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Waste Management xxx (2017) xxx-xxx

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Sustainable use of tannery sludge in brick manufacturing in Bangladesh

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ARTICLE INFO

Article history: Received 20 July 2016 Revised 3 December 2016 Accepted 29 December 2016 Available online xxxx

Keywords: Tannery sludge Heavy metals TCLP Bricks Waste management Recycling

ABSTRACT

Chromium-rich tannery sludge generated from tanneries has the potential to become a serious environmental burden in Bangladesh and a promising avenue for disposal of this sludge is by stabilizing it in clay brick products. But for sustainable industrial application of such technique it needs to be ensured first that the engineering properties of bricks as a building material are not diminished by addition of sludge, the process becomes energy efficient compared to alternatives and the use of such bricks do not pose any harmful environmental effects in the long run. In this study, clay bricks were prepared with different proportions of sludge (10%, 20%, 30% and 40% by dry weight) in both laboratory-controlled and field conditions and their suitability as a construction material was assessed based on their strength, water absorption, shrinkage, weight-loss on ignition and bulk density. For the sludge incorporated bricks, the compressive strength ranged from 10.98 MPa to 29.61 MPa and water absorption ranged from 7.2% to 20.9%, which in most cases met both the Bangladesh and ASTM criteria for bricks as a construction material. Volumetric shrinkage, weight loss and efflorescence properties of sludge-amended bricks were found to be favorable and it was estimated that an energy saving of 15-47% could potentially be achieved during firing with 10-40% tannery sludge-amended bricks. The quality of sludge-amended bricks made in the brick kiln was relatively inferior compared to bricks produced in the laboratory due to operating in a less-controlled environment with respect to maintaining adequate compaction and optimum moisture content. The leaching behavior of several heavy metals (Cr, As, Cu, Ni, Cd, Pb and Zn) from sludgeamended bricks has been found to be insignificant and far below the Dutch regulations and USEPA regulatory limits. Results from this study indicate that tannery sludge can be sustainably stabilized in clay bricks and large-scale application of this technique can be envisaged in the context of Bangladesh where brick remains a dominant building material.

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1. Introduction

Chromium-rich tannery sludge (TS) generated from the effluent treatment plants of leather industry has the potential to contaminate soil, surface water and groundwater and pose a threat to the environment and natural resources if it is not disposed properly (Thomson et al., 1999). Usually, about 100–150 kg of sludge is generated per ton of hides/skins processed (UNIDO, 1998) which is composed mostly of chemically precipitated dissolved chromium, different types of spent chemicals, sulfide, salt, proteins, polyphenolic compounds, surfactants, dyes and syntans in the treatment process (Swarnalatha et al., 2006; Chang et al., 2001). Tannery sludge (TS) contains elevated concentrations of heavy metals like Cr, As, Ni, Co, Cu, Zn, Fe, Cd due to use of basic chromium salt,

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http://dx.doi.org/10.1016/j.wasman.2016.12.041 0956-053X/© 2016 Elsevier Ltd. All rights reserved. different syntans, dyes, pigments, retanning agents etc. in the tanning process (Houshyar et al., 2012; Kiliç et al., 2011). These heavy metals are very harmful, because of their non-biodegradable nature, long biological half-lives and their potential to accumulate in biological systems (Wilson and Pyatt, 2007; Singh et al., 2004). Immobilization of heavy metals is the primary objective for stabilization of a hazardous waste such as tannery sludge.

A technique to treat or stabilize hazardous waste is by valorisation in construction materials such as brick or concrete which has been applied in several instances quite successfully for the cases of sewage and textile sludge and arsenic-rich filter materials (Patel and Pandey, 2012; Cusidó and Cremades, 2012; Praveen et al., 2015; Chiang et al., 2009; Hassan et al., 2014). Though this form of sludge utilization in such materials does not necessarily involve a chemical interaction between the waste product and the materials, it has proven to be an effective method to reduce the potential hazard of the waste by converting it into a less toxic and mobile

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form though the performance can vary depending on the type of waste product used, its particle size distribution and the technique for stabilization. The stabilized waste can be used as a construction material provided that the material possesses the necessary engineering properties and leaches toxic pollutants to an acceptable degree (Hassan et al., 2014; Rouf and Hossain, 2003). Several studies have shown that tannery sludge can be effectively stabilized in construction materials such as concrete, ceramic tiles and other engineering materials (Montañés et al., 2014; Basegio et al., 2002; Giugliano and Paggi, 1985) but not all these processes have been developed at a level suitable for industrial applications and treatment processes practiced are still potentially hazardous for the environment (Alibardi and Cossu, 2016). In order to offer tannery sludge as a viable recycling option in construction industry, the properties of the sludge-stabilized construction material must conform to local building material standards. In addition to that, the material must pose no long-term harm to the environment in terms of leaching of heavy metals. Therefore, although proof-ofconcept study of the stabilization of various waste products in building materials are available (Mohajerani et al., 2016; Ukwatta et al., 2015; Quesada et al., 2015; Chiang et al., 2009), whether or not these techniques can produce bricks of desired quality adhering to standards for industrial applications are somewhat unexplored. The aesthetic quality of the building material, energy consumption during production and the possibility of leaching harmful constituents for prolonged period of usage are also important considerations to make the product sustainable. In this paper, we primarily address these issues in the context of Bangladesh while attempting to stabilize tannery sludge in clay bricks, a dominant construction material in the country.

