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# Effect of the incorporation of residual sludge from water treatment on the technological properties of ceramic bodies: A review

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**Abstract.** The disposal of sludge generated by water treatment by chemical coagulation is considered an environmental problem that has attracted attention of researchers worldwide, whose objective is to promote strategies for their productive use. This study addresses results of studies conducted in the recent 7 years on the aluminous sludge use in the manufacture of ceramic products. The literature shows that the proportion of sludge and the firing temperature are two key factors that affect the final ceramic products technological properties: water absorption, mechanical resistance, porosity and bulk density. It is concluded that it is feasible to incorporate water treatment sludges in partial replacement of one of the constituent materials of clay bodies, with additions up to 10% sludge and firing temperatures above 1000 °C.

## 1. Introduction

The application, capacity and treatments that may need to be installed for water treatment depend on the reduction achievable by treating the concentration of chemical substances present in the water. Water treatment processes can be classified from lower to higher technical complexity and costs in the following way. [1,2]: simple chlorination, simple filtration, precloration and filtration, aeration, chemical coagulation [3], absorption treatment with granular activated carbon [4], ion exchange [5], ozonation, advanced oxidation processes [6] and membrane treatment [7]. Scientific literature review shows that conventional methods for waste management of water treatment or combination of several of them, are not efficient in the reduction of potentially toxic heavy metals, nitrogen, phosphorus, etc., from raw water and coagulant agents [8], being classified as a potential environmental pollutant, since in general, this sludge is discharged directly, after dehydration, into nearby water bodies or landfills [9-11].

The existence of a greater number of treatment plants, with which by 2015 access to a source of drinking water has been given to 91% of the world population [12], generate more and more volumes of residual sludge, with a load of pathogenic microorganisms, which due to their disposition constitute a risk to health, whether through ingestion, inhalation or contact [13]. This reinforces the need to search for more efficient processes for residual sludge treatment, disposal and reuse, at worldwide scale. In addition to this scenario, some industries according to the needs of their product, submit the water that enters their plant to purification treatments, according to specific characteristics requirements [14,15], having primary attention in the productive process and not in the use efficiency of this resource [16].



This tendency to search for more sustainable processes after the adoption of ecological practices in the supply chain has spread to different productive sectors, representing not so much a positive relationship between their environmental performance, as in the financial one [17]. A worldwide striking sector for the implementation of sustainable practices is the construction sector, which according to the United Nations Environment Program, is responsible for about 40% of global energy consumption, more than 30% of emissions of greenhouse gases (GHG), 30% of raw material extraction, 25% of solid waste, 25% of water use and 12% of land use [18]. For the millionaire amounts in weight and ceramic constructive units manufactured every year in the world, studies have focused on the development of sustainable construction solutions to minimize the aforementioned impacts, such as interventions to reduce GHG emissions, optimization of the processes of manufacturing and new products, which either by their geometries or by the use of certain residues as clay replacement contribute to reduce the consumption of natural resources, the energy consumption of the buildings, and their equivalent CO<sub>2</sub> footprint [19]; however, despite the good technological results obtained, these have not been scaled up to mass production [20].

The objective of the present work was to carry out a review on the existing processes for the management of the residual sludge of drinking water treatment plants, its composition and studies of its possible application in manufacture processes of ceramic construction materials with a view to reducing its environmental impact, and individually analyzing its most relevant technological properties: plasticity, linear shrinkage, water absorption, apparent density and mechanical strength, to define the feasibility of using this type of waste in the manufacture of ceramic materials.

## 2. Obtaining and disposing of sludge by chemical coagulation of waters

The water cycle at urban level includes extraction of surface water from natural water sources, treatment to meet the minimum quality standards required for different uses, distribution, consumption, collection, transport and treatment of wastewater before returning them to natural ecosystems [21]. In addition to pathogenic microorganisms, natural waters contain three types of non-sedimentable contaminating solids, which according to their size and condition are classified as: suspended, colloidal and dissolved solids [22-24]. In order to purify these contaminants from the water, it is necessary to add an electrolyte that immediately forms complex hydroxides while neutralizing the electric charge of the colloid; this neutralization, known as chemical coagulation, is usually done by applying coagulants to water, mostly iron or aluminum salts; aluminum sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) is the most used in water treatment plants, due to its stability, solubility, efficiency, low cost and easy access [25,26]. During the flocculation process, trivalent aluminum or iron cations neutralize the negative electric charges of the colloidal particles, forming aggregates of greater sedimentation rate, called flocs [23,25,27,28].

