APPLYING OF ALUMINIUM SCRAP RECYCLING WASTE (NON-METAL PRODUCT, NMP) FOR PRODUCTION ENVIRONMENTAL FRIENDLY BUILDING MATERIALS

Diana Bajare[1], Aleksandrs Korjakins[2], Mareks Zakutajevs[3], Filips Voroneko[4]

[1] *Riga Technical University , Kalku str. 1 LV-1658, Riga, Latvia. E-mail diana.bajare@rtu.lv*
[2] *Riga Technical University , Kalku str. 1 LV-1658, Riga, Latvia. E-mail aleks@latnet.lv*
[3]*Riga Technical University , Azenes str. 16/20 LV-1048 Riga, Latvia. E-mail mareks.zakutajevs@gmail.com*
[4]*Riga Technical University , Azenes str. 16/20 LV-1048 Riga, Latvia. E-mail philip.voronenko@gmail.com*

Abstract. An aluminium scrap recycling waste called non-metal product (NMP) was tested as raw material for production of expanded clay aggregates. Processing of NMP created in the aluminium scrap recycling companies is one of the most challenging tasks due to its toxic nature - in accordance with the Basel Convention, Annex III, marking of this waste is H 4.3 (reaction with water results in highly inflammable substances) and H 10 (reaction with water results in increased concentration of toxic gases, for instance, ammonia). The main phases of the starting waste are spinel ($FeAl_2O_4$) or ($MgAl_2O_4$) and aluminium nitride (AlN). The main task of the present research is to investigate the possibility of non-waste utilization of NMP and reprocessing it into environmental friendly material. The heat treatment in the temperature higher than 1100 ^oC was selected as appropriated method in order to eliminate impurities of NMP, like ammonia and others. It is approved by results obtained from chemical and mineralogical investigations of expanded clay aggregates. The phenomenon of gasses emission during the heat treatment of NMP is used as initiator for creation of extra porous structure of sintered ceramic bodies. Physical, mechanical and microstructural properties of sintered aggregates were determined as well as chemical and mineralogical composition of raw materials and end product were fixed.

Keywords: Lightweight expanded clay aggregates, aluminium scrap, recycling waste, non-metal product (NMP), environment.

1. Introduction

Recycling aluminium currently makes up a third of the total aluminium used in the world. Recycling is an essential of the aluminium industry, given that this process makes economic, technological and ecological sense (Gil 2005). From one side the aluminium recycling waste has a toxic nature and land filing is not suggested, from other side these waste should be used as raw material for production of new building materials. In many countries the utilization and recycling of secondary raw materials is subsidized by the state. In this way the natural resources are saved, environmental pollution decreased, more resources for safe storage and management of waste allotted and cost-efficiency of enterprise increased.

The consumption of aluminium recycling waste has been rising continuously worldwide, which is great stimulus for developing non-waste technology (Shinzato and Hypolito 2005; Samuel 2005; Shen and Forsberg 2003; Lucheva *et al*. 2005). Aluminium dross represents a residue from primary and secondary aluminium production. Drosses are classified according to aluminium metal content into white and black dross. White dross has higher metal aluminium content and it is produced from primary and secondary aluminium smelters, whereas black dross has a lower metal content and is generated during aluminium recycling (secondary industry sector). Black dross typically contains a mixture of aluminium oxides and slag with recoverable aluminium content ranging between 12–18% (Gil 2005; Shen and Forssberg 2003; Lucheva *et al*. 2005). The conventional rotary furnaces heated with a fuel or a gas burner is used to recover the extra aluminium from black or white dross. This treatment process produces the non-metal product called aluminium recycling waste containing alumina, salts, impurities and a little amount (3–5%) of metallic aluminium (Gil 2005; Tzonev and Lucheva 2007). This case study investigates the use of aluminium recycling waste (non-metal product NMP) generated from dross processing. The terms aluminium recycling waste and non-metal product (NMP) are used interchangeably in this case study.

