

# Recycling Possibilities of Sewage Sludge from Water Purification

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The sewage sludge from a water purification plant was analysed for recycling to the dense glass-ceramic materials. The sewage sludge containing aluminium compounds and circa 50 % of organic matter was used as a filler, but as the matrix of glass-ceramic was examined an illitic clay from Liepa deposit (Latvia). The raw materials were investigated by differential-thermal (DTA) and X-ray diffraction analysis (XRD). The dense glass-ceramic was produced from the water purification sewage sludge and limeless clay in ratio 10-40 : 90-60. The optimal values of bulk density (2.28 g/cm<sup>3</sup>) and thermal shrinkage (29 %) for novel materials were observed for glass-ceramic with the sludge additive at amount of 20 wt %, obtained by sintering at temperature 1140 °C. The relationship between thermal treatment conditions and maximal treatment temperature was established. The following crystalline phases for novel materials were detected by XRD analysis: microcline (KAlSi<sub>3</sub>O<sub>8</sub>), hematite (Fe<sub>2</sub>O<sub>3</sub>), quartz (SiO<sub>2</sub>) and corundum (Al<sub>2</sub>O<sub>3</sub>). The physical-chemical properties of novel materials correspond to the dense glass-ceramics composite while the material itself could be applied as building material.

Keywords: *recycling, sewage sludge, glass-ceramics.*

## 1. Introduction

Annually increasing amount of dry sewage sludge from water purification gradually increases their storage problem. Partly sewage sludge is used as fertilizer in the agriculture or forestry, while still unusable is the part of it containing the harmful admixtures or heavy metals.

Activated sludge from water purification equipment is assemblage of microorganisms (aerobous and anaerobous) having the ability to absorb the organic pollution and decompose it at the presence of air oxygen or without it. This ability is used for purification of waste or drinking water. Activated sludge contains the biologic organisms – bacteria's, protozoans and other microorganisms where the bacteria's create the main content in the activated sludge. In most cases the chemical composition of activated sludge can be described by chemical formula C<sub>3</sub>H<sub>7</sub>O<sub>2</sub>N (Ledins 2007).

As one of the most cheapest and safe methods of elimination of pathogenics in the purification sludge is their composting. The compost produced is used as fertilizer in agriculture and forestry if the pollution of dry matter with hazardous elements does not exceed the threshold limit value (Shinogi et al. 2003, Lu et al. 2008). As one of the possibilities for recycling of sludge containing large amount of heavy metals or other hazardous elements is their addition to concrete, for example, mix from half-and-half of Portland cement and sludge kept either in the autoclave or under pressure. The end-product could be used

as building material (Cheilas 2007). Other possibility for recycling of sewage sludge is to apply it as the filler for clay bricks, described in literature (Chih-Huang et al. 2003), whereas the limited amount of sludge in bricks is until 20%, thus reaching the requirements of brick's mechanical and physico-chemical properties.

Another possibility for recycling of industrial waste, i. e. sewage sludge, is the production of glass-ceramic materials using powder technology and sintering by thermal treatment. Only specific glass compositions are suitable precursors for glass-ceramics. Some glasses are too stable and difficult to crystallize, such as ordinary window glass, whereas others crystallize too readily in an uncontrollable manner resulting in undesirable microstructures. Secondly, heat treatment is a critical factor in attaining acceptable and reproducible products (Rawlings et al. 2006, Suzuki and Tanaka 1999). As the main methods for recycling of sewage sludge the powder technology and one-stage thermal treatment for production of glass-ceramic is selected. The raw materials used for the given studies were sewage sludge from drinking water preparation plant and limeless clay from deposit Liepa (Latvia). The limeless clay is suitable for press powder-mix while it can form the liquid phase during sintering process which is necessary for creation of glassy matrix to incorporate the part of sewage sludge (Sedmalis et al. 2002, Sedmale 2010, Rawlings et al. 2006, Svinka et al. 2011). The advantages of this method – it is simple,

economically profitable, necessary equipment is available in already running technologies and obtained products usually are characterized with high mechanical strength.