In Bangladesh, 85,000 tons of wet salted hides and skins are processed annually (Paul et al., 2013) and it is estimated that 19,000 tons of partially dried sludge will be generated by the effluent treatment plants if the treatment systems of all tanneries become operational. On the other hand, 45,000 brick kilns in Bangladesh together produce about 17.2 billion bricks per year with an estimated sale value of around US\$1.2 billion which is almost 1% of Bangladesh's GDP (World Bank, 2011). Due to unavailability of stone aggregate, brick has become the principal building material for the country's construction industry and will continue to be so in future. Therefore, exploring the brick manufacturing industry as a potential avenue for stabilization of the huge amount of tannery sludge in Bangladesh can be a viable option for commercial application.

In this study, we determined the characteristics of sludgeincorporated clay bricks with respect to its engineering properties as well as its environmental implications. Clay brick specimens were prepared with different proportions of sludge in both laboratory-controlled and field conditions (i.e. in a brick kiln) and their suitability as an engineering material was assessed based on their strength, bulk density, weight loss on ignition, shrinkage, water absorption and firing energy characteristics. Leaching test of sludge-incorporated bricks was also carried out to demonstrate the effectiveness of the stabilization technique against the release of heavy metals in the environment.

2. Experimental

2.1. Raw materials

Tannery Sludge (TS) samples were collected from the Effluent Treatment Plant of Apex Tannery Ltd, unit-2, Gazipur, Bangladesh (Fig. 1(a)). This tannery employs a conventional combination of chemical and biological treatment following rapid sand filtration and uses lime and ferrous sulfate as coagulant. The clay sample used to prepare bricks in the laboratory was obtained from a local brick manufacturing plant.

2.2. Characterization of sludge

The moisture content and organic content of sludge and clay were determined by APHA method (APHA, 2012). X-ray fluorescence (XRF-1800, SHIMADZU) was used to determine the chemical composition of tannery sludge. For heavy metal analysis, 5 g lightly ground dried sample was digested with acid (HNO₃: HCl = 1:3 vol. ratio) for 24 h, then 350–400 ml distilled water was added and the sample was boiled for 2.5 h to prepare a 500 ml solution. Finally, the solution was filtered through a 0.45 μ m filter paper and the filtrate was collected to determine the concentration of heavy metals (Cr, As, Pb, Cd, Ni, Cu, and Zn) by using Atomic Absorption Spectrophotometer (AAS) (Shimadzu AA 6800) (Juel et al., 2016; Saha and Hossain, 2011).

2.3. Brick preparation and tests

Both sludge and clay were oven dried and ground by a crushing machine. Atterberg limit tests were conducted according to ASTM (ASTM D 4318, 2000) to determine the plastic nature of the sludgeclay mixture. Optimum Moisture Content (OMC) was determined according to AASHTO (AASHTO T- 99, 1982).

Total 75 brick samples (length 12 cm. width 6 cm and height 3.5 cm) of sludge-clay mixture in varying proportions (0%, 10%, 20%, 30% and 40% by dry weight) at OMC were prepared in the laboratory (Fig. 1(b)). Additional 2–3% water above the OMC was added (based on the dry mass of each brick) to all the mix ratios to facilitate the hand molding process while mixing. Three 100% clay samples were prepared as a reference specimen. After 24 h of natural drying and 48 h of oven-drying (at 105 °C), these samples were heated in an electric furnace (Nabertherm, LH 60/14, Germany) at the rate of 5 °C/min up to experimental temperatures of 900 °C, 950 °C and 1000 °C and were held for 3 h (Fig. 1(d)). The objective of experimenting with brick with varying sludge proportion was to determine the effect of sludge quantity on various physical and mechanical properties of bricks and also to roughly find out the threshold sludge quantity that would allow the bricks to have desirable engineering properties as a building material.

A series of tests such as firing shrinkage, weight loss on ignition, water absorption and compressive strength were conducted on the bricks according to ASTM (ASTM C 67-02c, 2002). The engineering properties of the bricks were compared with the ASTM and Bangladesh Standards (see Table 1)(BDS 208, 2009). Efflorescence test was carried out following BDS 208 (2009) (procedure described in Supplementary materials). Leachability test of all brick samples for specified heavy metals was carried out in accordance with USEPA 1311 (USEPA, 1992) and Netherland Tank Leaching test NEN 7345 (NEN 7345, 1993). The test results of all the parameters were taken as an average value from three replicate tests.

A separate set of sludge-incorporated bricks was prepared in a commercial brick kiln ('field condition') following their typical protocols (Fig. 1(c)). Two types of brick molding procedures are primarily adopted in Bangladesh – hand molding and automatically pressed molding. In this study hand molding method was used for brick production. For the bricks prepared in 'field condition', the sludge content is chosen as 10% (by weight). The bricks prepared in 'field condition' underwent the same set of tests for mechanical properties and leaching as described above.

2.4. Firing energy saving

Brick production industries are considered to be one of the highest energy consuming sectors and have a large negative impact on the environment related to energy use (Koroneos and Dompros, 2007). So savings in energy consumption by incorporating tannery sludge may lead to sustainable brick production. The

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(c)

(d)

Fig. 1. (a) Raw tannery sludge in the sludge-drying bed of Apex Tanneries Ltd. at Gazipur (b) Raw TS bricks prepared in the laboratory (c) Raw TS bricks prepared in the brick kiln (Field condition) and (d) TS bricks fired at 950 °C and 1000 °C. In Fig. 1(d) the markings 0, 1, 2, 3, 4 indicate 0%, 10%, 20%, 30% and 40% sludge amended bricks and M & N indicate firing temperature of 1000 °C and 950 °C respectively.

