Construction and Building Materials 314 (2022) 125702

Contents lists available at ScienceDirect

Construction and Building Materials

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

Stabilization of liming sludge in brick production: A way to reduce pollution in tannery

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A R T I C L E I N F O

Keywords:
Liming sludge
Tannery
Construction material
Eco-friendly
TCLP
Waste management

A B S T R A C T

In leather processing, hair burning liming operation is employed to remove the hair/wool, epidermis, fat, grease, and non-structural proteins. The discharged wastewater from the hair burning step produces the most of the sludge in the tannery which is a hazardous waste that has a serious impact on the environment. Thus, managing of the liming sludge has become a great challenge for the leather industry. Using liming sludge as a raw material in building materials has the potential to reduce environmental pollution. In this novel work, building bricks were made under field conditions, where liming sludge (w/w) were incorporated by 2%, 4%, 6%, 8%, 10%, and 12%, and fired in a kiln at 1000 °C. Results reveal that 6% liming sludge incorporated fired brick showed the best engineering properties, microstructure, and chemical characteristics. It also offered the best properties such as compressive strength (ASTM C 67-02c), area shrinkage (ASTM C326), weight loss, bulk density, and water absorption (ASTM C373-88) which were found to be 27.50 MPa, 5.03%, 7.79%, 2.18 g/cm^3, and 10.46%, respectively. Moreover, Fourier-Transform Infrared (FT-IR) spectroscopy and Scanning Electron Microscope (SEM) analysis showed that liming sludge content has a profound effect on the structure of fired bricks. The Netherlands tank (NEN 7345) and Toxicity Characteristic Leaching Procedure (TCLP) (USEPA 1311) leaching test results exhibited insignificant leachability of metals from the sludge incorporated fired bricks. Hence, the utilization of liming sludge as an ingredient for producing construction materials will reduce the pollution loads of tannery industries.

1. Introduction

Sustainable waste management in the tannery industry has become a great challenge due to the incessant generation of huge amounts of pollution throughout the world [1]. Annually 6960.3 thousand tons of hides and skins are processed worldwide, of which 6 million tons of solid waste and 120 million tons of liquid waste are generated [2]. For instance, during the processing of 1000 kg of raw hide/skin, typically 50,000 kg wastewater and 150–250 kg sludge are produced [3]. Different types of toxic heavy metals like Zn, Cu, Pb, Cr, As, Ni, Hg, and Cd are found to be present in the tannery sludge [4,5].

In beamhouse operations, unhairing and liming (termed as liming) wastewater contains a high amount of biochemical oxygen demand (BOD), sulfides, chemical oxygen demand (COD), chlorides which contribute 60–70% of the total pollution load (TPL) created by tanneries [6]. In liming, hide/skin is treated with lime and sodium sulfide [7] to dissolve epidermis, non-fibrous protein, and fat/grease [8], which produce most of the pollution load in leather processing [9]. Wastewater ejected through liming operations comprises soluble sulfide, total suspended solids, high BOD, and COD due to rupturing of the epidermis, non-structural protein, and hair/wool [10]. As a result, wastewater expelled from the unhairing and liming operation is toxic to the aquatic environment [11] and forms a huge amount of sludge [6]. Hu et al. [12] reported that in typical liming for every 1000 kg of rawhide produces 5000–7000 kg of total effluents.

Doble and Kumar [7] stated that the wastewater from liming accounts for 45% of the entire effluent volume and contributes to 30% of the overall BOD and COD. The solid waste containing liming wastewater is directly discharged through the drain or to the effluent treatment plant (ETP). It harms the aquatic environment and human health because liming sludge contains different non-biodegradable toxic metals like zinc (Zn), copper (Cu), chromium (Cr), cadmium (Cd), nickel (Ni) [13-15], and keratin (wool, hair, epidermis) [16]. Hashem et al. [17] estimated that in Bangladesh, yearly 5,100–8,925 tons of liming sludge is generated and these huge amounts of toxic sludge are disposed of indiscriminately without proper management. Therefore, proper

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https://doi.org/10.1016/j.conbuildmat.2021.125702
Received 13 April 2021; Received in revised form 7 November 2021; Accepted 12 November 2021
Available online 20 November 2021
0950-0618/

disposal of the liming sludge considering the environmental conservations has become a serious challenge in Bangladesh.

For liming sludge stabilization, different techniques, for instance, incineration, landfilling, pyrolysis, and bio-oFil have been explored [18,19]. Incineration can reduce the volume of sludge by more than 30% and helps to reduce heavy metals' evaporation by volatilizing the organic content and forming stable compounds with the inorganic components like Al_2O_3, ZnO in the presence of oxygen [20]. Incineration can contaminate the environment by generating different toxic gases such as ammonia (NH_3), nitrous oxide (N_2O), carbon dioxide (CO_2), and ash, which is another harmful waste [21]. Because of the high operating temperature and inert gas atmospheric condition, the gases emitted during pyrolysis contain more detrimental compounds than incineration [22]. Another disadvantage of pyrolysis compared to incineration is the maintenance costs and less advanced technology [23]. Suitable landfilling is difficult and there is a possibility of heavy metal leaching from the sludge into the surrounding soil [24]. For example, a high concentration of mercury was observed in a landfilling area in Florida [25]. Liming sludge can be used as a substitute for artificial fertilizer and a nutrient for plant growth [26] but due to the high pH (8–9) of liming sludge, fertilizer from liming sludge can trigger the growth of fungus and other micropollutants in the soil, subsequently hampering plants' growth [27].

A group of researchers produced environmentally friendly cement from liming sludge [28]. Construction material like cement from liming sludge could be a viable solution to reduce pollution and help with building design. Among construction materials, brick is one of the major elements for making a building. At present, the demand for bricks is rising globally every day [29]. Some researchers have attempted to make eco-friendly bricks using waste materials like fly ash [30], granite sawing waste [31], municipal solid waste [32], and municipal solid waste incineration fly ash [33]. A feasible method for cleaner building materials could be implemented by manufacturing bricks from waste materials which will reutilize the waste as well as reduce the consumption of clay [34].

In this work, liming sludge generated from the expelled liming wastewater is used as raw material for brick production that could reduce the pollution load in the tannery. In-situ liming sludge was formed in a tannery and a brick sample was prepared by mixing liming sludge with clay at different ratios. The quality of bricks was examined by assessing compressive strength, bulk density, leaching test of sludge incorporated brick, water absorption capability, efflorescence test, weight loss on ignition, Fourier-Transform Infrared (FT-IR) Spectroscopy, and Scanning Electron Microscope (SEM) analysis were carried out on developed brick products.

