Applied Clay Science 155 (2018) 95–102

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Research paper

Influence of kaolin clay on mechanical properties and on the structure formation processes of white ceramics with inclusion of hazardous laundry sewage sludge

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ARTICLE INFO

Keywords:
Kaolin clay - laundry sludge composites
Hazardous waste
Structure formation
White ceramics
Mechanical properties enhancement
Environment efficiency

ABSTRACT

The present study deals with the development of new composites, which use kaolin clay for strong chemical bonding of heavy metals from industrial laundry water cleaning sludge (LS) after the washing of heavily soiled industrial uniforms. Compositions of these materials with up to 20 wt% content of LS were sintered at temperatures of 1100 - 1150 - 1200 - 1250 and 1300 °C. The raw materials and ceramics were analyzed by XRD, XRF, SEM, EDS and LAMMA methods. The flexural resistance of the compositions ranged from 2.80 to 12.52 MPa, linear shrinkage values varied from 4.52 to 14.06%, water absorption values diverged from 13.03 to 20.33% and density floated from 1.65 to 1.83 g/cm^3. Sintering at temperatures of 1150 °C and 1300 °C, with a partial melting of components, decomposed the Kaolin, Gibbsite, Dickite, Dolomite, and Almandine crystalline structures in the chemical environment of the compounds mixture and led to the synthesis of mainly amorphous vitreous substances with inclusions of crystalline structures such as Mullite, Cristobalite, and Alumina. The most important aspect of this research is the use of hazardous industrial waste as raw material for the consequent reduction the environmental burden caused by the indiscriminate disposal of these waste and thus abating the extraction of natural resources for the production of ceramic materials.

1. Introduction

The treatment of water for human consumption usually generates a large amount of waste in the form of sludge. According to Richter (2004), the main part of this waste is their discharging onto the soil as landfills. Mymrin et al. (2006, 2012) proved that any use of industrial waste as a raw material can solve the problem of the depletion of natural resources. McGrath and Cegarra (1992) came to the same solution after 30 years studying the process of increasing the degree of pollution of natural soils by Pb, Cu, Zn, Ni and Cd of sewage sludge of industrial laundry. Ceramic's composites (Dominguez and Ulmann, 1996) from wastes with high Zn, Pb, Cd, Ni, and Cr contents were tested for the formulation of a ceramic body and they met Argentina environmental regulations. Barredo-Damas et al. (2010) stated that some laundry sludge contained sand and grit, lint, free and emulsified oil and grease, heavy metals and volatile organic compounds. The incorporation of industrial wastes or by-products up to 30 wt% in bricks and tiles is becoming common (Segadaes et al., 2005). The study of some researchers (Acchar et al., 2009) reported the properties changes in ceramics containing 20 wt% of industrial sludge. Jordan et al. (2005) came to the conclusion, that the addition of sewage sludge decreased the bending strength. Herek et al. (2012) also fabricated ceramic bricks with different quantities of textile laundry sludge. They examined the properties of compressive strength, water absorption, solidification, and stabilization. Conversely, Ramirez et al. (2008) showed increased strength in concrete and mortar with the inclusion of waste sludge. Ceramic blocks containing 10% of sludge mixed with the clay showed better properties than blocks made only of clay.

But depending on the chemical composition of dirt can be used not only for the production of ceramics. Pietrobon et al. (2004) prepared sludge/cement test specimens containing until 30% of sludge; in general, the presence of sludge in the solid matrix of the sludge/cement paste negatively altered its structure.

Low-cost adsorbents were tested by Schuten et al. (2007) to remove anionic surfactants from the laundry rinsing water in order to allow the water reuse. Silva et al. (2016) used sewage sludge of industrial laundry by slow pyrolysis followed by physical activation with CO_2 for the preparation of mesoporous activated carbon.

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https://doi.org/10.1016/j.clay.2018.01.006
Received 12 September 2017; Received in revised form 10 January 2018; Accepted 11 January 2018

The mixtures of sewage sludge from different industrial processes in composites with clay were successfully adopted by Martínez-García et al. (2011). The authors studied the influence of each of them on the mechanics and structure of the developed ceramics.

The researcher of the Federal Technological University of Paraná, Brazil, has wide experience in this field. Mymrin (2012) have developed new compositions and technologies for about 80 types of industrial solids, sludge, and liquid rejects utilization for the production of market materials.

Therefore, the objectives of this research were: 1. Study of the possibility of using laundry water sewage sludge (LS) from heavily soiled industrial uniforms as a raw material combined with kaolin clay (KC) for the production of white ceramics whose mechanical properties meet the criteria established by Brazilian technical standards; 2. Study of the processes associated with the formation of the white ceramics structures during their sintering; 3. Development of new laboratory-level sustainable composites and technology in order to produce white ceramics using hazardous sewage sludge from industrial laundries.

2. Research methods and raw materials

2.1. Methods

The chemical composition determination of the raw materials was conducted by X-Rays Fluorescence method, using Spectrometer of Philips/Panalytical, model PW2400.

The mineralogical composition of the raw materials and ceramics was studied by powder method in a Philips X-Rays Diffractometer, model PW1830, at a 2Θ range of 2–70° with monochromatic wavelength λCu-Kα; the results were interpreted with Super-Q X'Pert High Score software (database PDF-2). To characterize the morphological structures, it was used scanning electron microscopy (SEM), model FEI Quanta 200 LV. The microchemical analyses were carried out by the energy dispersive spectroscopy method (EDS) in an Oxford equipment (Penta FET-125 Precision). The isotopic compositions of the new formation were performed by LAMMA-1000 laser micro-mass analyzer, model X-ACT. The comparison of the environmental impact of the LS and the developed ceramics was performed by solubility determination and metals lixiviation from liquid extracts (standard NBR 10004/2004), using the Atomic Absorption Spectroscopy (AAS) method in a Perkin Elmer 4100 spectrometer. The flexural resistance strength of the ceramics was controlled in the universal testing machine EMIC DL10,000. Linear shrinkage was determined with a digital caliper of DIGIMESS and water absorption (WA) - after ceramics complete immersion in water for 24 h. The particles size distribution was executed by laser diffraction analysis on a CILAS 1064 Granulometer.