The composition of NMP is highly variable and usually unique to the plant generating the waste, hence finding potential application for this material is often seen as a difficult task (Gil 2005; Bajare *et al*. 2007; Bajare *et al*. 2008). The task becomes more challenge due to toxic nature of NMP. It emits flammable gases, such as acetylene, or it is liable to give off toxic gases, such as ammonia, in dangerous quantities in contact with water (Proposal to regulate salt slag under the Hazardous waste (Regulation of Exports and Imports) Regulations 1996). Beside this, the waste contains compounds like soluble salts, oxides, carbides and sulphides as well as metallic aluminium (Gil 2005; Tzonev and Lucheva 2007; Bajare *et al*. 2007; Bajare *et al*. 2008).

However there is a lot of patents and scientific approving for possibilities to used NMP as source of aluminium compounds for cement (Aza 2003; Diaz 2005), metallurgy (US Patent 4252776 1981), chemical (Lucheva *et al*. 2005; US Patent 5407459 1995; US Patent 5132246 1992; US Patent 5424260 1995; US Patent 4252776 1981) and building industry (Shinzato and Hypolito 2005; Lucheva *et al*. 2005; US Patent 5045506 1991). NMP could be utilized together with other alternative materials to produce premixes for clinker and ceramic products (Yoshimure *et al*. 2008; Garcia-Valles *et al*. 2008) as well as it can be used as raw material for production of concrete, mineral wool and materials with ultra high resistance to fire (Shinzato and Hypolito 2005; Yoshimure *et al*. 2008; US Patent 7015167 2006; Maschio 1988). The aim of research is a same - to develop processes for converting of NMP to high value-added products. Scientific justification of successful application of NMP in the different fields is changes in the mineralogical and chemical composition of NMP during high temperature treatment.

The main goal of research is investigation of possibilities to recycle the NMP in order to produce useful commercial product - porous ceramic building materials with extra high porosity and unique pore structure.

The expanded clay aggregate (ECA) is important building material in the regions where natural lightweight aggregates are not available. From the other hand ECA should be manufactured from widely available material - clay. The properties of final product mainly depend on the type of clay, the type and amount of pore – formatting agents and the sintering temperature of aggregates. The main aim of this experimental work is to examine the effect of the NMP usage as a pore formatting agent on the physical and micro structural properties of ECA produced at different sintering temperatures.

2. Experimental study

2.1 Characterisation of raw materials

In the current study, lightweight expanded clay aggregates (ECA) were produced from clays with high content of carbonates and aluminium scrap recycling waste (NMP) in the different compositions. In this experimental investigation NMP was used as extra pore forming agent. The chemical composition of clay is given in Table 1, beside that dolomite (21.6 %) is determined in clay composition. X-ray diffraction (XRD) analysis of clay is shown in Figure 1, which indicates that clay is composed of quartz, calcite, dolomite, iolite, kaolinite and anorthoclase. According analyses clay used in experimental studies is typical carbonate clay.

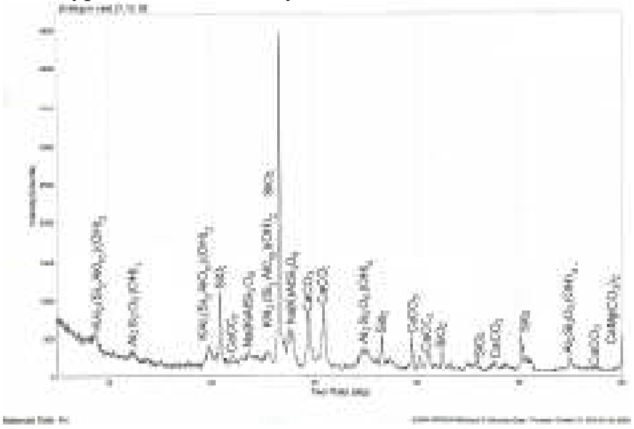

Fig. 1. The mineralogical composition of clay used in the experimental investigation.