2. Materials and methods

2.1. Estimation of liming sludge

To estimate the amount of sludge generation during liming operation, six pieces of wet salted cowhide and twenty-seven pieces of goatskin was purchased randomly from a local slaughterhouse, Khulna, Bangladesh for pilot scale liming sludge (LS) production. The average weight of the wet salted cowhide and goatskin was 6.83 kg and 1.86 kg, respectively. Batch-wise triplet unhairing and liming were carried out on cowhide and goatskin. The sequential beamhouse operations, i.e. presoaking, soaking, and liming were carried out following the conventional method [35]. After completing the liming and unhairing process, the generated wastewater was culled into a bucket. For sludge formation, coagulant (aluminum sulfate) was added gradually into the bucket with continuous stirring; pH was adjusted to 8–9 and kept for 12 h to settle down. After decantation, liming sludge was collected from the bottom of the bucket. The sludge was sun-dried and then oven-dried for 24 h at 105 °C. The dried liming sludge was weighed to estimate the sludge generation per kg of wet salted cowhide and goatskin.

2.2. Liming sludge preparation

A large scale of liming sludge was made for use in brick preparation in the Superex Leather Limited which is situated in Khulna, Bangladesh. After the liming operation had finished, the generated wastewater was culled into 125-liter capacity plastic drums. For the coagulation process, coagulant (aluminum sulfate) was added to the drum, stirred for 10 min, and pH was adjusted 8–9. At pH 8–9, coagulant (aluminum sulfate) destabilized most of the suspended solids from the wastewater and settled down. The suspension was kept in a static condition for 12 h for settling. After decantation, liming sludge was collected from the bottom of the drum.

2.3. Characterization of liming wastewater

During sampling, the standard method was followed carefully. The glassware and accessories were washed with diluted nitric acid and properly rinsed with deionized water to avoid contamination which may change the results. Moreover, the experiments were carried out in a set of three to get a standard deviation and mean value. The pH of the sample was measured using a pH meter (UPH-314, UNILAB, USA). Electrical conductivity (EC) was measured by using a conductivity meter (CT-676, BOECO, Germany). Total dissolved solids (TDS) and total suspended solids (TSS) were determined by following American Public Health Association (APHA) [36] methods. Determination of BOD and COD was carried out by following APHA-5210B [37] and APHA-5220C [38], respectively. The chloride content (Cl^-) of the sample was determined by following the analytical method APHA-4500B [39]. SLTC 202 [40] was followed to measure the sulfide content of the liming wastewater.

2.4. Liming sludge and clay ground for brick production

The collected liming sludge was sun-dried for 72 h to remove the excess moisture. About 1000 kg clay samples were collected from Super Bricks Limited Khulna, Bangladesh, and sun-dried for 72 h. After that, both liming sludge and clay were oven dried for 24 h at 105 °C. To obtain identical particle size, the clay and liming sludge were ground to a particle size of 0.1 mm with a grinder machine (Pulverisette, Germany).

2.5. Characterization of sludge and clay

Moisture content and pH of the raw clay and liming sludge were measured by applying the British Standard method BS 1377–1 [41]. APHA [36] method was followed to determine the organic content of the clay and sludge. EPA 3050B method was employed for sample preparation to determine the heavy metal contents of the liming sludge and clay. About 5 g of each dry sample was dissolved with concentrated nitric acid and heated at 150 °C until the brown-colored fume was produced. After adding 200–250 mL of deionized water, the samples were heated at 70 °C for one and half hours. Then the solutions were sanctioned to chill at room temperature and filtrated through a filter paper of 0.45 μm pore size. Finally, the volume of the filtrate samples was made up to 100 mL by adding deionized water and the heavy metal content of the filtrate samples was determined by using atomic absorption spectroscopy (AAS) (SpectraAA 240FS, Agilent, USA). Sieve analysis was carried out according to ASTM D 422–63 [42] in determining the particle size distribution of the liming sludge and clay. The Atterberg limit test was performed according to ASTM D 4318 [43] standard method.

2.6. Fabrication of bricks

Different percentages of liming sludge 2%, 4%, 6%, 8%, 10%, and 12% were mixed homogeneously with the clay. In each batch, the amount of dry clay and liming sludge mixture was around 80 kg. Later, 25.21%, 25.30%, 26.17%, 27.81%, 28.96%, and 31.95% water were respectively added for 2%, 4%, 6%, 8%, 10%, and 12% liming sludge incorporating batches during the mixing stage. Then a clean wooden mold was filled with the brick material (premixed liming sludge and clay) and after one minute this mixture was demolded without applying any pressure by turning it upside down. In this way, a total of 100 molded bricks were prepared and their dimensions were 23.5 cm × 11.5 cm × 6.5 cm [44] (Fig. 1(b)). Then the molded solid bricks were allowed to dry at room temperature for 48 h. After that, they were sun-dried for 15 days. The dried bricks were burned in a kiln at 1000 °C. Finally, the burned bricks were withdrawn from the kiln and left at a normal temperature (Fig. 1(c)). All the procedures were carried out in a commercial setting.

Fig. 1. Liming sludge and clay mixture (a) fabricated raw bricks (b) and fabricated fired bricks (c).

2.7. *Testing of produced bricks*

Physicomechanical tests were carried out to evaluate the quality of bricks. Three bricks were used for each test and the mean value, as well as standard deviation, were calculated. The compressive strength of the produced bricks was determined by following the standard method [45]. The shrinkage area on firing was determined according to ASTM C326 [46]. ASTM C373-88 [47] was employed to determine the water absorption capacity and bulk density of the produced bricks. Weight loss on ignition was measured by calculating the before and after weight of the bricks. BDS 208 [48] served to carry out an efflorescence test.

2.8. *Microstructure and chemical characteristics of raw material and fired bricks*

The chemical composition of the raw liming sludge and clay sample as a percentage of oxide was determined by using an X-Ray Fluorescence Spectrophotometer (XRF-1800, Shimadzu). The functional group of the raw samples and the fired bricks was identified using an FT-IR spectrometer (Spectrum 100, PerkinElmer, USA). For every sample, 60 scans were run to avoid the noise at an intensity of 4 cm^{-1} in the wave range of 450–4000 cm^{-1}. The morphology of the fired bricks was investigated with a SEM (S3400, Hitachi, Japan). For SEM analysis the sample was spread on carbon tape and coated with platinum to prevent overcharging. SEM images were obtained at 1000X magnification with an accelerating voltage of 15 kV.

2.9. *Leaching test*

The environmental compatibility of the fabricated bricks was analyzed by carrying out two leaching methods: Toxicity Characteristic Leaching Procedure (TCLP) according to USEPA 1311 [49] and the Netherlands tank leaching test according to NEN 7345 [50]. In USEPA 1311 method, 25 g of dried sample (size less than 1 mm) was placed into a plastic cylinder to which 500 mL acetic acid solution was added. The pH of the mixture was 2.81 ± 0.01. Then, the cylinder was set on a rotary agitator device and the solution was agitated at 32 rpm for 18 h. After agitation, the extracted fluid was filtered through a filter paper of 0.45-μm pore size, and the filtrate samples were analyzed for determining the heavy metal concentration by using AAS.