Table 1

BDS 208 (2009) and ASTM C216-10 criteria for bricks for specific use.

Name of test	BDS 208 (2009)	BDS 208 (2009)			
	Grade S	Grade A	Grade B	Grade SW	Grade MW
Compressive strength (MPa) Water absorption (%)	≥24 ≼10	≥15 ≼15	≥10.3 ≼20	17.2 20	15.2 25

Note: Grade S: This type of bricks may be used for breaking into aggregate for plain and reinforced concrete and for making base course of pavement. Grade A: This type of bricks may be used in construction of buildings of long duration. Grade B: This type of bricks may be used for one storied building, temporary shed, where intended durability is not very long; Grade SW: Resistance to severe weathering. Grade MW: Resistance to moderate weathering.

firing energy saved due to incorporating sludge can be calculated using Eq. (1) derived by Mohajerani et al. (2016).

Energy saved,
$$\Delta E = \frac{q.m_1 - (q.m_2 - CV.m_3)}{q.m_1} \times 100\%$$
 (1)

where q = Specific energy for brick firing (firing energy per unit mass of brick), MJ kg⁻¹, m_1 = mass of clay in control brick (kg), m_2 = mass of TS brick (kg), m_3 = mass of sludge in TS brick (kg), CV = calorific value of tannery sludge MJ kg⁻¹.

In this study, the specific firing energy was assumed to be 5 MJ kg^{-1} which is within typical values reported by Whittemore (1994) and Prasetsan (1995).The calorific value of tannery sludge was measured using Bomb calorimeter (Parr 6100).

2.5. Leaching test of building materials

The toxicity and leachability tests were carried out using two different methods: toxicity characteristic leaching procedure (TCLP) according to USEPA 1311 (USEPA, 1992) and the Nether-

lands tank leaching test according to NEN 7345 (NEN 7345, 1993). In TCLP test, dried samples are ground and passed through 9.5 mm standard sieve. An acetic acid solution (0.57% v/v) was added to samples at a constant liquid to solid ratio of 20:1. After rotating the samples at 30 ± 2 rpm for 18 h the leachate was filtered with 0.45 µm filter paper and analyzed for Cr, As, Cu, Ni, Cd, Pb and Zn using AAS (Shimadzu A 6800). The Cr(VI) concentration was determined using a UV-vis spectrophotometer (HACH, DR/4000) at 540 nm using 1,5-diphenylcarbohydrazide method which is equivalent to USGS method I-1230–85 for water.

The NEN 7345 method is generally suited to building materials and mostly followed in Netherlands and EU. In this test, each of the samples was introduced into a polyethylene container and filled with acidified water (with HNO_3 at pH = 4) (Fig. 2). Volume of extractant fluid in each recipient was approximately 5 times the volume of sample, and the sample was completely submerged with the fluid level not less than 5 cm high from the top of the sample. For each sample, eight extractions were done. The leachate was removed and replaced with fresh extractant fluid after 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days as per NEN 7345. Leachate obtained



Fig. 2. An arrangement of the leaching test (according to NEN 7345) being carried out in the laboratory with sludge-amended bricks.

from each extraction was filtered through 0.45 μm filter, and analyzed using AAS (Shimadzu A 6800) for the above-mentioned heavy metals. Eq. (2) was used to compute leachability of each pollutant (heavy metals) at the *i*-th extraction.

$$E_i = \frac{(C_i - C_o)V}{1000A} \tag{2}$$

where E_i = leachability of a pollutant at the i-th extraction (mg m⁻²); C_i = pollutant concentration at the i-th extraction (mg/l); C_o = pollutant concentration in the blank (mg/l); V = volume of extractant agent (L); A = surface area of the sample (m²).

After eight extractions, Eq. (3) was used to compute the leachability, *E*, for the targeted heavy metals.

$$E = \sum_{i=1}^{8} E_i \tag{3}$$

3. Result and discussion

3.1. Characteristics of sludge

Table 2 shows the pH, moisture content, organic content and metals content (mg/kg) for the tannery sludge and clay. The moisture content of the tannery sludge and clay was $70 \pm 8.34\%$ and $2.42 \pm 0.9\%$, respectively. The organic content of tannery sludge and clay was $28.75 \pm 3.68\%$ and $3.72 \pm 0.8\%$, respectively. Chromium was the principal heavy metal in the tannery sludge (Table 2). Mean concentration of the metals were: Cr: 19,229 mg/kg; As: 1.8 mg/kg; Pb: 63.8 mg/kg; Cd: <0.01 mg/kg; Cu: 385.7 mg/kg; Zn: 250.6 mg/kg; Ni: 139.5 mg/kg. Tannery sludge composition can vary from country to country depending on the tanning processes applied and chemicals involved. However, chromium has been the predominant constituent in the sludge as can be seen in other studies. For example, Alibardi and Cossu (2016) reported Cr in dry sludge to be 44,900 mg/kg which was several orders of magnitude higher than Pb, Cu, Zn and Ni.

Particle size distribution and chemical composition of tannery sludge and soil as percentage of major oxides are shown in Tables 3 and 4 respectively. The sludge consisted of particles of a somewhat higher size fraction than that of the soil (sludge: clay and silt 62.3%, sand 37.7%; soil: clay and silt 89.6%, sand 10.4%). The main components of tannery sludge were Fe₂O₃ (34.32%), SO₃ (28.71%), CaO (26.26%), SiO₂ (3.55%), Cr₂O₃ (3.36%). The clay that was used for brick manufacturing consisted of similar components present in the tannery sludge but in different quantities. So clay can be partially replaced by tannery sludge as raw material for brick production.