According to the element analysis resulted from inductive coupled plasma optical spectrometry (ICP-OES), atomic absorption spectroscopy (AAS) and potentiometer titration analyses, the NMP contains: aluminium (Al) – 34.4%, silicon (Is) – 4.4% magnum (Mg) – 2.4%, calcium (Ca) – 1.32%, sodium (Na) – 1.69%, potassium (K) – 2.31%, sulphur (S) – 0.07%, chlorine (Cal) – 4.23, ferric (Fe) – 3.6%, copper (Cu) – 0.99%, zinc (Zn) – 0.6%. These data corresponds to chemical composition of aluminium recycling waste, which is given in Table 1. From the point of chemical composition analyzed wastes content also aluminium nitride (LAN) - on average 5%, aluminium chloride – ($AlCl_3$) - on average 3%, potassium and sodium chloride (Nail +ACL) – totally 5% and ferric sulphide ($FeSO_3$) - on average 1%.

Table 1. Basic chemical composition of clay and aluminium recycling waste (amount %)

	Clay	Waste
Al_2O_3	14,34	63,19
SiO_2	50,22	7,92
CaO	8,54	2,57
SO_3	0,07	0,36
TiO_2	0,56	0,53
Na_2O	0,43	3,84
K_2O	3,09	3,81
MgO	3,07	4,43
Fe_2O_3	5,74	4,54

The mineralogical composition of the NMP is determined by using the XRD analysis (Figure 2). According to the analysis data, the NMP contains corundum (Al_2O_3), silica (SiO_2), ferric sulphite ($FeSO_3$), aluminium chloride ($AlCl_3$), calcium ferric oxide ($Ca(FeO_3)$), calcium, magnesium or ferric carbonate ($Ca(Mg,Fe)(CO_3)_2$), gibbsite ($Al(OH)_3$), spinel ($FeAl_2O_4$) or ($MgAl_2O_4$) and aluminium nitride (AlN).

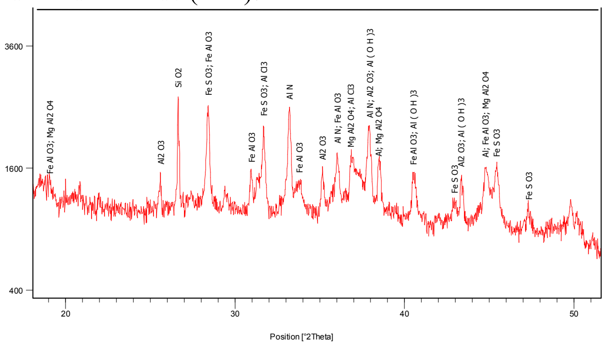

Fig 2. The mineralogical compositions of ground aluminium scrap recycling waste (non-metal product, NMP)

2.2 Preparation and synthesis of lightweight expanded clay aggregates (ECA)

Clay was mixed with NMP in different ratio ranging from 9.1, 16. 7 to 23.1%. The raw materials were ground and mixed together in the planetary ball mill. The average particle size of mixes was 54 µm. Plastic mass was prepared by adding 20–25% water and rounded by shaping operation. Prepared aggregates were dried in an oven at 105 °C to avoid reaction between aluminium recycling waste and water used for preparation of plastic mass.

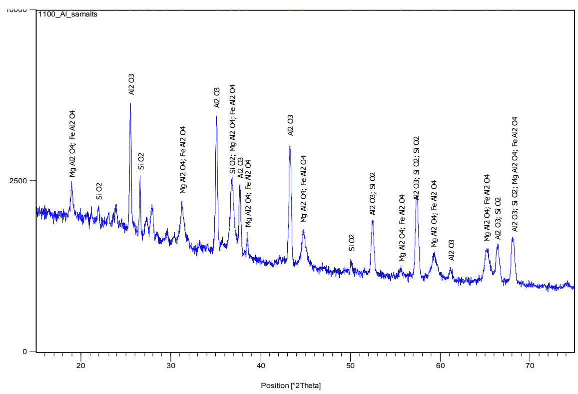

Fig 3. The mineralogical composition of the ground aluminium scrap recycling waste (non-metal product, NMP) treated at the temperature of 1100 °C.

Green aggregates were treated at sintering temperatures from 1170 to 1210 °C in electrically furnace. The rate of temperature increase in furnace was kept constant as 15 °C/min.

The RXD analysis was used for determination of changes in the mineralogical composition of NMP after the heat treatment. NMP was treated separately at temperatures from 1100 to 1500 °C.