The Netherlands Tank Leaching Test was performed by following the norms of NEN 7345 [51]. In this test, each specimen was submerged in acidic water (maintaining pH 4 by adding HNO_3) in such a way that the top layer of the bricks had to remain at least 5 cm deep from the water surface (FS1). In total, eight extractions (at 0.25, 1.0, 2.25, 4, 9, 16, 36, and 64 days) were carried out for each sample and the volume of the extracting fluid volume was 5 times of the bricks. After every extraction further submersion was done similarly as before. The extracted fluid was filtered through a filter paper of 0.45-μm pore size then the filtrate was analyzed using AAS. The leachability at the t^{th} extraction was calculated via the following equation (i):

$$E_t = \frac{(C_t - C_0)V}{1000 \times A} \quad (1)$$

where E_t, pollutant leachability at the t^{th} extraction (mgm^{-2}); C_t, the concentration of the pollutant at the t^{th} extraction (mgL^{-1}); C_o, the concentration of the pollutant in the blank (mgL^{-1}); V, the volume of extracting fluid (L); A, surface area of the brick (m^2). After eight extractions, the cumulative leachability was calculated by using the following equation (ii):

$$E = \sum_{t-1}^{8} E_t \tag{2}$$

3. Results and discussion

3.1. Sludge estimation

The production of liming sludge per unit (w/w) of cowhide and goatskin was analyzed on a pilot scale. For this experiment, conventional pre-soaking, soaking, and liming operations were conducted for 6 pieces of wet salted cowhide and 27 pieces of wet salted goatskin. After the liming operation, the collected wastewater was produced 4.4 kg and 16.11 kg of liming sludge from cowhide and goatskin, respectively. It is calculated that during liming operation for one kg of wet salted cowhide and one kg of wet salted goatskin produces 0.10 kg and 0.30 kg liming sludge, respectively. It can be said that compared to cowhide, goatskin produces 3 times higher liming sludge. This huge amount of liming sludge reveals an outstanding opportunity for waste-to-wealth utilization for brick production as well as will significantly reduce the pollution load of the wastewater.

3.2. Characterization of liming wastewater

The discharged wastewater from the liming operation is composed of different pollutants with high pH values. The pollution load parameters like electrical conductivity (EC), chloride, TSS, TDS, BOD, and COD released from the liming process also vary from each other. Characterization of wastewater is the key step for taking the initiative to manage the wastewater released from the liming process. Table 1 depicts the physicochemical parameters of liming wastewater before and after treatment. The pH of the liming wastewater was 11.2 due to the use of lime which was necessary for the operation; liming wastewater produces the highest pH among leather processing operations [53]. During coagulation, the pH of the wastewater was adjusted to 8.4. Before and after treatment, the electrical conductivity of the wastewater was found to be 37.3 and 22.4 mS, respectively, which were far above the permissible limit of ECR (1997). The high EC level indicates the wastewater may contain different soluble salts. The chloride content of the raw wastewater was 5432.2 mg/L which was much higher than the standard value (600 mg/L). This might be because of the usage of common salt (NaCl) in the preservation process which was not completely removed from the hide/skin during the previous soaking process [54]. Although the coagulation process removes 51.6% chloride, still the chloride content is beyond the permissible limit. Since liming wastewater contains a huge amount of grease, protein residue, and dissolved hair, consequently, the total suspended solids (TSS) (18357.2 mg/L) were greater than the total dissolved solids (TDS) (13173.9 mg/L). During coagulation, most of the solids (e.g., dissolved solids and suspended solids) settled down at the bottom of the container and were eventually removed from the wastewater [55].

Table 1
Physiochemical parameters of liming wastewater.

Parameters	Unit	Raw	Treated	Removal Efficiency (%)	Permissible limit[52]
pH	–	11.2 ± 0.5	8.4 ± 0.3	–	6–9
EC	mS	37.3 ± 3.3	22.4 ± 2.2	40.0	1.2
Cl^-	mg/L	5432.2 ± 58.4	2629 ± 23.1	51.6	600
TDS	mg/L	13173.9 ± 79.4	1449 ± 28.2	89.0	2100
TSS	mg/L	18357.2 ± 86.3	1268.2 ± 35.02	93.1	150
BOD	mg/L	11473.6 ± 93.2	2130.5 ± 23.4	81.4	100
COD	mg/L	21783.9 ± 175.8	3848.7 ± 43.01	82.3	400

The removal efficiencies of TDS and TSS from the liming wastewater were 89% and 93.1%, respectively. It seems that the TDS of the treated wastewater was 1449 mg/L which was within the permissible limit (2100 mg/L) but the TSS of the treated wastewater was a little bit higher than the value of the standards. The BOD of raw liming wastewater was 11473.6 mg/L which was above the permissible limit. The reason behind this high BOD is that the liming wastewater still contains a huge amount of organic substances and different unabsorbed chemicals [56]. After coagulation as well as treatment, BOD level was 2130.5 mg/L and removal efficiency was 81.4%, which was much better than what other studies documented [57]. The COD is the most reliable parameter of tannery wastewater as it represents both organic and inorganic substances present in the wastewater [8]. However, liming wastewater contains a large amount of COD and it is estimated that liming wastewater contributes 54% of total COD in the tannery effluent. Obtained results imply that the amount of COD present in raw liming wastewater was 21783.9 mg/L which was copiously higher than the permissible limit. The coagulant (aluminium sulfate) removes 82.3% COD from liming wastewater which shows more effective results than other coagulants, for example, ferric chloride ($FeCl_3$) and ferrous sulfate ($FeSO_4$) [28]. Though all the pollution parameters are far above the permissible limit; coagulant (aluminium sulfate) could remove a significant amount of pollutants.

The liming sludge produced from liming wastewater in the coagulation process significantly reduces the EC, Cl^-, TDS, TSS, BOD, and COD parameters. The treated wastewater would require further minor treatment before discharge. However, this technique remarkably lessens the pollution load for the treatment of the wastewater in the Central Effluent Treatment Plant (CETP).

3.3. Characteristics of liming sludge and clay

The composition of liming sludge and clay is variable since it depends on the sampling location, treatment methodology, and tanning technology. Table 2 represents the characteristics of the liming sludge and clay sample of this study. The pH of the liming sludge was in an alkaline state (8.3 ± 0.2) whereas the pH of the clay sample was in a slightly acidic condition (5.7 ± 0.3). The liming sludge contains higher moisture content (22.7%) than the clay sample (5.5%). The production of liming sludge from liming wastewater might be responsible for the high moisture content in it. Liming sludge contains organic matter (47.6%) and the possible source of this organic content is dissolved hair, non-structural protein, and other organic substances that are removed from the cowhide and goatskin during liming operation. The metal Cr, Cu, Ni, Pb, and Zn contents in the liming sludge were 108.01, 18.04, 95.7, 14.8, and 86.7 mg/kg, respectively, which are quite similar to other studies [58]. Though Zn, Ni, and Cr were leading metals present in clay samples (63.3, 19.4, and 17.8 mg/kg, respectively), however, these were far below the metal content in the liming sludge. The Cu and Pb contents were almost the same in both liming sludge and clay samples.

