wellbore proppants that keep the slots open all the time. After finishing the portion of the injected fracturing the fluid returns to the surface, then a portion, together with proppants, goes inside the rock. Commonly used proppants are resin-coated silica sand, sintered bauxite, and ceramic. However, ceramic proppants significantly increase the cost of fracturing. For one such fracturing in a typical situation, about 70-100 tons of proppants are used. One shale gas mine consumes 5,000-15,000 tons of proppants [74].

The glass beads made from waste materials will be much cheaper than ceramic, and at the same time provide good mechanical properties and chemical stability. Large surface smoothness and regular shapes allow for increasing gas flow efficiency and reducing the time needed to complete the extraction of the deposit.

Conclusions

The main result of vitrification of ash from combustion of sewage sludge is to solve the problem of neutralization of heavy metals by transforming them into very stable forms, eliminating their environmental presentation. Another result is to obtain specific shapes and dimensions of glass products, mainly in the ceramic and construction industries. Successful efforts for processing of ash from thermal power plants and municipal waste incinerators containing sulfur compounds have already been conducted, with lead and other elements transformed in such a way as to be neutralized.

The last result of vitrification is developing a methods for commercial production of vitrified aggregates and regular glass beads as well. Vitrified pellets of ashes were found as substitutes of mineral and artificial aggregates used in road construction. Mini-glass balls were applicable, inter alia, as additional proppants in hydraulic fracturing of shale gas and oil.

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