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Valorization of stabilized river sediments in fired clay bricks: Factory scale experiment

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ABSTRACT

The objective of this study is to demonstrate the practical use of polluted river sediments after treatment into brick production. Consequently, a full-scale industrial experiment was conducted at a brick factory in the north of France. Polluted sediment was stabilized by the Novosol® process and then was introduced in the mix-design with a substitution ratio of 15% as a partial replacement of quartz sand. Approximately 15,000 perforated sediment-amended bricks were produced. The produced bricks were then subjected to several qualification tests (compressive strength, freeze and thaw resistance, water absorption). The results obtained showed that the substitution of quartz sand by treated sediment resulted in a significant increase in brick compressive strength and firing shrinkage, and in a decrease in porosity and water absorption. Moreover, leaching tests performed according to different standards on substituted brick samples showed that the quantities of heavy metals leached from crushed bricks were within the regulatory limits. Thus substituted bricks can be regarded as non-hazardous material.

1. Introduction

Over the next 15 years, a volume of 3 million m^3 of river sediments is to be dredged in the north of France in order to maintain a normal river activity in this region [1]. An important part of these sediments is actually polluted. This pollution is linked to the industrial history of the region where certain activities, like iron and steel industries, metallurgy of nonferrous ores and energy sectors remain polluting. Contamination is mainly due to organic (PAHs, PCBs, TBT and dioxins) and inorganic (heavy metals: lead, chromium, zinc, copper) pollutants, which can end up in drains, rivers, and coastal waters, thus contaminating water resources, soils and polluting the environment. Dredged material has to be managed and since international and European laws have become more stringent, its management has become an environmental and economical concern for a large number of countries [2]. Various alternatives to the disposal of the processed material have been investigated like sea deposit, landfilling and treatment processes. The effect of disposal in open-water has been largely studied [3,4]. Landfilling requires large spaces and long-term monitoring; however, it is less accepted by the public opinion (NIMBY). On the other hand treatment processes permit a reduction in toxicity and volume of dredged material, but in comparison with open-water and upland disposal, the treatment cost is not yet competitive enough [5]. This underlines the necessity to find ecological valorization paths for processed material to make these alternatives economically competitive. Moreover, the beneficial use of polluted sediments offers a practical contribution to maximising the conservation of traditional brick-making raw materials. The heterogeneity of clay-based materials accommodates a variety of waste materials, thus the incorporation of industrial wastes in bricks and tiles is becoming common practice [6,7]. In recent decades several types of waste materials have been assessed as raw material for brick making, for example lightly contaminated harbour sediments [8–10], waste bricks [11], limestone dust and wood sawdust [12], processed waste tea [13], reservoir sediments, mixed with fly ash [14], dried sludge collected from industrial wastewater treatment [15–17], incinerated sewage sludge ash [18–20], fly ash [21], granite sawing waste material [22], water treatment residual with excavation waste soil [23] and steel dust [24]. Considering their perpetual availability, particle sizing and their chemical composition, sediments are regarded as a suitable raw material for brick production.

The application of polluted river sediments as a partial replacement material in brick making has been previously investigated. Indeed, during the last 6 years our laboratory has realised a large number of experiments in order to study the feasibility of using polluted river sediments, after treatment, in construction materials (mortar, road materials, clay bricks). In brick making, treated sediment (TS) was used as a partial sand and clay substitute. It has

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Table 1
Total concentrations of heavy metals in raw river sediments in mg/kg on dry material

Element	Cadmium (Cd)	Chromium (Cr)	Copper (Cu)	Lead (Pb)	Zinc (Zn)
Raw sediment	12.8	413	150.7	1373	5032
Level N1	1.2	90	45	100	276
Level N2	2.4	180	90	200	552