Table 2

The basic properties and metal content of tannery sludge and soil samples.

Properties	Sludge	Soil
рН	7.5 ± 0.2	5.5 ± 0.15
Moisture (%)	70 ± 8.34	2.42 ± 0.9
Organic content (%)	28.75 ± 3.68	3.72 ± 0.8
Total metal content (mg/kg)		
Cr	19,229 ± 6303	-
As	1.8 ± 0.3	-
Pb	63.8 ± 5.6	-
Cd	< 0.01	-
Ni	139.5 ± 1.91	-
Cu	385.7 ± 170	-
Zn	250.6 ± 68	-

indicates not available.

Table 3

Particle size distribution of tannery sludge and soil sample.

Particle size (µm)	Tannery sludge (%)	Soil (%)
<75	62.3	89.6
75–150	11.3	5.1
150-420	15.2	5.3
420-841	7.8	-
>841	3.4	-

Table 4

Chemical composition (dry weight,%) of tannery sludge and soil.

Analytes	Tannery sludge (%)	Soil (%)
SiO ₂	3.55	63.47
CaO	26.26	2.19
Al_2O_3	0.48	13.08
Fe ₂ O ₃	34.32	8.94
SO ₃	28.71	0.56
MgO	1.69	3.9
Cr_2O_3	3.36	0.04
TiO ₂	0.41	1.25
MnO	0.2	0.11
ZnO	0.08	0.02

3.2. Compaction and Atterberg Limit Tests of clay-sludge mixtures

Moisture content has the potential to affect the properties of bricks. Up to a certain point, additional water replaces air from the soil voids, but, after a relatively high degree of saturation is reached, the water occupies space which would otherwise be occupied by soil particles (Lambe, 1993). Therefore, an optimum amount of mixing water in brick manufacturing process is usually sought which would give the maximum level of compaction for a given amount of soil sample. This will result in increased bulk density, lower tendency of water absorption and eventually the highest compressive strength. The results of the compaction test from Table 5 shows that OMC increases with the increase of sludge incorporation into the mixture. The results of Atterberg tests of sludge-clay mixtures indicate that the value of Plasticity Index (PI) decreases when sludge content increases in the mixture. The PI value of soil that is used in brick manufacturing process is 18.2 which can be classified as high plastic material. The plasticity index of sludge - clay mixture was 17.6, 15.54, 13.92 and 11.6 for the addition of 10%, 20%, 30% and 40% TS respectively into mixture. According to the soil properties stated in Table 6, by applying 10% of sludge by weight, a high enough plasticity can still be maintained in the mixture that would be suitable for mixing and molding process.

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Sludge proportion (% by weight)	0	10	20	30	40
Optimum moisture content (%)	18	21	26	29 42	33
Plastic limit (%)	17	20	24	43 29	35
Plasticity index (%)	18	18	16	14	12

Table 5

Effect of moisture content on OMC, Liquid limit, Plastic limit, and Plasticity Index of sludge-clay mixtures.

Table 6

Plasticity Index and properties of soil (Panjaitan, 2014).

PI range	Plastic nature	Soil properties
0	Non plastic	Sand
<7	Low plastic	Silt
7–17	Medium plastic	Silty clay
>17	High plastic	Clay

3.3. Properties of fired bricks under laboratory conditions

3.3.1. Water absorption

While many factors such as type of clay and methods of brick manufacturing affect the durability of bricks (Liew et al., 2004), the tendency of bricks to absorb water is also an important factor. Less the amount of water penetrates into the brick, more will be its resistance to weathering and hence more will be the durability of the bricks (Begum et al., 2013; Weng et al., 2003). Fig. 3 shows the results of the water absorption test as function of sludge content and firing temperature. It has been found that the water absorption of the bricks increased with increased sludge addition thereby potentially increasing its susceptibility to weathering action. On the other hand, water absorption was found to decrease when firing temperature was increased. This can be due to the formation of the amorphous phase at high firing temperature (Cultrone et al., 2004). Generally, sludge contains high amount of organic content which generates pore spaces within the brick when fired at high temperatures and these pore spaces favor water absorption. Similar trends in water absorption with sludge fraction in bricks and various construction materials have been observed in other studies (Basegio et al., 2002; Ukwatta et al., 2016; Monteiro et al., 2008; Quesada et al., 2015). Table 7 shows the comparison of water absorption of 10% TS bricks with similar building materials manufactured with different types of sludges. In each of these cases, there was an increase in water absorption with respect to



Fig. 3. Water absorption of bricks as function of sludge content and firing temperature. The dashed line represents maximum water absorption requirements for different grades of bricks according to BDS 208.

the reference brick. Considering water absorption standards stated in BDS 208 (2009) (Table 1) bricks made with 10% sludge burnt at 1000 °C can be regarded as Grade-S category and bricks with 20– 30% sludge fired at 1000 °C and 10–20% sludge fired at 950 °C as well as 10% TS bricks fired at 900 °C fall within Grade-A category bricks. All bricks except the one having 40% TS fired at 900 °C satisfy the ASTM requirement of water absorption for Grade SW and MW category bricks.