Obtained floc, with a humidity generally superior to 80% weight, is constituted by the precipitate of hydroxide, sand, silt, clay, organic substances of the water and undesirable components, like pathogenic microorganisms [9,10]. The treated water is removed through subsequent processes such as sedimentation or flotation, rapid filtration by gravity or pressure, or a combination of methods [2,29], being an effective method to eliminate colloidal and suspended particles, but not enough to eliminate the dissolved components [11]. The most used methods for an adequate final disposal of dry sludge, include incineration and their use in the production of construction materials and soil improvement [30].

### 2.1. Chemical and mineralogical characterization

Chemical composition of the residual sludge from drinking water treatment plants evaluated in previous studies is shown in Table 1. The variation in the concentrations of each compound is conditioned by the origin of the water, the treatment process, and the analysis method, reason why they cannot be directly compared, since they have been obtained in different non-standardized conditions, and the statistical error analysis is generally not available [29,31]. In general, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> make up most of the sludge, with other oxides such as CaO, MgO, Na<sub>2</sub>O, K<sub>2</sub>O, P<sub>2</sub>O<sub>5</sub> and TiO<sub>2</sub> being found in a small percentage. The amount of Al<sub>2</sub>O<sub>3</sub> or Fe<sub>2</sub>O<sub>3</sub> in the mud is associated with the applied coagulant (Al or Fe salts), and the concentration of these metals in the untreated water.

Mineralogical composition of sludge from water treatment plants at different temperatures indicates the majority presence of phases of quartz (50%), illite, calcite (21%) and albite (30%); This last phase is an aluminosilicate  $\text{Na}(\text{AlSi}_3\text{O}_8)$ , a type of sodium feldspar [32,33], authors also report the presence of calcite [34]; a chemical composition similar to clay, so it is considered as a potential additive in the manufacture of clay products. This is reported in the literature, in which these sludges have been reused in the manufacture of light aggregates [35], in bricks and ceramics production [36,37], as raw material for concrete and mortar [38,39], in cement production [40], in agricultural practice and other uses based on the land [29,41,42], on the recovery and reuse of coagulants [43,44], as an adsorbent of heavy metals such as phosphorus, hydrogen sulfide, boron, fluorides, perchlorate, glyphosate, mercury, arsenate, lead and selenium [32,45,46], and as a substrate in wetlands Noises [47].

**Table 1.** Chemical analysis of sludge (%).

Concentration (5)										Reference
SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	TiO <sub>2</sub>	PPF	
54.10	28.84	9.92	3.10	0.64	0.30	0.75	-	1.28	-	[40]
10.90	1.34	68.65	8.23	0.61	-	-	0.39	-	-	[36]
54.80-	19.60-	4.91-	0.72-	1.06-	0.08-	0.78-	-	-	3.77-	[35]
66.90	23.00	11.30	4.26	4.15	10.99	2.08	-	-	13.00	[48]
29.60	28.02	8.05	1.48	0.38	-	0.84	-	0.85	22.60	[37]
37.50	30.10	12.30	0.20	0.40	0.20	0.90	-	1.00	17.10	[49]
27.12	62.66	1.16	1.25	0.37	0.24	0.83	0.19	0.16	5.11	[10]
52.78	14.38	5.20	4.39	3.08	0.97	3.62	0.17	0.61	-	[50,51]
40.33	31.84	6.43	0.09	0.48	0.04	1.32	0.20	0.46	18.50	[52]
46.37	20.33	8.55	11.15	2.19	0.36	3.25	5.89	0.85	0.05	[32]
36.51	22.21	5.65	2.66	1.34	1.35	0.49	-	-	28.10	[53]
40.60	41.31	8.56	1.55	0.86	0.18	2.15	2.76	-	26.90	[54]
29.59	31.18	10.21	0.34	-	-	1.27	-	1.04	24.50	[33]
59.70	10.52	4.38	6.01	2.20	1.53	1.16	-	-	11.10	[55]
17.01	24.46	13.00	0.30	0.15	0.02	0.18	0.44	0.40	39.77	