2.2. Raw materials characterization

A sample of LS was obtained from an industrial laundry specialized in the washing of heavily soiled industrial uniforms in the city of Curitiba, Brazil. The KC sample was obtained from the company Farmanil Quima, Curitiba. The LS sample was black and oily, with a dense bitumen-like appearance. This comparison was confirmed by its high gross calorific value - 5284 Kal/kg. The sludge had a moisture and oily substances content of 71.8%.

2.2.1. Chemical composition of the raw materials

In order to study the chemical composition (Table 1) of the sludge, it was burned to 800 °C and the ash (28.2% of weight) was analyzed by XRF method. The chemical composition of the raw materials indicated a higher concentration of SiO_2 (71.50%), Al_2O_3 (12.74%) and Fe_2O_3 (10.34%) in the LS, with small inclusions of other elements. The very high ignition loss (I.L.) (71.8%) was probably due to grease, oil and all types of oily components emulsified, resin, tar, paints, organic volatile compounds, carbonates, etc.

Table 1
Chemical composition of the raw materials (by XRF method).

	Fe_2O_3	CaO	MgO	SiO_2	Al_2O_3	K_2O	Ba O	P_2O_5	Total	I.L.
LS	10.34	2.51	0	71.50	12.74	1.85	0.74	0.32	100.00	28.76
KC	0.3	8.68	6.17	45.13	38.58	0.94	0	1.41	100.00	14.98

Note: I.L. – Ignition Loss.

Table 2
Results of leaching and solubility tests of laundry sludge (LS) in comparison with ceramics 7 (Table 2) after sintering at 1150 °C.

Elements	Leaching, mg/L			Elements	Solubility, mg/L		
	LS	Comp. 7	NBR 10004		LS	Comp. 7	NBR 10004
As	9.92	0.15	1.0	As	12.44	< 0.01	0.01
Ba	88.28	4.17	70.0	Ba	97.15	< 0.1	0.7
Cd	9.15	0.018	0.5	Cd	17.11	< 0.005	0.005
Pb	6.32	0.21	1.0	Pb	8.56	< 0.01	0.01
Cr	22.15	1.04	5.0	Cr	25.72	< 0.01	0.05
Hg	2.04	< 0.001	0.1	Hg	3.84	< 0.001	0.001
Se	2.75	< 0.01	1.0	Se	3.74	< 0.01	0.01
Al	20.13	< 0.10	[a]	Al	29.28	0.01	0.2
Cu	23.52	< 0.05	[a]	Cu	33.44	< 0.05	2.0
Fe	39.63	0.07	[a]	Fe	53.87	0.15	0.3
Mn	74.18	< 0.10	[a]	Mn	92.74	0.03	0.1
Zn	78.18	< 0.10	[a]	Zn	92.73	0.10	5.0

[a] No demands yet in the Brazilian sanitary norms NBR 10004.

KC also consisted (Table 1) predominantly of SiO_2 (45.13%) and Al_2O_3 (38.58%) with an extremely low inclusion of Fe_2O_3 - 0.3%, therefore, it was used as a raw material for the production of white ceramics. A rather high value (14.98%) of the ignition loss (I.L.) was probably due to the decomposition of clay minerals, dolomite and the combustion of organic matter.

2.2.2. Environmental impact of the laundry sludge

The comparison of the results of the LS leaching and solubility tests with the Brazilian sanitary standards (NBR 10004) demonstrated higher contents of hazardous elements in these raw materials (Table 2). The value of As was almost 10 times higher than the allowed level by these norms. According to the same standards, the levels of Cd, Pb, and Hg were also 18.3, 6.3, and 20.4 times higher, respectively, etc. The determination of the solubility of LS showed a much more dangerous discrepancy with the standards: As – in 1244 times, Ba – almost in 139 times, Cd – in 3422 times, Pb – in 856 times, Hg – 3840 times, etc. These hazardous washing sludges cannot be managed in dumps or run into municipal sewage water as it is currently done. Their complete neutralization can only be accomplished through chemical interaction, with selected suitable components, in high temperature or vitrification processes.

All metals are out of the permitted levels: As – in 9.92 times, Pb – in 6.32 times, Cr – 4.4 times, etc. Therefore, LS is included in the list of hazardous wastes and should not be located in industrial evictions in accordance with the Brazilian sanitary regulations (NBR 10004).

2.2.3. Mineralogical composition and structure of the raw materials

The X-ray diffractogram of the KC (Fig. 1A) revealed the following mineralogical composition: Kaolinite $Al_4(Si_4O_{10})(OH)_8$, Gibbsite $Al(OH)_3$, Dickite $Al_4(Si_4O_{10})(OH)_8$, and Dolomite $CaMg(CO_3)_2$ with rather high contents of amorphous materials.

Deciphering the DTA and TG curves (Fig. 1B) of the sample consisting of four minerals was always ambiguous due to the overlap of endo and exothermic effects of these three minerals, masking them to one another in the final curve. The proximity of the thermal effects of three minerals containing aluminum - Kaolinite, Gibbsite, Dickite, and