Table 2
Physicochemical and metal content in liming sludge and clay

Properties	Liming sludge	Clay sample	Unit
pH	8.3 ± 0.2	5.7 ± 0.3	–
Moisture	22.7 ± 2.67	5.5 ± 1.02	%
Organic content	47.6 ± 0.21	2.6 ± 0.12	%
Cr	108.01 ± 1.3	17.8 ± 1.7	mg/kg
Cu	18.04 ± 3.07	15.6 ± 4.01	mg/kg
Ni	95.7 ± 1.12	19.4 ± 1.2	mg/kg
Pb	14.8 ± 1.7	14.09 ± 1.2	mg/kg
Zn	86.7 ± 0.5	63.3 ± 0.7	mg/kg

3.4. Particle size distribution and Atterberg limit test

Particle size distribution of the raw materials plays a vital role in the bricks' structure and its efficacy. The particle size distribution of the liming sludge and clay is presented in Table 3. The clay is composed of thinner particles compared to liming sludge. The clay & silt percentages in the liming sludge and clay were 49.01% and 74.69%, respectively. Nearly 51% of the particles of the liming sludge were of a size above 75 μm which is correspondingly called 'sand fraction' (d greater than 75 μm). In contrast, the amount of 'sand fraction' in clay is around half of the liming sludge 'sand fraction' (25.31%). The finest particle size distribution in the clay might provide compactness in the fired bricks and in the moistened condition it also helps in increasing the plasticity of the bricks [59].

The properties of fired bricks are sometimes regulated by the moisture content of the raw materials mixture. The addition of water in the raw material mixture replaces the air present in the mixture and after a certain saturation level, the water also occupies the space which was filled by the raw particles [60]. Hence, an optimum amount of moisture content is necessary for the brick manufacturing process that would give a higher degree of compactness for the raw samples. This will help in lowering the water absorption tendency and increasing the compressive strength and bulk density as well. Results presented in Table 4 showed that with the increasing liming sludge content, the optimum moisture content of the liming sludge-clay mixture is gradually increased.

Table 4 represents the Atterberg limit test results. The results indicate that both the liquid limit and plastic limit is gradually increased with increasing the liming sludge percentage in the mixture. The maximum liquid limit and plastic limit of the liming sludge-clay mixture were 48.28% and 41.29%, respectively for the maximum liming sludge percentage (12%) of the mixture. On the contrary, the plastic index was gradually decreased with increasing liming sludge percentage in the mixture, which also points out the lowering of the bonding ability of the mixture. For incorporating 2%, 4%, 6%, 8%, 10% and 12% liming sludge in the mixture, the plastic index of the mixture was 10.01%, 9.48%, 8.14%, 7.6%, 6.94% and 6.49%, respectively. According to Sarani et al. [61], the ideal plastic index range is 7–18% for good quality bricks materials. Therefore, 2%, 4%, 6%, and 8% liming sludge could be used for good quality bricks production.

3.5. XRF analysis

Table 5 summarizes the XRF results of liming sludge and clay in oxide form. The major composition of clay is SiO_2, Al_2O_3, Fe_2O_3, CaO, and MgO with respective concentrations of 64.75%, 10.29%, 5.37%, 4.26%, and 3.59%, which are very much similar to the standard clay sample. The major constituent of liming sludge was SO_3 (30.86%) which is quite dissimilar from standard clay samples. The high SO_3 content might come from the liming process where Na_2S is used as a sharpening agent. At low pH, Na_2S produces toxic H_2S gas which is further converted into SO_3 at atmospheric conditions through a chemical reaction [63]. Unlike standard clay samples, liming sludge contains a high amount of CaO that may appear in the sludge body. The possible source of CaO could be from $Ca(OH)_2$ which is also used in the liming operation to swell up the hide/skin [64]. Rukijkanpanich and Thongchai [65] reported that a large amount of CaO is beneficial for the fabrication of clay bricks because it helps to decrease porosity which will increase the compressive strength.

The concentrations of SiO_2 and Al_2O_3 in the liming sludge were 15.03% and 9.51%, respectively which are far below the standard clay sample. Other oxides viz. P_2O_5, K_2O, ZrO_2, MnO, and Cr_2O_3 are also present in both liming sludge and clay samples but in a trace amount. During the burning process of clay bricks, these oxides help to increase the density as well as assist to obtain better vitrification [66]. Based on the metal content, physical, and chemical composition of liming sludge and clay samples, it is suggested that they are suitable for brick production.

3.6 wt. loss on ignition

The weight loss of bricks on ignition depends on the amount of organic and inorganic materials present in both clay and liming sludge. The percentage of weight loss with respect to liming sludge content is

Table 3
Particle size distribution of liming sludge and clay

Particle size (μm)	Liming sludge (%)	Clay (%)
less than75	49.0	63.7
75–150	24.6	28.1
150–300	11.2	8.2
300–600	7.8	–
greater than1180	7.4	–

Table 4
Atterberg limit test of liming sludge and clay mixture

Parameters	Liming sludge (%)						
	0	2	4	6	8	10	12
Optimum moisture content (%)	6.02	8.01	10.38	13.24	16.73	20.34	24.81
Liquid limit (%)	30.27	31.01	33.88	35.44	43.40	47.22	48.28
Plastic limit (%)	18.69	21.00	24.40	27.63	35.80	40.28	41.79
Plasticity index (%)	11.58	10.01	9.48	8.14	7.61	6.94	6.49

Table 5
Chemical composition of raw materials for brick

Analyte	Chemical composition (%)		
	Liming sludge	Clay sample	Standard clay [62]
SiO_2	15.0359	64.7538	55
Al_2O_3	9.5137	10.2953	30
SO_3	30.8607	2.4462	0
CaO	30.4512	4.2575	1
Fe_2O_3	1.5613	5.3662	8
MgO	1.9646	3.5942	5
P_2O_5	1.6598	1.0958	0
K_2O	0.727	0.0356	0
ZrO_2	0.1211	0.0744	0
MnO	0.0374	0.0195	0
Cr_2O_3	0.1187	0.0251	0

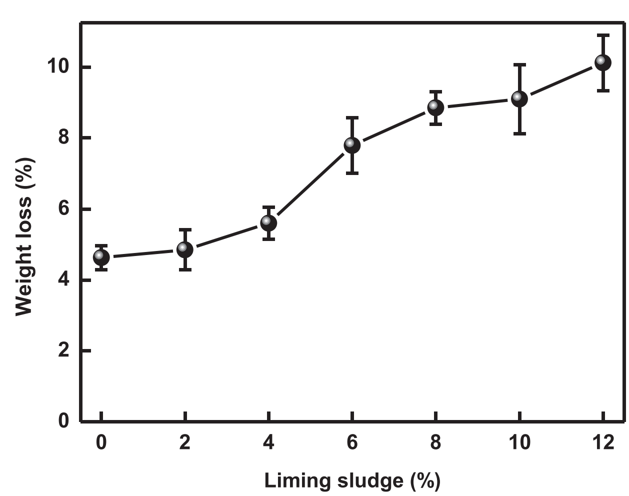

Fig. 2. Percentage of weight loss of liming sludge incorporated fired bricks.

presented in Fig. 2. With the increased liming sludge percentage, the weight loss of brick on ignition also increases. The minimum weight loss was found at 4.63% in clay bricks. The organic content in the clay and sludge was 2.6% and 47.6%, respectively. Since the weight loss significantly depends on the organic content, therefore, the percentage of weight loss increases with the increase of liming sludge percentage in the brick. To maintain the ASTM C62-17 standard of weight loss (≤15%), the percentage of liming sludge in the clay mixture should be carefully maintained.