Aluminium nitride (AlN), ferric sulphide ($FeSO_3$) and metallic aluminium (Al) were not determined any more, but the amount of ferric oxide (Fe_2O_3) is considerably reduced, in the composition of NMP treated at temperature 1200 °C. Whereas, the amount of corundum (Al_2O_3) and spinel ($FeAl_2O_4$) is considerably increased (Figure 3).

The gasiform substances and new minerals like spinel and alumina are originated during the heat treatment process of NMP. The phenomena of origination of gasiform substances during the heat treatment of NMP should be use as pore creator for obtaining porous structure of lightweight ceramic aggregates (ECA). As hazardous compounds of NMP are transformed in to new ones, non-hazardous compounds, NMP don not have toxic nature any more and it is become environmental friendly and it can be a constituent part of new, environmental friendly material.

This is approved by RXD analyses of expanded clay aggregates (ECA), which is made by using NMP in the different proportions. The mineralogical composition of clay aggregates with NMP additions no depends of proportion and without it is almost same from the qualitative point of view. According to quantitative analyse of mineralogical composition ECA has higher amount of minerals such as spinel and corundum compare with aggregates made from pure clay.

2.3 Characterization of lightweight expanded clay aggregates (ECA)

2.3.1 Particle density

The density of ECA made by using NMP from 9.1 to 23.1% sintered at temperature 1170 °C is in the range from 0.5 to 0.7 g/m^3 (Table 2). The composition and sintering temperature of ECA has fundamental relationships with the particle density, which is also function of the pore structure and physical – mechanical properties.

ECA made by using NMP in the amount of 9.1 and sintered at temperature 1170 °C has density 0.59 g/m^3. The lower density (0.4 g/m^3) of aggregates (16.7% of added NMP) was reached then they were sintered at temperature 1190 °C, but lower density (0.45 g/m^3) of aggregates (23.1% of added NMP) was reached then sintered temperature was increased to 1210 °C.

The evaluation of aggregate expansion due to increasing of sintering temperature was determined by measurement of single particle density and volume. Density of ECA noticeable depends from two factors: amount of added aluminium recycling waste and sintering temperature. Density of green, un-sintering aggregates is in the average 1.85 g/cm^3, but it can decrease to 0.45 g/cm^3 by modification of composition and production temperature of ECA.

2.3.2 Water absorption capacity

The water absorption capacity increases with increasing sintering temperature of ECA due to increases of volume and porosity of aggregates. The maximum water absorption (after 30 min. immersion in water) of ECA sintered at the optimal expansion temperature is in the average 6-12%. The water absorption of ECA increases of 1-3% then immersion in the water was prolonged to 24 h. The highest water absorption after 30 min. (10.2%) was measured on ECA produced from clay mass with addition of 23.1% aluminium waste and sintered at temperature 1210 °C.

2.3.4. Mechanical properties of the ECA

Strength of the ECA is very important property of obtained material form point of view of application this one in the civil engineering.

Specimens of the ECA have been prepared with different proportion – 9.1, 16.7 and 23.1% of NMP, which were added to clay and treated at the temperature 1170 °C. Since the produced expanded clay pieces have round form, the specimens have been prepared in cubic form for evaluation compression strength (Figure 4).

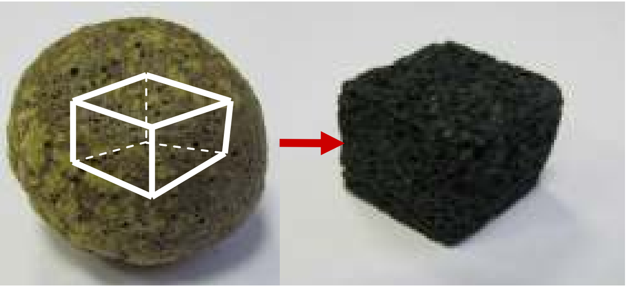

Fig 4. Cat specimen with size ~ 25 x 25 mm

The specimens have been tested by universal testing system Zwick Z100. The obtained results are presented in the Table 2.

Table 2. Physical-mechanical properties of ECA sintered at the temperature 1170 °C.