3.3.2. Weight loss on ignition

The Effect of sludge content and firing temperature on weight loss of bricks are shown in Fig. 4. It has been found the weight loss of bricks increased as percentage of sludge is increased. The weight loss of bricks also depends on the firing temperature. With the increase of firing temperature weight loss increases. The reference bricks fired at 900 °C showed the lowest weight loss of 5.52%, while the 40% sludge incorporated bricks sintered at 1000 °C showed the highest weight loss of 16.5%. This weight loss could be due to the combustion and decomposition of the organic and inorganic matter present in both the tannery sludge and clay during the firing process (Liew et al., 2004; Weng et al., 2003). Similar characteristics of weight loss have been observed in bricks incorporating sewage sludge (Ukwatta et al., 2016; Monteiro et al., 2008), textile effluent sludge (Begum et al., 2013) and arsenic-rich sludge (Rouf and Hossain, 2003) (see Table 7). Though BDS and ASTM do not propose any criteria on weight loss in ignition, a 15% maximum limit is often recommended (Begum et al. 2013). In that respect, most of the bricks tested satisfy the criteria. Light weight bricks are actually a favorable property for a construction material because of their lower dead load, ease in handling, savings in cost during transportation and better thermal insulation.

3.3.3. Compressive strength

The compressive strength is the key parameter for ensuring the engineering quality of building materials. The results of the compressive strength test of the bricks are shown in Fig. 5. The compressive strength of TS incorporated bricks ranged from 10.98 MPa to 29.61 MPa depending on the sludge content and sintering temperature. Compressive strength has been found to be inversely proportional to the sludge content and directly proportional to the firing temperature. This may be due to decrease in porosity and increase in bulk density resulting from increased firing temperature (Bhatnagar and Goel, 2002). Similar trend of compressive strength reduction with sludge content has been previously observed in other studies (Basegio et al., 2002; Begum et al., 2013; Ukwatta et al., 2016; Monteiro et al., 2008; Hassan et al., 2014) (see also Table 7). The addition of 10%, 20%, 30% and 40% sludge into the mixture reduces strength approximately by 19%, 31%, 48% and 56% respectively compared with the reference bricks fired at 1000 °C. According to BDS (2009) standards, 10% TS bricks burnt at both 950 °C and 1000 °C can be considered as Grade-S and 20-30% TS bricks fired at 1000 °C, 20% TS bricks fired at 950 °C as well as 10-20% TS bricks fired at 900 °C fall within Grade-A category bricks (Fig. 5). However, all the samples satisfied the minimum criteria of compressive strength for bricks which is 8.6 MPa and 10.29 MPa according to ASTM (ASTM C 62, 2012) and Bangladesh standards (BDS 208, 2009) respectively.

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Table 7

Performance comparison for 10% sludge incorporated bricks with various waste stabilization attempts involving sludge and building materials. The % increase/reduction in the parenthesis indicate how much the specific parameter changed compared to the reference brick (i.e. 0% sludge).

			Previous studies (10% sludge incorporated bricks)				
Physical and Mechanical Properties	Present study (10% TS bricks)	Tannery sludge in ceramic tiles (Basegio et al., 2002)	Sewage sludge in bricks (Ukwatta et al., 2016)	Sewage sludge in bricks (Liew et al., 2004)	Textile sludge in bricks (Begum et al., 2013)	Arsenic-iron sludge in bricks (Hassan et al., 2014)	
Compressive strength (MPa)) 21.2–29.6 (17–25% reduction)	8 [°] (33% reduction)	35 (16% reduction)	9 (43% reduction)	5.5" (30% reduction)) 9–11	
Water absorption (%)	9.1-14.2 (20-33% increase)	20 (55% increase)	9.1 (15% increase)	26 (8% increase)	13	17-20	
Firing shrinkage (%)	6.3-9.1 (23-30% reduction)	5 ^{•••} (No change)	6.5 (32% reduction)	2 (20% reduction)	-	-	
Weight loss on ignition (%)	8.5-10.1 (35-40% increase)	-	5.26 (14% increase)	9 (28% increase)	4	-	
Bulk density (kg/m ³)	1580-1687 (10-12% reduction)) 1800 (18% reduction) 2075 (2.5% reduction)) 2440 (5% reduction)	2100	-	
Firing energy saving (%)	26	-	25	-	-	-	

^{*} Bending strength.

** Test carried out after 7 days wetting in water.

Linear shrinkage.



Fig. 4. Weight loss of bricks as function of sludge content and firing temperature.



Fig. 5. Effect of sludge content and firing temperature on compressive strength of bricks. The dashed line represents minimum strength requirements for different grades of bricks according to BDS 208.

3.3.4. Firing shrinkage

Shrinkage in bricks occurs as chemically and mechanically bound water is lost during firing (Karaman et al., 2006). High shrinkage is an undesirable property in any engineering material. Fig. 6 shows the volumetric shrinkage of bricks due to firing as a function of firing temperature and sludge content. The shrinkage



Fig. 6. Shrinkage of bricks as function of firing temperature and sludge content.