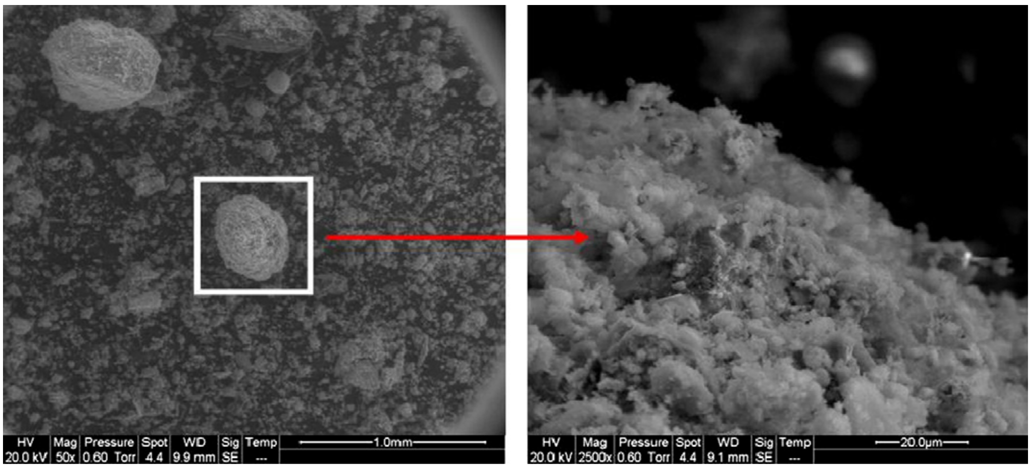

Fig. 1. SEM photomicrograph of a typical particle of treated river sediments.

been constructively and successfully incorporated into bricks with different proportions ranging from 25 to 45% (dry basis). The mix-designs (sediment, water, and natural clay) were first dry-blended in a Z-blade mixer. After tempering, the four (0, 25, 35 and 45%) plastic bodies were formed into test specimens using a laboratory extruder. A vacuum machine was used to expel the air from the mixture to avoid cracking during firing. Brick specimens were firstly dried off in a tunnel dryer and then fired in the tunnel kiln of the local brick factory. The results show that the sediment proportion in the mixture has had an important impact on the quality of the brick. As an example, the increase in sediment proportion resulted in a decrease in compressive strength, but the latter of all sediment-amended brick samples was still comparable to that of standard bricks. The substituted bricks have successfully passed the different tests required by French Standards in order to assess the suitability of a brick to be used in construction, for example: it was observed that the weight losses for all substitution ratios are less than 1% (the limit given by the French Standard). In addition, neither cracking, nor breakage occurred on all the specimens tested. Moreover, after drying, samples were carefully examined and no efflorescence was observed for all tested specimens. Considering physical, mechanical and chemical results, the 35% ratio of substitution of treated sediments in bricks was selected to be the most effective one in laboratory scale. For more details see [25]. Subsequently, a factory proving experiment was carried out at the Briqueterie du Nord (BdN) to confirm their full-scale practical use as a brick raw material. The current paper presents and discusses the results obtained. In the first part a characterisation of treated sediment is introduced. Then, the manufacturing process of the BdN brick factory is described. Finally, the results of qualification tests undertaken on sediment-amended bricks are presented compared to those of the standard one.

2. Materials and methods

2.1. Characterisation of sediments

Sediments in this study come from Dampremy-Charleroi region (Belgium). This area is marked by several industrial activities like coal mining, iron and steel industry, glassworks, chemicals and electrical engineering which explains the high heavy metal concentrations in raw sediment.