### 3. Sustainable use of sludge in the ceramic materials field

Martínez, Eliche, Pérez, Iglesias, and Corpas [52] evaluated the effect of the incorporation of sludge from wastewater treatment on the properties and microstructure of clay used for the manufacture of bricks, replacing clay with additions of sludge in 1%, 2.5%, 5%, 7.5%, 10% and 15% by weight. The clay bodies were molded by pressing and fired at 950°C showed that, with additions up to 5% in weight, mechanical properties of the product remain in acceptable ranges. Similar results were reported by Torres, Hernández and Paredes [56], who conclude that sand replacement by sludge at 10% in weight is suitable in the manufacture of ceramic bricks at a firing temperature of 900 °C, and additions in a higher percentage significantly compromises water absorption and compressive strength.

Anyakora [48] showed that sludge collected from a drinking water treatment plant could be used as a dye and clay replacement in the manufacture of bricks for various engineering and construction applications. Five different proportions of clay mixtures were studied with additions of sludge at 0%, 5%, 10%, 15% and 20% of the total weight of the mixtures, the molded bodies were sintered at 850 °C, 900 °C, 950 °C, 1000 °C and 1050 °C. The results of the tests indicated that the firing temperature and the sludge ratio are two key factors that determine the quality of the bricks; as the sludge content increases, it results in lower resistance to compression, lower density, and greater water absorption, in addition to improving the physical appearance (color) of the bricks.

Kizinievic, Zurauskiene, Kizinievic, and Zurauskas [36], used a freshwater treatment sludge (WTS) composed mainly of Fe<sub>2</sub>O<sub>3</sub>, with a particle size up to 138 μm, incorporating it into a ceramic paste between 5% to 40% by weight; the molded and fired bodies at 1000 and 1050 °C showed that the physical and mechanical properties deteriorate as larger amounts of WTS are added; with additions of 5% the ceramic density increases 5% -14% and the compressive strength by 36% - 97%, water absorption decreases 29% - 61%, and open porosity by 19% - 35%, while with additions of 40% WTS,

ceramic density decreases 10% - 22%, and compressive strength of 34% - 43%, water absorption of 70% to 100%, and open porosity 45% - 50%, compared to a reference mix without WTS.

With the same objective, Benlalla, Elmoussaouiti, Dahhou, and Assafi [49], prepared clay mixtures with additions between 5 and 30% weight of aluminous sludge from a water treatment plant. Extruded mixtures were fired at 800 °C, 900 °C, and 1000 °C. The technological properties of the sintered bodies showed that to produce a dense brick with high mechanical strength, the mixtures must contain less than 20% in sludge, and be sintered at temperatures between 900 °C - 930 °C. Results very similar to those obtained by Tantawy and Ramadan [33], who suggest additions of 15% - 30% by weight of these sludges for the production of dense clay bricks, with firing temperatures between 700 °C and 1000 °C. Likewise, Barón, Montaña and González [57], conclude that replacement of clay by 10% - 20% of aluminous sludge guarantees physical and mechanical properties for the manufacture of non-structural bricks, with a firing temperature of 1100 °C.

Ling and others [50], reformulated the basic composition of pottery clay with freshwater treatment sludge, observing that the mixture loses plasticity with addition of sludge higher than 20%, contraction of the bodies was uniform, and it was possible to improve the mechanical properties of the fired product, e.g. hardness increased 21.6%. The authors conclude that because of the minimum shrinkage, and the improved hardness, sludge from water treatment is an additive potential in red clay mixtures [51].

Amin, *et al.* [34] studied the possibility of using silver mud from municipal wastewater treatment in the manufacture of ceramic paving tiles, adding dry mud in proportions from 5% to 35% to a mixture of standard tiles; mixtures pressed uniaxially at 30 MPa, and fired at 1050-1150°C allow obtaining tiles of type BII<sub>b</sub> (water absorption < 10%) with mud additions up to 7% and firing temperature of 1150 °C, and tiles BIII (water absorption > 10%) with sludge additions of 5% - 10% sintered at 1150-1100 °C, with acceptable physical and mechanical properties according to the UNE-EN 14411: 2016 standard.