## 2. Methods

The information about used raw material – clay is based on literature data (Svinka and Lindina 1994, Svinka *et al.* 2011, Sedmalis *et al.* 2002), while the information concerning the used sewage sludge is based on producer's data. The mineralogical composition was determined by XRD analysis, while the determination of moisture, organic matter and ash content – performed according to the Latvian Standard (LVS EN Report 13039:2012). The raw materials were dried for 24 h by 100 °C temperature and the mixtures were created from 60 to 90 wt % of clay and 40 until 10 wt % of sewage sludge. The compositions were milled in mechanic mill RETSCH RM100 for 1 hour in order to get the powder fraction with particle's  $\varnothing \leq 0.5$  mm. Discoid samples ( $\varnothing = 10$  mm) were prepared using WEBER PW-10 hydraulic press by pressure 6 kN (19.1 MPa) and treated thermally in NABERTHERM electric furnace (max temperature – 1300 °C) with following high-temperature treatment rate: increasing rate 3 °C/min until maximal temperature interval 1100 - 1180 °C and keeping time by maximal temperature - 1 h. The bulk density, water uptake and porosity were measured using standard method (LVS EN Standard 10545-3:2002). The samples of sewage sludge and sintered glass-ceramic materials made from the already mentioned Latvian industrial wastes and clay were analysed by XRD analysis. Powdered glass-ceramic materials were used for XRD patterns, recorded on a RIGAKU ULTIMA+ X-ray diffractometer with 50mA and 40 kV, Cu K  $\alpha$  radiation. The mineralogical identifications were made using an XRD pattern database (International Centre for Diffraction Data, ICDD).

## 3. Results and Discussion

### Investigation of raw materials

The sewage sludge contains biogenous elements needed for bacteria's in the activated sludge – nitrogen, phosphorous, oxygen, carbon and hydrogen, as well as some trace of aluminium (Al), while as the coagulant in

the water purification process the aluminium sulphate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ ) was used (data from producer). During the entering into the water the hydrolysis of aluminium takes place which resulted with aluminium hydroxide and its polymers.

The calculated content of moisture is 84.11 wt %, part of organic matter corresponds to the 55.60 wt %, while the ash content of sewage sludge is 44.03 wt %.

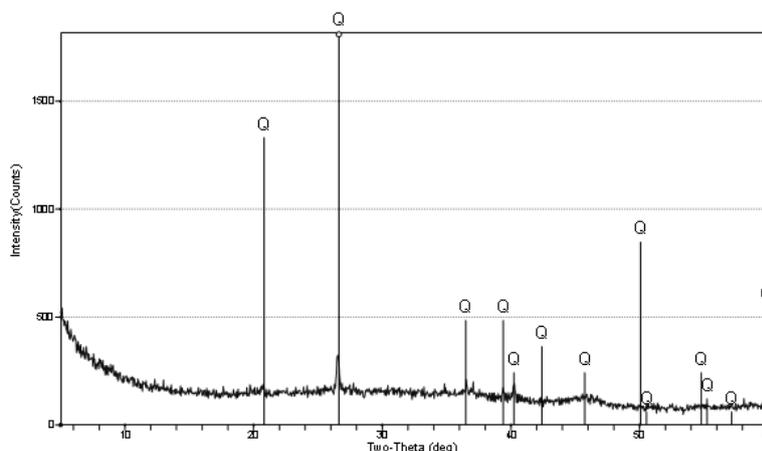
**Table 1.** Chemical composition of applied clay

Oxides	Content, wt %
$\text{Al}_2\text{O}_3$	16.75
$\text{SiO}_2$	63.18
CaO	0.82
$\text{TiO}_2$	0.81
MgO	2.07
$\text{Fe}_2\text{O}_3$	7.64
$\text{Na}_2\text{O}$	0.14
$\text{K}_2\text{O}$	3.46
Organic substances	0.24
LOI (1000 °C)	4.89

By the XRD analysis for sewage sludge treated at temperature 700 °C as main crystalline phase the quartz ( $\text{SiO}_2$ ) was detected (see in Fig. 1). The clay used as a matrix for glass-ceramic shows the high amount of silica ( $\text{SiO}_2$ ), alumina ( $\text{Al}_2\text{O}_3$ ) and iron oxide ( $\text{Fe}_2\text{O}_3$ ) (see in table 1) (Sedmalis *et al.* 2002).