Dai et al. [67] reported that during brick burning, all organic matter is completely volatilized below 950 °C. The carbonate in the clay and sludge becomes deformed and releases CO_2. A similar kind of phenomenon was observed in several other studies. Making bricks with tannery sludge [4], sewage sludge [68], and paper mill sludge [69] showed a maximum of 16.5%, 14%, and 18.84% weight loss, respectively. In the present study, maximum weight loss was 10.12% which is within the ASTM C62-17 criteria (≤15%). The weight loss could provide additional benefits during the transportation and construction process.

3.7. Area shrinkage

When bricks are burned at elevated temperature it tends to show shrinkage properties to some extent due to the loss of water. Fig. 3 depicts the shrinkage characteristic of bricks incorporated with liming sludge. There is a linear relationship between the percentages of area shrinkage with the addition of sludge. In the control bricks, the lowest amount of area shrinkage is 3.96%. However, with the addition of sludge, the percentage of area shrinkage gradually increased which is analogous with textile sludge [70] and water treatment plant sludge [71]. One possible reason is that since there is more organic matter in the liming sludge, it emancipates more gases during burning, and reduces the final area. On the other hand, it is clear from Table 4 that with the increase of liming sludge percentage the moisture content in the mixture was also increased. During the burning process of the sludge containing bricks, the moisture is completely evaporated and thus the final area of bricks is shrunk. Due to the higher degree of shrinkage, there is a strong possibility of cracks development, deformation of dimensions, and decline in aesthetic appearance [72]. As seen from Fig. 3, the maximum area shrunk for fired bricks containing 10% liming sludge was 5.24% which was below the standard value of 8% [73]. Moreover, in the final brick, there is no sign of crack development or dimensional deformation.

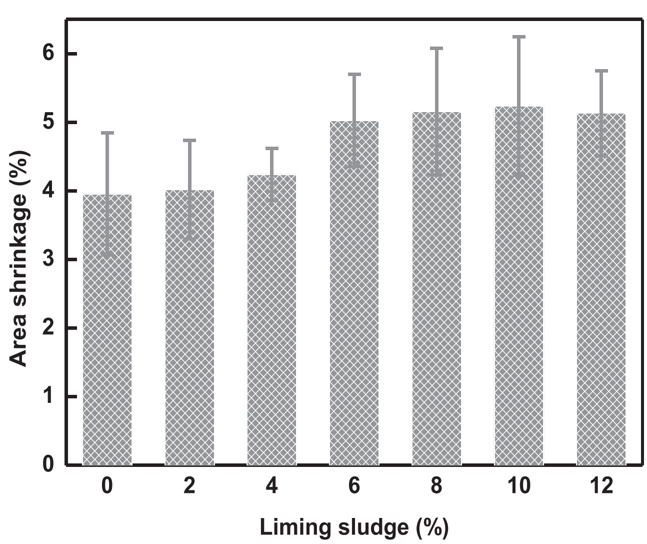

Fig. 3. Area shrinkage of fired bricks with different percentage of liming sludge.

3.8. Bulk density of bricks

Raw materials, burning temperature, and the manufacturing process are the key parameters on which the bulk density of normal clay bricks depends [74]. The bulk density of normal clay bricks varies within 1.8–2.5 g/cm^3 [75]. Fig. 4 demonstrates the bulk density of fired bricks with different percentages of liming sludge. The results stipulated that by increasing the liming sludge percentage, the bulk density of fired bricks gradually diminished. The bulk density of control bricks was 2.53 g/cm^3 which was finally reduced to 1.63 g/cm^3 after adding a fixed percentage of liming sludge. A possible explanation is that there is more hydrate and organic matter in liming sludge which might reduce the bulk density after burning. Making bricks with sewage [76] and tannery sludge [77] showed similar kinds of results.

The reduction of the bulk density might be related to the area shrinkage and weight loss. It can be seen that with the increase of liming sludge percentage in brick, the area shrinkage and weight loss also increase. Conversely, it leads to the reduction of the bulk density of the brick. The reason behind the area shrinkage and weight loss, i.e. the evaporation of the organic matter and moisture possibly lead to the reduction of the bulk density.

3.9. Water absorption

The durability of brick depends on its water absorption capacity. It also represents the sustainability of bricks against the moist environment [78]. The bricks having higher water absorption capacity were found less durable against external weathering conditions [79]. Fig. 5 depicts the effect of liming sludge addition on water absorption property during brick production. The percentage of water absorption varied from 8.63% to 25.12%. For control brick (without liming sludge), 8.63% water absorption was observed but with the addition of 2%, 4%, and 6% liming sludge, water absorption rose at a slow rate up to 9.76%, 10.39%, and 10.46%, respectively. However, with further increase of liming sludge at 8%, 10%, and 12%, the water absorption increased in a faster way to 13.06%, 16.68%, and 25.12%, accordingly. The possible explanation of these phenomena is that generally, liming sludge contains a high amount of organic content; therefore, during firing liming sludge incorporated bricks at high temperature generate pore space which favors water absorption. Singh [79] reported that 10% paper mill sludge incorporated brick showed 17.25% of water absorption capacity. In this study, water adsorption for 10% liming sludge incorporated brick was 16.68%. For severe weathering conditions, water absorption should be less than 17% [80]. Except for 12% liming sludge embedded brick, all the other bricks showed less than 17% water absorption that can be used

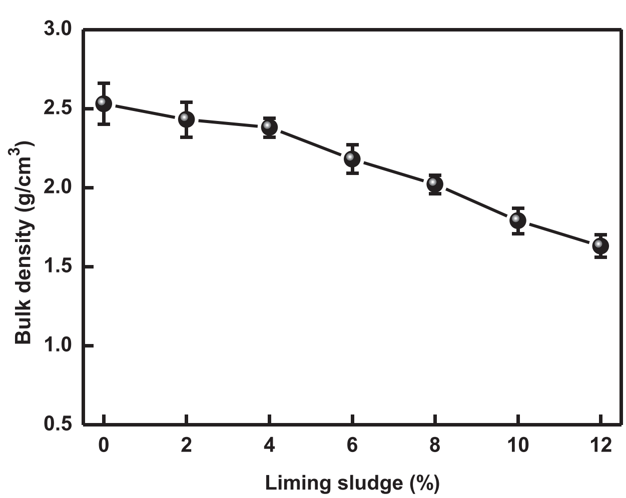

Fig. 4. Bulk density of fired bricks as a function of liming sludge percentage.