of bricks was found to increase proportionally with firing temperature but the firing shrinkage was found to decrease with the increase of sludge content in bricks which is in congruence with previous studies by Ukwatta et al. (2016) for ETP biosolid, Quesada et al. (2015) for oil refining sludge and Liew et al. (2004) for sewage sludge. For 40% sludge amended bricks fired at 1000 °C, the volumetric shrinkage reduced by up to 50.8% compared to the control bricks. There are two hypotheses as to why this has happened. Firstly, the decreasing trend obtained here may be due to nonplastic nature of dried sludge. Rhodes and Hopper (2000) reported that low plastic soil shrinks less than high plastic soil. The Plasticity Index of tannery sludge-clay mixtures was lower for high sludge amendments (Table 4) rendering the mixture to become more and more non-plastic. Secondly, the decreasing trend may be due to expansion in the bricks during the firing stage. Sludge-amended bricks could release more gases compared to control bricks due to burning of organic matter, and the gases could not escape out from the clay matrix during sintering probably due to lack of connectivity among the pore spaces. This phenomenon generated voids which may cause a net expansion within the sludge-clay matrix and eventually resulting in a lower net shrinkage (Ukwatta et al., 2016). In order to confirm the existence of pores, we analyzed and compared the internal structure of the brick specimens using a Scanning Electron Microscope (SEM) (Fig. 7). It was revealed that the control brick had a much denser and smoother inner structure than that of the TS-amended bricks. The TS-amended bricks showed pores that were larger in size and numbers than those of the control bricks which basically lends evidence in support of the above hypothesis.

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Fig. 7. SEM images captured at 2000× magnification of the internal structure of the (a) control brick and (b) 40% TS-amended brick. The TS-amended bricks showed the existence of pores which are higher in number and larger in size compared to the control bricks.

3.3.5. Bulk density

The bulk density for different percentage of TS-amended bricks fired at three defined temperatures are shown in Fig. 8. An inverse relationship was observed between the bulk density of bricks and the amount of sludge added to the mixture. In this study, bulk density decreased from 1872 to 1505 kg/m³ (reduction of 19.6%) as the TS content increased from 0% to 40% at 1000 °C firing temperature. The firing temperature can also have an effect on the bulk density of the bricks. The results show that increasing the temperature results in a slightly higher bulk density. A similar trend of bulk density with sludge content has also been shown in the case of ceramic containing tannery sludge (Basegio et al. 2002) and brick containing sewage sludge (Ukwatta et al. 2016; Liew et al. 2004)

3.3.6. Efflorescence

Waste material such has sludge contains inorganic salts that can get dissolved in water and exhibit visible deposits on the surface of the engineering material that contains them. This phenomenon of efflorescence can diminish the aesthetic quality and performance of an engineering material. Efflorescence test of the TS-amended bricks fired at 1000 °C were carried out according to BDS 208 (2009), and it was found that there were no visible salt deposits and change in aesthetic appearance in any of the samples having TS content between 10% to 40%. (Efflorescence can be reported as 'Nil' according to BDS208 classification). The appearance of bricks before and after efflorescence test is shown in Fig 9. This suggests that TS-amended bricks fired at 1000 °C are not susceptible to efflorescence and can be exposed to a wide range



Fig. 8. Bulk density of the bricks as function of sludge content and firing temperature.

of climatic conditions and will be able to maintain its aesthetic qualities.

3.4. Energy saving during firing

The calorific value of tannery sludge used in this study has been found to be 5.85 ± 0.76 MJ/kg. The estimated amount of energy saved during firing of TS bricks was calculated using Eq. (1) which has been interpreted in Table 8. It can be estimated that approximately 15-47% energy savings can be achieved for 10-40% TS-amended bricks respectively compared to reference bricks containing no sludge. Similar nature of energy savings was reported in Ukwatta et al. (2016) for ETP biosolid- incorporated bricks and at Mohajerani et al. (2016) for bricks incorporating cigarette butts. Tannery sludge used in this study contained about 28.75\% organic content which could facilitate heat input to the furnace and reduce the amount of energy required for firing.

3.5. Comparison of properties with kiln-fired bricks

By applying linear interpolation in Figs. 3 and 5, it is theoretically possible to find the maximum TS content that would exactly satisfy the criteria for Grade-S quality bricks in Bangladesh according to BDS 208(2009). Accordingly, in order to satisfy both the criteria for water absorption and compressive strength, a TS content of 2%, 9% and 12% would be required for bricks sintered at 900 °C, 950 °C and 1000 °C, respectively. However, in the interest of choosing a particular set of bricks from those prepared in the previous experiments and accounting for margin of error in experiments, we propose that 10% TS bricks fired at 1000 °C could be proposed as an ideal model to apply in this case. Therefore, 10% TS was used to prepare bricks in the field condition ('brick kiln'). Table 9 shows the properties of 10% TS-amended brick along with control bricks prepared in the brick kiln. For the purpose of comparison, the properties of 10% TS bricks in the laboratory under various sintering temperatures was used as these resembled the closest possible reference values under the given operating conditions in the kiln. The compressive strength of 10% TS-amended brick ("field condition") was found to be 16.3 MPa which was 22-45% lower in strength compared to that made in laboratory conditions. Despite the reduction in compressive strength, these bricks still satisfied the ASTM standard and BDS requirement for building construction (Table 1). The difference in quality between the bricks manufactured in two different conditions was expected as there was a variation in molding process and OMC. During brick molding at laboratory, OMC was accurately maintained and compaction by hand was applied very carefully for individual bricks under controlled conditions which may have contributed to

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Fig. 9. Appearance of TS-amended bricks before (a) and after (b) efflorescence test.

Table 8

Calculation for energy saved during firing.

Sludge content (%)	Mass of control brick, m1 (kg)	Mass of TS brick, m ₂ (kg)	Mass of TSs in TS brick, m ₃ (kg)	Energy used for control brick, Q1 (MJ)	Energy used for TS brick, Q ₂ (MJ)	Energy saved (Q ₁ - Q ₂), MJ	% of energy saved
10	0.505	0.489	0.049	2.525	2.158	0.367	15
20	0.505	0.473	0.095	2.525	1.809	0.716	28
30	0.505	0.492	0.146	2.525	1.606	0.919	36
40	0.505	0.498	0.198	2.525	1.332	1.193	47

Table 9

Comparison of properties of sludge incorporated bricks made in kiln and laboratory.