The data of chemical and mineralogical compositions of applied clay as well as the ability to make light weight aggregates was obtained from literature (Sedmalis *et al.* 2002, Svinka and Lindina 1994, Svinka *et al.* 2011). The XRD analysis indicates the presence of illite, kaolinite and quartz, as well as hematite and trace minerals – orthoclase and microcline in the clay (Svinka *et al.* 2011). The ratio of clay and sewage sludge for selected four optimal compositions mixed from the raw materials S1, S2, S3 and S5 are given in table 2.

The mentioned mixtures were created in order to detect the possible volume of each addition for production of dense ceramic composites with exact complex of properties – to create a glassy and crystalline phases during thermal treatment.



**Fig. 1.** The XRD pattern for sewage sludge treated at temperature 700 °C Abbreviation: Q – quartz ( $\text{SiO}_2$ )

**Table 2.** Content of raw materials in the waste mixtures

Components, wt %	S1	S2	S3	S4
Clay from deposit Liepa	90	80	70	60
Sewage sludge	10	20	30	40

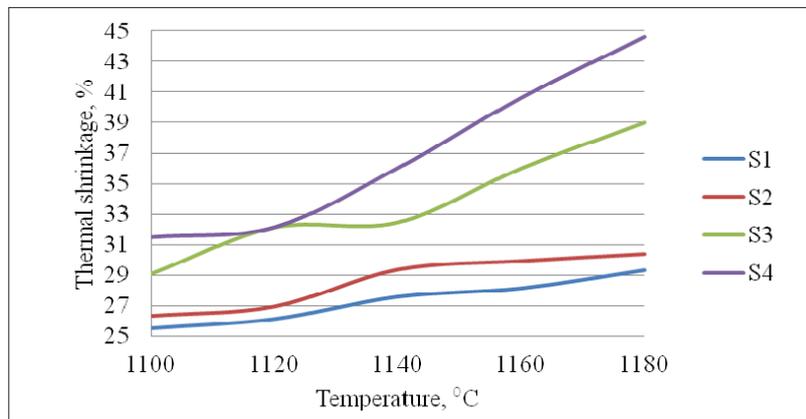
*Physical-chemical properties of glass-ceramics*

Fig. 2 shows the increase of thermal shrinkage of all investigated series of mixes versus the increasing temperature of thermal treatment. The highest thermal shrinkage values indicate the mixtures S3 and S4 – 35 and 45%, respectively. The curves of shrinkage for composition S3 and S4 became to be sheerer with temperature 1140 °C, while the shrinkage curves for compositions S1 and S2 are shallow (Fig. 2).

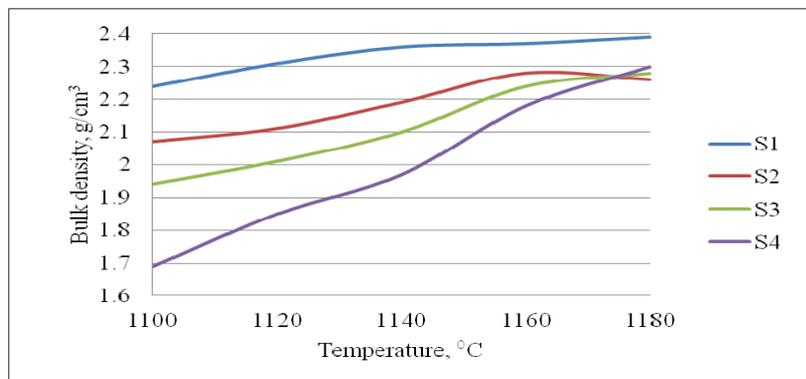
The highest bulk density (2.36 – 2.39 g/cm<sup>3</sup>) shows the composition S1 (with 10 wt % of sewage sludge) in temperature range 1140 – 1180 °C when probably starts the melting process of some crystalline phases in the clay (see Fig. 3).

Whereas for all compositions the similar increase of bulk density until temperature 1160 °C could be observed.