Added amount of NMP, %	Density, g/cm^3	Compression strength, MPa
9.1	0.59	15
16.70	0.54	6
23.1	0.67	5

The strength of the material is not linearly proportional to density of these one. It may be explained by non-regular forms of the pores in the obtained specimens. The second reason is the occurrence NMP in the ECA isn't reacted with clay.

2.3.3 Microstructure of ECA

Microstructures of the lightweight aggregates (ECA) produced at different temperatures were observed by an optical microscope. The pore structures of ECA are illustrated in Figure 5. It is clear that the pore structure of ECA significantly affected by the amount of added aluminium recycling waste and sintering temperature.

Two types of pores were observed due to microstructural studies of ECA. One type of pores, which can be called macropores have a diameter larger than 1 mm and are interconnected through smaller porous. The macropores have rounded morphology and mainly are closed. They are typical of aggregates sintering at the maximal expansion temperature, which depends from amount of added aluminium waste (Figure 5 a). These pores are typical also of aggregates made from pure clay and sintering in the maximum expansion temperature (1160 °C) where decomposition process of carbonates (calcite and dolomite) is completed

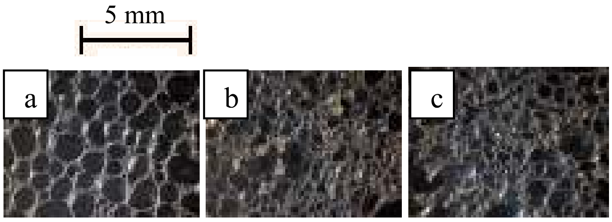

Fig 5. Microstructure of ECA made by adding: a) 9.1% of NMP; b) 16,7% of NMP; c) 23,1% NMP – sintered at temperature 1160 °C.

The second type of pores (micropores) measures smaller than 1 mm. Decomposition of aluminium recycling waste at the temperature, which is maximal expansion temperature of ECA, is the main reason for appearing of interconnected small size (<0,2 µm) pores. Micropores are not observed in the microstructure of aggregates made from pure clay and sintered at the maximal expansion temperature (1160 °C). Micropores also are not clearly detected for ECA, which are not sintered at the maximal expansion temperature (Figure 5b and 5c). In that case pore structure of ECA is not completely developed and pores, which become larger than 1 mm at the maximal expansion temperature, still is ranged from 10 to 1000 µm (pore size distribution was detected by Hg-porosimeter Pore Master 33 Quanta-chrome).

Mechanical properties mainly depend from the pore structure of aggregates. ECA (made by using 9.1% of NMP and sintered at temperature 1170 °C) with completely developed structure (Figure 5a) has three times higher compression strength (Table 2) compare with ECA, which have un-developed pore structure (Figures 5b, 5c).

Conclusively it was found that the amount of aluminium scrap recycling waste (non-metal product, NMP) and the sintering temperature were very important for the properties of lightweight expanded clay aggregates (ECA). The test results show that utilization of NMP as pore forming agent may influence the pores structure of ECA and reduce the production costs as well as promote possibilities to produce lightweight aggregates with different physical and mechanical properties. Since the clay is locally available material the production of ECA is perspective in the countries, where natural lightweight aggregates sources are unavailable.

3. Conclusions

The elimination of toxic nature of NMP was realized by heat treatment at the temperature higher than 1100 °C. The hazardous compounds of NMP are transforming in to new ones, non-hazardous compounds and NMP don not have toxic nature any more.

NMP without previous heat treatment should be use as raw material for production of new, environmental friendly building material in the case then the new product must be sintered at temperature higher than 1100 °C according to technology.

NMP is suitable for production of expanded clay aggregates (ECA) with density from 0.4 to 0.7 g/m^3. Density of ECA noticeable depends from two factors: amount of added aluminium recycling waste and sintering temperature.

ECA with different amount of NMP has unique optimal sintering temperature where the pore structure becomes completely developed.

Mechanical properties of ECA depend from the proportion of NMP and sintering temperature. Compression strength is larger for ECA with fully developed pore structure. The best compression strength may be reached by optimisation of technological process of expanding.

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