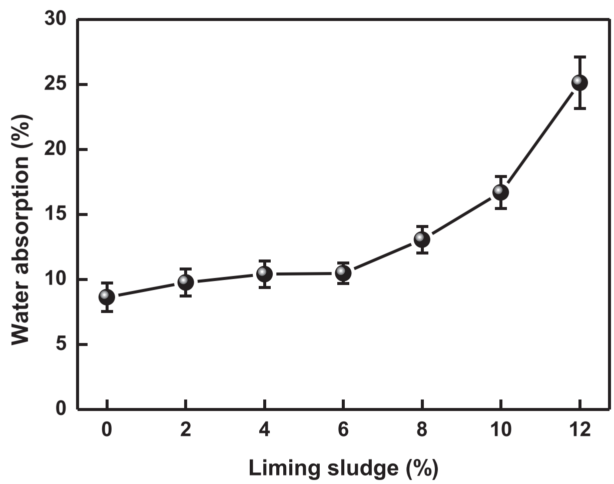

Fig. 5. Water absorption property of liming sludge incorporated fired bricks.

as severe weather-resistant bricks. According to Bangladesh Standard, BDS 2008 [48] brick incorporating 2% liming sludge is considered to be 'S' grade brick, whereas bricks comprising of 4%, 6%, and 8% liming sludge is said to be 'A' grade brick and brick with 10% liming sludge can be regarded as a 'B' grade variety. Several researchers disclosed the upper limit of water absorption at 30% for burned clay bricks [81]. Therefore, liming sludge incorporated brick burned at a high temperature can be used in any kind of weather.

3.10. *Compressive strength*

The average compressive strength value for the liming sludge incorporated fired bricks is shown in Fig. 6. For each combination, three bricks were tested for the average value. The control brick manifested a compressive strength of 10.20 MPa. At the initial stage of introducing liming sludge, the compressive strength increased remarkably and the maximum compressive strength (27.50 MPa) was found by incorporating 6% liming sludge. In contrast to this phenomenon, for other studies like electroplating sludge [67], paper mill sludge [69], water treatment sludge [71], textile sludge [77], arsenic–iron sludge [82], the compressive strength declined when the sludge content increased. The possible explanation is that a certain amount of liming sludge may be able to form a strong bond with the earth metal which exhibits mechanical resistance. Based on what the figure shows, it is clear that when the brick fabricated with more than 6% liming sludge, the compressive strength reduces dractically.

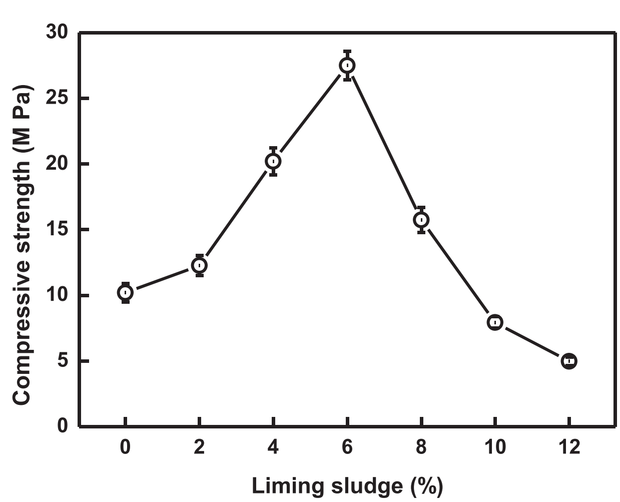

Fig. 6. Compressive strength of liming sludge incorporated fired bricks.

The compressive strength of fired bricks is firmly governed by the characteristics and combination of raw materials [83]. When the percentage of liming sludge is too high, it could not form a strong bond with the clay which might be why compressive strength has fallen. The compressive strength of 7.94 MPa and 4.98 MPa was found for the liming sludge incorporated fired bricks of 10% and 12%, respectively. Hence, the mixing of liming sludge with the clay could play a vital role in attaining maximum compressive strength. The minimum requirement of compressive strength for fired bricks of different building standards varies from 3 MPa to 24 MPa [84]. However, in this study, the lowest compressive strength (4.98 MPa) was shown by the brick specimens that fulfill the minimum requirements of Australian Standards [85]. Considering the Bangladeshi Standards [48] bricks containing 6% liming sludge can be graded as 'S' category; 4% and 8% liming sludge contained in bricks can be categorized as 'A' grade bricks, while bricks made with 2% liming sludge fall into the 'C' category. Based on the analysis, it is seen that the liming sludge-clay mixed brick could form a strong bond when the liming sludge is used in a limited percentage (6%). Therefore, liming sludge containing burned brick could provide sufficient strength that can be used as a construction material.

3.11. *Efflorescence results*

Efflorescence test results describe whether there is any crystalline salt or powdery material deposited on the surface of building materials. Calcium oxide and inorganic salts are the two major components that cause efflorescence and liming sludge contains large amounts of these two components [86]. The efflorescence test result is classified as 'nil', 'slight', 'moderate', 'heavy', or 'serious' as per ASTM C67-08 standard.

The efflorescence is nil if there is no deposition on the surface of the specimens. If less than 10% surface area of the tested brick is covered with a thin layer of salt, it is classified as slight efflorescence. When the deposition is heavier than slight and covers up to 50% of the surface area, it will be moderate efflorescence. While white or gray deposit covers more than 50% of the surface area of the tested bricks it is deemed to be heavy efflorescence. When a thick layer of powdery salt is observed over the exposed surface area of the tested bricks then it is declared as serious efflorescence. Table 6 represents the efflorescence test result for a varying percentage of sludge incorporation bricks. The efflorescence test result of 12% sludge incorporated bricks is classified as "Slight" which means about 10% surface area of the tested brick is covered with a thin layer of salt. As sludge contains inorganic salts that can get dissolved in water and exhibit visible deposits on the surface of the bricks that contain them [4]. However, the efflorescence test result of 0 to 10% sludge incorporated bricks is classified as "nil". This means that the bricks having liming sludge from 2 to 10% can be used as construction materials in any type of climate as there is no sign of cracking, flaking, and powdering on the surface of the tested bricks.