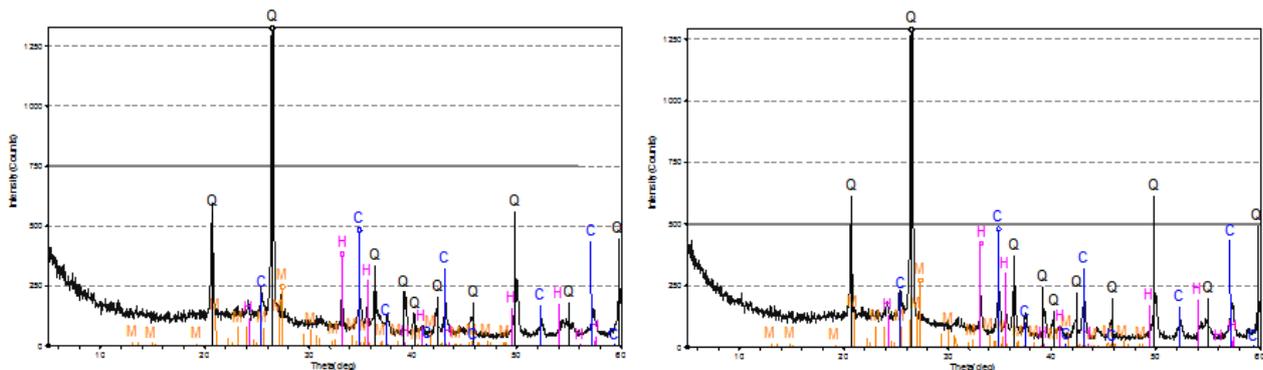
The increase of bulk density until the maximum 2.28 g/cm<sup>3</sup> and subsequent slow decrease (from the maximum 2.28 g/cm<sup>3</sup> at 1140 °C temperature) until 2.25 g/cm<sup>3</sup> at temperature 1180 °C could be observed for composition S2 containing 20 wt % of sewage sludge (see in table 2). For all other compositions (S1, S3 and S4) the bulk density does not have a maximum, while the curves continue to rise even at the higher temperatures than 1180 °C (Fig. 3).



**Fig. 2.** The thermal shrinkage of materials versus thermal treatment temperature



**Fig. 3.** The bulk density of materials versus thermal treatment temperature



**Fig. 4.** The XRD pattern for glass-ceramic S4 sintered at temperature 1100° C (left) and 1160° C (right) Abbreviations: Q – quartz (SiO<sub>2</sub>), M - microcline (KAlSi<sub>3</sub>O<sub>8</sub>), H - hematite (Fe<sub>2</sub>O<sub>3</sub>) and C - corundum (Al<sub>2</sub>O<sub>3</sub>)

In general the mineralogical composition of material S4 for both sintering temperatures – 1100 °C and 1160 °C (Fig. 4) show similar crystalline phases – microcline ( $\text{KAlSi}_3\text{O}_8$ ), quartz ( $\text{SiO}_2$ ), hematite ( $\text{Fe}_2\text{O}_3$ ) and corundum ( $\text{Al}_2\text{O}_3$ ) with insignificant higher peaks of intensity for quartz and hematite for material sintered at temperature 1160 °C (Fig. 4 right).

The discussions on the chemical and mineralogical composition of Liepa deposit clay is given in literature (Svinka and Lindina 1994, Svinka et al. 2011). By the XRD analysis in the untreated clay the illite, kaolinite and quartz, as well as hematite and trace minerals – orthoclase and microcline was detected (Svinka et al. 2011).

The XRD analysis for sewage sludge treated at temperature 700 °C as the main crystalline phase shows the quartz (see Fig. 1). Whereas according to the producer's data in the composition of sewage sludge a big amount of alumina due to coagulation's agent added during the water purification process is present. Thus it could be conclude that aluminium in the composition of mixtures after thermal treatment is not involved into the crystalline structure of material.

According to literature data (Svinka and Lindina 1994, Svinka et al. 2011) and results of investigation of sewage sludge, the largest amount of quartz, microcline and hematite could be formed on the basis of high-temperature mineralogical transformation of used clay (Sedmalis et al. 2002, Svinka et al. 2011), while the microcline and corundum partly were formed probably during the reactions between the clay as a matrix and the sewage sludge with high aluminium and some trace of iron content. The orthoclase and microcline due to their fluxing behaviour could be basis of formation of liquid phase during sintering process resulted with high density during sintering of mixtures (Wattanasiriwech and Wattanasiriwech 2011). The limeless clay is suitable for press powder-mix while it can form the part of liquid phase during sintering process which is necessary for the creation of glassy phase for incorporation of sewage sludge into the glass matrix (Sedmale 2010, Rawlings et al. 2006, Svinka et al. 2011).