	Bricks ma	ade in kiln	
Parameters	10% sludge	Control (0% sludge)	Laboratory condition (10% sludge fired at 900–1000 °C)
Compressive strength (MPa) Water absorption (%) Weight loss on ignition (%) Firing shrinkage (%) Bulk density (kg/m ³)	$16.3 \pm 2.9 \\19 \pm 1.12 \\10.8 \pm 1.18 \\10.5 \pm 1.3 \\1500 \pm 50$	$16.8 \pm 2.5 \\ 16 \pm 0.6 \\ 8.5 \pm 0.8 \\ 11.2 \pm 1.45 \\ 1520 \pm 30$	21–29.6 9.1–10.2 8.5–10.1 6.3–9.1 1580–1687

improved compaction of clay-sludge mixture resulting in increased compressive strength, lower water absorption, lower shrinkage and higher bulk density. Under field conditions in the kiln, the OMC was not maintained and limited compaction was applied to the clay mixture during molding. Quality of TS-amended bricks can be improved infield condition if automatic molding machines are used which applies pressure during molding.

3.6. Environmental aspects of sludge amended bricks

Leaching of heavy metal can be a concern as heavy metal contaminated sludge is introduced to the finished products. It should be ensured that leaching of heavy metal from bricks does not exceed the maximum permissible limit even under extreme conditions. The NEN 7345 leaching test was carried out for bricks fired at 1000 °C. The cumulative leachability values of targeted heavy metals from the sludge incorporated and control bricks fired at 1000 °C are summarized in Table 10. According to Dutch regulations, if cumulative results from the NEN 7345 leaching test do not exceed U₁ values, sludge incorporated bricks can be used in construction without any restrictions. If cumulative results exceed U₂ values, they cannot be used as construction materials and if the results are between U₁ and U₂ they can be used as construction materials but they need to be treated once their life cycle is ended (NEN 7345, 1993). The cumulative leachability values for all heavy metals analyzed in this study were insignificant as they were found to be far below U₁ (Table 10). The leached concentration of chromium was found to be 0.161 mg/m², 0.345 mg/m², 0.290 mg/m² and 0.499 mg/m² for 10%, 20%, 30% and 40% TS bricks respectively. Analysis showed that about 15-50% of total chromium was present in hexavalent form, which is the most toxic species of chromium. Zn, Cu, Pb, Ni, and As have been found to have leached in lower concentrations from sludge amended bricks compared to control brick (100% clav).

The leachate analysis according to USEPA 1311 from sludge amended bricks fired at 1000 °C, 950 °C and 900 °C temperature is given in Table 11. Pb, Cu, Zn and As leached from sludge-amended bricks in negligible amounts and far below the USEPA regulatory limits. The hexavalent chromium (Cr(VI)) in the leachate increased with the increased TS content in the brick (not shown in the table); in case of 1000 °C sintering temperature, the concentration in leachate was 0.002 mg/l, 0.612 mg/l, 0.472 mg/l

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Sludge content	Cr (mg/m ²)	Zn (mg/m ²)	Cu (mg/m ²)	Pb (mg/m ²)	Ni (mg/m ²)	As (mg/m ²)
0%	0.008	0.006	0.021	0.086	0.008	0.007
10%	0.161	0.004	n.d	0.027	0.002	0.002
20%	0.345	0.005	0.003	0.020	n.d	n.d
30%	0.290	0.001	0.003	0.055	n.d	n.d
40%	0.499	n.d	0.003	0.062	n.d	n.d
Leaching limits set by t	he Netherlands Tank Lead	ching Test (NEN 7345)				
U ₁	150	200	50	100	50	40
U ₂	950	1500	350	800	350	300

Results of the tank leaching tests in bricks samples after 8 extractions (64 days) according to NEN 7345.

"n.d" = not detected. Detection limit for Zn, Cu, Ni and As are> 0.001 mg/l.

Table 11

Table 10

TCLP test result of TS amended bricks fired at 1000 °C, 950 °C and 900 °C temperature.