3.12. *FT-IR analysis*

FT-IR analysis was executed to understand and observe the functional groups on the surface of raw materials and fired bricks. FT-IR

Table 6
Efflorescence test results of sludge incorporating bricks

Sludge content (%)	Efflorescence results
0	Nil
2	Nil
4	Nil
6	Nil
8	Nil
10	Nil
12	Slight

analysis of the raw liming sludge, clay, 6% liming sludge incorporated brick, and control brick (only clay) are illustrated in Fig. 7. FT-IR spectra show the bending vibration at 747 and 805 cm^{-1} for clay samples due to the generation of C–H bond which is associated with calcium substitutes like calcium carbide [87]. Two sharp bends appeared at 850 and 1000 cm^{-1} which reveal the presence of the Si-O bond in the clay sample [88]. A band was observed at 1311 cm^{-1} which is marked out as C-O stretching. The band at 3697 cm^{-1} shows evidence that bound water is present in the clay sample [89]. FT-IR results display that the bands range from 949 cm^{-1} to 1170 cm^{-1} for the liming sludge which is imputed to the Si-O stretching band [90]. The band fused at 1354 cm^{-1} was associated with S = O stretching. The intensity of the Si-O band is higher in clay samples compared to liming sludge samples. The bending ranging from 1435 cm^{-1} to 1803 cm^{-1} shows C = O bonds which indicates the presence of calcium carbonate in the liming sludge [91]. The peaks in the zone between 2931 and 3726 cm^{-1} could be employed for O–H bending which indicates that liming sludge is a hydrothermal compound [92]. Compare to liming sludge the clay sample contains more –OH group which indicates that liming sludge is hydrophobic in nature. However, the FT-IR spectrum of both the control and liming sludge containing bricks show the same types of banding at different intervals. The bending at 877 cm^{-1} and 921 cm^{-1} indicates that the C = C bond is present in both the control and 6% liming sludge containing bricks. A sharp bending at 1000 cm^{-1} revealed that both fired bricks and the clay sample contain Si-O-Si stretching. Hafez et al. [93] observed the same types of stretching for the fired bricks made from rice husk waste. For both the control bricks and the 6% liming sludge incorporating bricks the band shifted to 1256 cm^{-1} which indicates that the ratio of CaO/SiO_2 is changed [94]. The C-N stretching is attributed to a band at 1340 cm^{-1}. Due to the low reactivity character of the liming sludge, the band Si-O-Si and Si-O-Al present in the liming sludge containing bricks are weaker than those are presented in control bricks. It has been observed from the FT-IR spectrum that there is no –OH bending for both the fired bricks and it can be concluded that fired bricks do not contain water.

3.13. SEM analysis

The morphological structure of the sintered products wields a great impact on their mechanical properties. Microstructure images of control (without liming sludge) and 6% liming sludge incorporated fired bricks are shown in Fig. 8. The SEM images affirmed significant distinctions in the morphological structure. The control bricks showed a smoother and agglomerated layered structure compared to 6% liming sludge incorporated bricks. Due to this fine agglomeration control bricks showed higher bulk density (discussed in section 3.8). The bricks fabricated with 6% liming sludge revealed a more porous structure compared to control bricks (Fig. 8). The reason behind this porous structure is that liming sludge contains more organic material and moisture content compared to the clay sample [4]. During the burning process, both the organic material and moisture are evaporated leaving behind a porous structure. Moreover, $CaCO_3$ also facilitates the formation of pores to a little extent by decomposing into calcium oxide (CaO) and releasing gaseous carbon dioxide [69]. Due to this porous structure, 6% liming sludge containing bricks showed more water absorption property in comparison with control bricks (discussed in section 3.9). The homogeneous and cohesive structure of the bricks fabricated with 6% liming sludge was also observed from microscopic images. This could be since liming sludge particles are finer than the clay particles; it allows the liming sludge particles to fill the gaps between the liming sludge-clay mixtures, which yield a stronger shape [95]. This could improve the brick's physical properties such as giving it superior compressive strength.

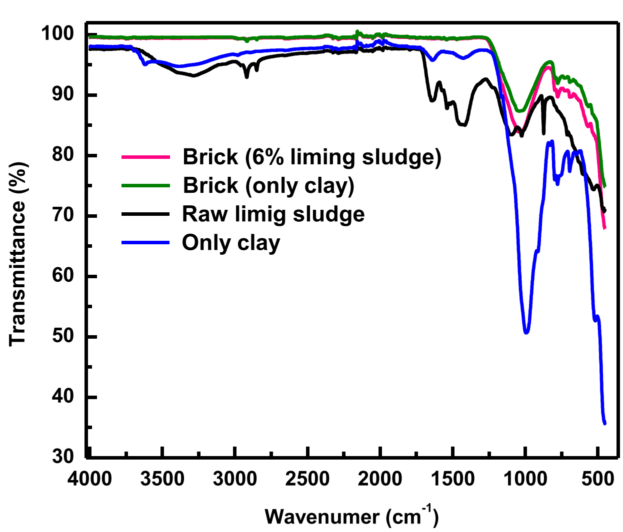

Fig. 7. FTIR analysis of raw liming sludge, clay, lime sludge (6%) incorporated brick, and control brick (only clay).

3.14. Environmental aspect

By carrying out the leaching test the environmental aspect of the fired bricks was analyzed. In this study, we have carried out the leaching test by following two standard methods. The cumulative NEN 7345 leaching test results are presented in Table 7. The maximum leaching concentration was found for Cu, which varied from 0.356 mgm^{-2} to 0.027 mgm^{-2} for 0 to 10% liming sludge incorporated bricks. The maximum concentration of Cr, Pb, Zn, and Ni was 0.048, 0.092, 0.074, and 0.256 mgm^{-2} for 12%, 0%, 2% and 0% liming sludge incorporated bricks, respectively. Results showed that the cumulative leaching value for all the heavy metals was insignificant as the values were far below the maximum leaching value set by Dutch standards U_1 and U_2 (Table 7). It indicates that without any treatment liming sludge incorporated bricks could be used as construction materials for a long period [51]. Cusidó and Cremades [61] also conducted the same leaching test for sewage sludge incorporated bricks.

Table 8 represents the leaching concentration of heavy metal in the liming sludge incorporated bricks which are analyzed by following the USEPA 1311 method. The leached metals Zn, Cu, Ni, and Cd were insignificant as all the values were far below the USEPA regulatory limit. Among the heavy metals, Cr was maximum but within the permissible level. The possible explanation is that the concentration of Cr in the liming sludge was the highest of all the other metals. Judging by this result, it has been observed that with the increased liming sludge percentage in brick, the leachability of Pb gradually decreases. The leachability values of Cr and Pb were higher in the control bricks compared to the liming sludge incorporated fired bricks. Nevertheless, the number of heavy metals leached from the fired brick was far below than it is in liming sludge as well as clay samples. This could be due to the oxidation of heavy metals in the course of the firing process of the bricks at elevated temperatures [96]. Taha et al. [97] and Lin et al. [98] have also observed that during the burning process of liming sludge incorporated into bricks, heavy metals tend to be immobilized. On the other hand, liming sludge and the clay sample contain a higher amount of silica that could amplify the creation of bonding and reduce the metals leachability [99]. However, the amount of leached metals in both leaching test were found far below the respective permissible limit. Although, there was the presence of some heavy metals in raw liming sludge and clay (Table 2), after brick production, the heavy metals did not leach into the acidic solution according to the NEN 7345 and TCLP leaching test. This indicates that the liming sludge-embedded bricks could be widely used without imposing any effect on the environment. Also, it will help to reduce the pollution load caused by the tannery liming sludge.