Mymrin, *et al.* [55] used municipal sewage sludge as the main component to produce ceramics with combinations of acid neutralization salts, glass debris, sand and clay. The ceramic bodies obtained at 900 °C, 950 °C, 1000 °C, 1050 °C, and 1100 °C have a flexural strength up to 2.5 times higher than ceramics without additions, as the firing temperature is increased above 950 °C; the water absorption decreases while the density increases for all designed mixtures.

The highest sludge addition found in the literature was reported by Kizinievič, Kizinievič, Boris, Girskas, and Malaiškienė [58], who after replacing 40% - 60% clay with sludge, composed mainly of Fe<sub>2</sub>O<sub>3</sub>, conclude that with higher replacing percentage and lower firing temperature, the obtained product has lower density, greater water absorption and porosity, and a reddish hue. In a later study, Kizinievič and Kizinievič [59] investigated the properties of clay bricks with additions of 5% - 60% of the same sludge fired at 1000 °C, finding that with less than 20% addition, this residue is an additive that allows to obtain light ceramics pieces, with physical and mechanical properties within the normative parameters.

#### 4. Discussion

Different scientific studies show that residual sludge of water treatment, especially municipal water, contains numerous dangerous pathogenic organisms and toxic metals, among other substances that represent an extremely high ecological risk, which, depending on their disposition, represent a direct risk for people. Due to the increasing amount of sludge generated by wastewater treatment plants, there has been a strong demand for reuse in different industries. Throughout the body of the article, the results of the use of these sludges in the manufacture of ceramic materials are presented, as well as the effects on the ceramic piece properties.

##### 4.1. Effect of sludge addition on mixtures plasticity

The plasticity in clay mixtures, is a fundamental property that allows to project the changes so that a clay body with added water will suffer when subjected to an external force, and if this maintains the form after the force is removed or reduces [60]. The plasticity of the mixtures studied in different proportions of the system shows that there is an almost linear increase in plasticity with added sludge

percentage [34,48,49]. In the study by Amin et.al. [34] the plasticity of the mix almost doubled when the sludge percentage increased from 0% to 35%. This can be understood considering the dual effect of finer particle size and organic matter on plasticity [61]. Some studies reveal a contrary behavior, in which the addition of sludge decreases the plasticity of the mixture; this can be understood by considering that sludge is composed of silt particles with coarser grains and poor water absorption capacity [50,62]; however, the literature reports that this index can be controlled by increasing the grinding time [51].

#### 4.2. Characteristics of fired tiles samples

*4.2.1. Effect of sludge addition on linear shrinkage.* The quality of the ceramic products can be guaranteed among other things by its degree of linear shrinkage. Bricks generally show a contraction of less than 8% [52,49].

The literature reveals a great variety of results that do not allow for a clear trend in the relationship of this property and the percentage of sludge applied; some studies report that at higher proportions of sludge addition, contraction percentage increases, obtaining values suitable for additions equal to or less than 5% [52] while other authors [36] report that with sludge additions of 5% and above the ceramic body contracts above the suggested 8%; however, in the investigations carried out by authors such as Benlalla, Elmoussaouiti, Dahhou, and Assafi [49], the addition of sludge up to 30% allows obtaining contractions below 3%, and Tantawy and Ramadan [33] obtain contractions below 8% with up to 60% sludge additions. These last two studies also reveal that the higher the burning temperature, the greater the contraction of the ceramic body; giving that when the vitrification temperature is lowered, sintering in the liquid phase of the feldspathic materials present in the mixtures improves [63]. The variation in the contraction percentages of the mixtures is mainly due to chemical and mineralogical composition of the mixtures. The presence of quartz and kaolinite in the sludges, gives this material a low capacity of contraction-swelling, since these minerals have little affinity for water [64,65].