Metals	Firing temp.	0% TS brick	10% TS bricks	20% TS bricks	30% TS bricks	40% TS bricks	USEPA regulatory limit ^a
Cr (mg/l)	1000 °C 950 °C 900 °C	0.033 n.d 0.02	0.28 ± 0.12 0.665 ± 0.15 0.745 ± 0.05	0.908 ± 0.2 1.04 ± 0.31 1.18 ± 0.4	1.07 ± 0.23 1.25 ± 0.02 1.67 ± 0.03	1.08 ± 0.26 1.17 ± 0.03 2.18 ± 0.03	5
Pb (mg/l)	1000 °C 950 °C 900 °C	0.17 0.063 0.07	0.094 ± 0.03 0.096 ± 0.08 0.134 ± 0.02	0.129 ± 0.06 0.116 ± 0.05 0.146 ± 0.02	0.12 ± 0.002 0.122 ± 0.05 0.098 ± 0.01	$\begin{array}{c} 0.11 \pm 0.004 \\ 0.137 \pm 0.04 \\ 0.109 \pm 0.02 \end{array}$	5
Ni (mg/l)	1000 °C 950 °C 900 °C	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	11
Cu (mg/l)	1000 °C 950 °C 900 °C	0.214 0.085 0.216	0.211 ± 0.02 0.185 ± 0.07 0.273 ± 0.02	0.123 ± 0.1 0.182 ± 0.01 0.116 ± 0.05	0.234 ± 0.1 0.19 ± 0.06 0.16 ± 0.01	0.249 ± 0.07 0.12 ± 0.09 0.31 ± 0.03	100
Zn (mg/l)	1000 °C 950 °C 900 °C	0.181 1.625 1.48	0.312 ± 0.2 0.736 ± 0.1 0.56 ± 0.1	0.307 ± 0.3 0.243 ± 0.05 1.07 ± 0.03	0.587 ± 0.3 0.352 ± 0.03 1.26 ± 0.1	0.767 ± 0.4 0.24 ± 0.08 1.15 ± 0.07	500
As (mg/l)	1000 °C 950 °C 900 °C	0.017 0.047 0.067	0.124 ± 0.16 0.123 ± 0.02 0.127 ± 0.16	0.134 ± 0.16 0.117 ± 0.03 0.125 ± 0.16	$\begin{array}{c} 0.036 \pm 0.12 \\ 0.126 \pm 0.05 \\ 0.036 \pm 0.12 \end{array}$	0.084 ± 0.08 0.115 ± 0.02 0.084 ± 0.08	5
Cd (mg/l)	1000 °C 950 °C 900 °C	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	n.d. n.d. n.d.	1

"n.d." = not detected; Detection limit for Ni and Cd are >0.001 mg/l and >0.01 mg/l respectively.

^a United States Environmental Protection Agency (USEPA) (1996).

and 0.726 mg/l for 10%, 20%, 30% and 40% TS bricks, respectively which amounted to 1 to 67% of total chromium leached. But since the leached total chromium in the leachate for all the sludge amended bricks has been found to be below the EPA regulatory limit of 5 mg/l, it can be considered safe for disposal. These metals are believed to be locked inside the fired bricks as metal oxide (Liew et al., 2004). Firing temperature is a very important factor affecting leachability of Cr as the leaching of Cr reduced from 0.745 mg/l to 0.28 mg/l when firing temperature increased from 900 °C to 1000 °C (Table 11). Other researchers noticed a similar decrease in Cr leaching at high temperatures and several explanations have been provided for such behavior. Some authors explained that at elevated temperatures, Na₂CrO₄ and K₂CrO₄ could form an amorphous phase with SiO₂, thus preventing Cr from leaching (Verbinnen et al., 2013; Verbinnen et al., 2014). Others suggested that at higher temperatures (800 °C or above) the reduction of Cr(VI) to Cr(III) occurs, CaCrO₄ becomes the dominant crystalline phase in co-existence with magnesium chromite $(MgCr_2O_4)$ in which chromium is found as Cr(III) and the mobility of chromium becomes limited as these compounds are insoluble in water and weak acids (Wei et al., 2005; Abreu and Toffoli, 2009; Kavouras et al., 2015).

These experiments suggest that heavy metal contaminants can be effectively immobilized by incorporating the sludge in clay mixtures for brick manufacturing. Most of the heavy metal concentration from the TCLP tests were found to be slightly higher than that from the tank leaching test results. This can be attributed to the differences between the two test methods regarding sample preparation and leachate extraction. However, given the environment the samples undergo during experiments, the tank leaching test appears to present a comparatively realistic scenario of leaching for construction materials such as bricks.

4. Conclusion

In this study a range of properties of tannery sludgeincorporated clay bricks were studied to understand their behavior under different temperatures and sludge contents with an aim to determine their suitability as a viable construction material. Bricks were manufactured in the laboratory and in a brick kiln and the effect of varying sludge content in the clay mix and manufacturing environment were investigated. It was found that

• Similar to other studies with sludge-incorporated building materials, water absorption increased and the compressive strength decreased with increased TS content but TS-amended bricks have been generally found to meet both BDS and ASTM requirement for bricks to be used in construction. 10% TS by weight can be an optimum recipe for TS-amended bricks without diminishing its engineering properties. The 10% TS bricks

produced in the kiln for commercial purpose, though showing lower quality compared to the 10% TS bricks produced in laboratory, satisfied strength requirement for building construction materials.

- Reduced shrinkage, weight and bulk density as well as the absence of efflorescence of TS-amended bricks can make the use of these more appealing.
- The firing energy of bricks is estimated to be saved up to 15–47% by incorporating 10–40% TS content which can pave the way for its sustainable use.
- The leached concentrations of targeted heavy metals have been found to be insignificant and far below Dutch regulations and USEPA regulatory limits. The results from TCLP tests showed that an increase in firing temperature caused further immobilization of heavy metals such as chromium.

The results obtained from the present research indicate that by incorporating tannery sludge it is possible to produce good quality bricks that can satisfy all the required mechanical and physical properties as per ASTM and BDS standards and there will be minimal cause for concern for leaching of toxic metals in the environment. It is a promising venture for successfully recycling waste products particularly in the context of Bangladesh and the brick industry should be encouraged to adopt this practice in their operation.

Acknowledgements

Authors wish to express their gratitude to Committee for Advanced Studies and Research (CASR), BUET, Bangladesh and NGO FORUM for public health for their financial support to carry out this research work. Special thanks to Apex Tannery Ltd. Unit-2 for providing sludge sample and the Department of Glass and Ceramic Engineering, BUET for performing the SEM analysis.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2016.12. 041.

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