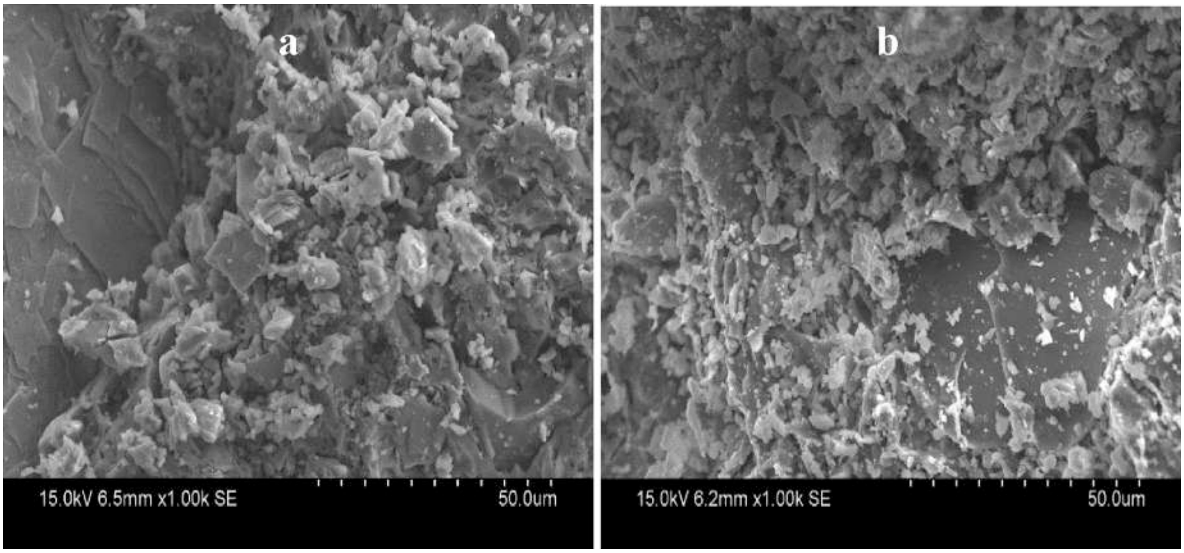

Fig. 8. SEM images of the fired bricks (a) control brick (only clay) and (b) 6% liming sludge incorporated brick.

Table 7
Cumulative leaching test result of liming sludge incorporating bricks as mgm^{-2} according to NEN 7345

Metal	Percentage of liming sludge used in brick							Standard limit	
	0	2	4	6	8	10	12	U_1	U_2
Cr	0.001	0.002	0.004	0.003	0.008	0.034	0.048	150	950
Pb	0.092	0.022	0.024	0.013	0.032	0.037	0.052	100	800
Zn	0.07	0.074	0.071	0.071	0.017	N/D	0.016	200	1500
Cu	0.356	0.348	0.321	0.202	0.189	0.027	N/D	50	350
Ni	0.256	0.212	0.173	0.026	0.003	N/D	N/D	50	350
Cd	0.002	N/D	N/D	N/D	N/D	N/D	N/D	–	–
ND → Not detected									

Table 8
TCLP test results of control and liming sludge incorporating bricks.

Metal	Liming sludge used in brick (%)							USEPA permissible limit	Unit
	0	2	4	6	8	10	12		
Cr	0.480	0.015	0.003	0.030	0.027	0.024	0.021	0.60	mg/L
Pb	0.108	0.018	0.018	0.009	0.036	0.027	0.054	0.75	mg/L
Zn	0.001	0.053	0.002	0.007	0.001	0.001	0.003	4.3	mg/L
Cu	0.010	0.015	0.044	0.020	0.001	0.013	0.001	–	mg/L
Ni	0.074	0.085	0.058	0.133	0.042	0.005	0.037	11	mg/L
Cd	0.016	0.026	0.063	0.071	0.034	0.024	0.018	0.11	mg/L

3.15. Implications of the study

The proposed investigation was carried out utilizing the liming sludge produced during liming operation in the tannery. Paul et al. [100] suggested that annually 85,000 tons of wet salted hides and skins are processed to produce finished leather. A report by FAO [101,102] showed that every year 34.4 and 27.6 thousand tons of wet salted cowhides and goatskins, respectively, are treated to produce leather. According to the estimation of this study, every year 3.4 × 10^3 MT and 8.2 × 10^3 MT lime sludge are generated during liming of wet salted cowhide and goatskin, respectively. On average, annually 11.6 × 10^3 MT liming sludge is produced exclusively from the liming operation. In general, the liming wastewater is combined with other wastewaters from tannery operations. As a result, the combined wastewater and combined sludge contain toxic heavy metals like Cr which enters mainly from chrome tanning operation. Due to the lack of proper management, this combined sludge has negligible commercial application as it requires a separate treatment process to remove heavy metals. The present study approaches separating the liming sludge from the combined sludge with the benefit of the absence of heavy metals and 60–70% reduction of the total pollution load of the tannery [28]. Moreover, if properly used, this frowned upon and concerning solid waste could produce more than 38 million bricks and reduce the clay load by 17.48%.

4. Conclusion

A novel eco-sustainable way was executed by utilizing tannery liming sludge in the production of eco-friendly construction bricks. The major consequences of the investigation can be encapsulated as follows:

- The engineering properties of fired bricks like percentage of area shrinkage, percentage of weight loss, and percentage of water absorption proportionally increased with the increase of sludge percentage but according to ASTM and BDS requirements, all the values were within the acceptable range.
- Although all the compressive strength values of the fired bricks fulfill various nations' construction industry requirements but 6% liming

sludge amended bricks provided with the maximum value of 27.50 MPa emerging the best possible recipe for liming sludge amended bricks.

- Although raw liming sludge contains a considerable amount of heavy metals, two leaching tests revealed that the leaching concentration of heavy metals from the liming sludge incorporating fired bricks was negligible and far below the two regulatory standard limits.

The results suggest that liming sludge could be used as raw materials for good quality brick production that could satisfy all the standard requirements. Therefore, brick industries could exploit the liming sludge as raw material for brick fabrication leading towards a fruitful way of waste management in the tannery.

Funding

This research did not receive any specific grant from funding agencies that operate in the public, commercial, or not-for-profit sectors.

CRediT authorship contribution statement

Md. Anik Hasan: Visualization, Conceptualization, Investigation, Methodology. **Md. Abul Hashem:** Data curation, Supervision. **Sofia Payel:** Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors greatly acknowledge Superex Leather Limited, Khulna, and SAF Leather Industries Limited, Jashore, Bangladesh for providing the liming wastewater for conducting the research work. Authors are also thankful to Mr. Abu Hasan, Leather Technologist, SAF Leather Industries Limited, Jashore, Bangladesh, and Mr. Md. Imamul Vuian, Proprietor, Super Bricks Limited, Jashore, Bangladesh for their assistance.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.conbuildmat.2021.125702.

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