*4.2.2. Effect of sludge addition on water absorption.* Like linear shrinkage, water absorption is a main property to consider when characterizing ceramic products, especially ceramic tiles, because the less water infiltrates the ceramic piece, the more durability and resistance is expected. Literature reveal that higher than 10% sludge addition causes an increase in water absorption of the clay body, fired above 850 °C [33,36], indicating that sludge content produces a more porous material, which leads to a lower mechanical resistance [66]. Anyakora [48] and Benlalla, Elmoussaouiti, Dahhou, and Assafi [49] obtained similar results in their study, in which they also point out that at higher firing temperatures the water absorption decreases, being able to obtain bricks with less than 15% water absorption, with sludge additions up to 10% by weight at 1000 °C firing temperature. As the firing temperature increases, clay minerals dehydrate to become vitreous minerals that close some of the open pores [67]. A similar behavior is observed in triaxial mixtures for the manufacture of tiles, of which it is reported that to obtain a water absorption lower than 10%, it is necessary to add no more than 15% of sludge and firing them at 1150 °C [34].

*4.2.3. Effect of mud addition on apparent porosity.* This property is not a standard requirement of ceramic products, although it is indicative of the percentage of open pores and, therefore, is strongly related to the water absorption. As can be anticipated by the results presented in the previous section, the addition of sludge causes an increase in the proportion of open pores, especially at temperatures below 850 °C - 900 °C, temperature at which the combustion of organic matter occurs, without forming the vitreous phase of the mineralogical components. Significant changes in this property occur in bodies with additions of more than 10% by weight of sludge [59]. Apparent porosity has an inverse relationship with density; while the apparent porosity decreases, the apparent density increases with the firing temperature, this because pores created by the mixture decomposition are closed by the formation of the vitreous phase in clay minerals, as firing temperature increases [68].

*4.2.4. Effect of sludge addition on mechanical strength.* The mechanical resistance is perhaps the most important parameter of the ceramic body, since with this its engineering quality is determined for applications such as construction materials; for ceramic tiles it must be formulated as two values: breaking strength and breaking modulus; for bricks it is determined as resistance to compression. According to what is observed in the literature, the resistance depends to a large extent on the quantity of sludge in the mixture and the firing temperature. Such is the case of the values of breaking strength and rupture modulus for tiles reported by Amin et.al (2017) in which it is evident that mixtures with additions of sludge lower than 7% and sintered at 1150 °C complies with the minimum resistance break (500 N) for tile thickness <7.5 mm and water absorption <10%. Similar proportions are reported [36,49] to ensure optimum compression resistance in bricks, where additions of less than 10% allow even more resistance compared to a mixture without sludge, achieving better results at temperatures above 1000 °C; being higher than the recommended minimum resistance of 100 kg/cm<sup>2</sup> [69] with additions of up to 20% in sludge.

## 5. Conclusions

One of the residues investigated for use in the ceramic network is the residual sludge from the raw and/or municipal water treatment plants, whose simple method of final disposal involves the contamination of water bodies and soil; this is the reason why the development of strategies for its reuse has aroused great interest. This study presents the review of the most recent options that have been identified worldwide for the use of these sludges in the manufacture of ceramic products, specifically tiles and bricks.

It was found that these sludges contain considerable amounts of Fe<sub>2</sub>O<sub>3</sub>, so it can be used as a natural pigment, dyeing the ceramic body in a darker and intense red color.

This work showed that the ceramic sector is a potential receiver of sludge from raw and residual water treatment plants. The proportion of sludge in the mixture for the manufacture of ceramic products and the burning temperature have been demonstrated as the two key factors that affect the technological properties of the final ceramic products. At higher proportions of added sludge, the water absorption clearly increases, the compressive strength is reduced for bricks, and the flexural strength and rupture modulus is reduced for tiles as well, due to the greater porosity caused by combustion of organic matter, which also implies the decrease in apparent density.

In general, although given that the increase in the incorporation of mud generates a growing trend in terms of water absorption and decreasing in terms of mechanical resistance, the percentage of more suitable mud that could be incorporated for the manufacture of the bricks and/or ceramic tile would be 10% at burning temperatures above 1000 °C.

The use of sludge in the manufacture of bricks and ceramic tiles is an alternative to eliminate an environmental problem, that although it is the objective of many investigations, with which they have achieved viable results, the real production at an industrial level is still very limited. It is proposed that for a wide production and application of ceramic products from industrial waste, more research and development is needed, not only in the technical, economic and environmental aspects, but also in standardization, government policies and the public education related to waste recycling and sustainable development.

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