The rapid increase of thermal shrinkage of mixtures S3 and S4 by temperatures higher than 1140 °C (Fig. 2) could be explained with the higher sewage sludge content in those compositions (30% for composition S3 and 40% for composition S4, respectively). Probably at these temperatures more liquid phase could be created due to the high content of aluminium and other metals in sludge composition. While for compositions S1 and S2 the content of sewage sludge is lower therefore the curves is more shallow, which is more suitable for production of dense glass-ceramic due to longer sintering interval (Rawlings et al. 2006).

The increase of bulk density until the maximum 2.28 g/cm<sup>3</sup> at 1140 °C temperature for composition S2 containing 20 wt % of sewage sludge (see in table 2) shows the beginning of sintering interval of composition, while the slow decrease of bulk density - from the temperature 1140 °C until 1180 °C (Fig. 3) shows the continuing of the sintering interval of composition. This composition S2

according to their presented relations between bulk density and thermal shrinkage could be the optimal for production of glass-ceramics while this shows the similar properties to other previously investigated glass-ceramics (Rozenstrauha et al. 2011) and could be suitable for running ceramic's production technologies without significant changes.

For compositions S1, S3 and S4 the bulk density curves does not have a maximum, but they continue to ascend. These processes resulted with high thermal shrinkage for compositions S3 and S4 – 35 until 45% (see Figure 2), where the shrinkage curves show rapid increase, which could be explained with the melting of crystalline phases containing alkali metals and vaporisation of volatile elements in the compositions content. In this temperature (from 1140 until 1180 °C) the amount of liquid phase is probably too high (due to the presence of large amount of sewage sludge 30– 40 wt. %, see in Table 2) which results in high thermal shrinkage and subsequent weakness of materials shape of compositions S3 and S4.

The results of XRD analysis of series S4 sintered at temperature 1100 °C indicates the following crystalline phases: microcline ( $\text{KAlSi}_3\text{O}_8$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), quartz ( $\text{SiO}_2$ ) and corundum ( $\text{Al}_2\text{O}_3$ ) (see Figure 4). If to compare the mineralogical composition of series S4 at both sintering temperatures (1100 and 1160 °C), then insignificant higher intensity peaks of quartz and hematite crystalline phases for materials sintered in temperature 1160 °C (Fig. 4, right) could be mentioned. Therefore it could be concluded that the formed crystalline phases in temperature 1100 °C are stable until the temperature 1160 °C and starts to melt at higher temperatures. This consequence can be suitable for production of investigated materials with longer sintering temperature interval for composition S4.

For further investigations of produced novel materials the mechanical properties tests and studies of microstructure are necessary in order to acquire the suitability of products for building ceramics.

#### 4. Conclusions

The present study was focused on the recycling of sewage sludge from water purification into the glass-ceramic materials. The novel glass-ceramic composites were produced from both raw materials – sewage sludge (10 – 40 wt %) and limeless clay (90 – 60 wt %) using a simple technology route – powder technology and sintering during one-stage thermal treatment.

The optimal bulk density 2.28 g/cm<sup>3</sup> observed for composition S2 containing the 20 wt % of sewage sludge, while the composition with highest content of sewage sludge (40 wt %) resulted with high bulk density ratio 2.30 g/cm<sup>3</sup> by temperature 1180 °C, subsequent increasing of density by higher temperatures until 1180 °C and at the same time the rapid increase of thermal shrinkage 35 – 45 %.

The results of XRD analysis of material S4 sintered at temperature 1100 °C indicate the following crystalline phases: microcline ( $\text{KAlSi}_3\text{O}_8$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), quartz ( $\text{SiO}_2$ ) and corundum ( $\text{Al}_2\text{O}_3$ ) which is base for crystalline structure of novel materials.

The direction of application of produced glass-ceramics – suitability for building materials could be better evaluated by microstructure studies and mechanical tests.

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