Table 1 gives the concentrations of heavy metals in raw sediments, where five metal species of daily concern were selected (cadmium, chromium, copper, lead, zinc). French levels of reference, given by the Official Journal [26], are also reported in this table. Below level N1, the potential impact is regarded, in principle, as neutral or negligible. Between levels N1 and N2, further investigations may prove necessary depending on the project considered and on the extent to which action level N1 is exceeded. Beyond N2 level, additional investigation is generally necessary since significant indices suggest a potentially harmful impact of the operations

Table 2
Concentrations of main heavy metals leached out of treated sediments according to French Standards (mg/kg on dry material)

Element	Treated sediment pH 11.4	Limit values for waste acceptable as inert $L/S = 10$ (l/kg)	Limit values for waste acceptable as non-hazardous $L/S = 10$ (l/kg)
Cd	<0.01	0.04	1
Cu	<0.1	2	50
Zn	0.18	4	50
Ni	<0.08	0.4	10
Pb	<0.2	0.5	10

Table 3
Concentrations of heavy metals leached out of TS according to the TCLP standard procedure (mg/kg on dry material)

Element	Treated sediment pH 4.65	Regulated TCLP limit
Cd	0.31	1.00
Cu	0.76	15
Zn	13.8	25.00
Ni	0.9	–
Pb	1.65	5.00

Table 4
Mineralogy of treated sediments and standard brick feedstock (sand 1, 2 and natural clay)

Treated sediment	Sand 1	Sand 2 (coarse sand)	Natural clay
Quartz: SiO_2	Quartz: SiO_2	Quartz: SiO_2	Quartz: SiO_2
Hematite: Fe_2O_3	Gypsum: $CaSO_4{\cdot}2H_2O$	–	Clay minerals (<2 μm): Smectite: 66% Illite: 16%
Calcite: $CaCO_3$	–	–	Kaolinite: 12% Chlorite: 6%
Feldspar: anorthite $(Ca,Na)(Si,Al)_4O_8$	Feldspar: anorthite $(Ca,Na)(Si,Al)_4O_8$	–	Feldspar: albite $NaAlSi_3O_8$
Mica: muscovite $KAl_2(Si_3Al)O_{10}(OH,F)_2$	–		Mica: muscovite $(K,Na)Al_2(Si,Al)_4O_{10}(OH)_2$
Glassy phase	–	–	–

[27]. From Table 1 we can observe that raw sediments exhibit high concentrations of heavy metals that largely exceed level N2. As an example the concentration of Zn (5032 mg/kg) is approximately ten times higher than N2 level, which is equal to 552 mg/kg. These results emphasis that polluted raw sediments have to be treated before being valorized. Moreover, raw sediments contain ≈15% (on dry basis) of organic matter, thus to avoid the uneven surface texture of bricks, they need to be calcined before being introduced into brick production. Thus, raw sediments were subjected to a preliminary treatment using the Novosol® process developed and patented by the Solvay Company. It is based on the stabilization of heavy metals in the solid matrix by phosphatation and the destruction of organic matter by calcination. During the phosphatation phase, raw sediments are mixed with phosphoric acid H_3PO_4 (2–3.5%) in a tubular reactor. The addition of phosphoric acid allows, in the presence of calcite, the formation of calcium phosphates minerals.

These minerals are known for their low solubility and their ability to fix heavy metals. The calcination phase consists of calcining the phosphated sediments at ≥650 °C in a rotary kiln, in order to break down the organic matter (polycyclic aromatic hydrocarbons, dioxins and pesticides). It increases the product toughness, reduces

Table 5
Composition of the BdN-standard Brick and the Factory Trial mix-designs (w% on dry basis)

Product	BdN-standard mix-design	Sediment-amended mix-design
TS	0	15
Sand 1	20	0
Sand 2	5	5
Natural clay	75	80

the volumes of processed materials after treatment and allows a better stabilization of metal phosphates. On the other hand, gaseous emissions resulting from the treatment process (essentially H_2S and CO_2 and traces of heavy metals) are chemically treated using activated charcoal and sodium bicarbonate. For more details concerning this process, see [28–30].

Treated sediment can be described as an odourless, fine grained powder of a particle density (≈2.8 g cm^{-3}) comparable to that of clay (2.6–2.7 g cm^{-3}). The granules making up the bulk material are generally angular to round in shape and composed mainly of agglomerates of fine sintered particles with a specific surface area (BET ≈ 6 m^2 g^{-1}), which gives them the ability to absorb a lot of water (Fig. 1).

Heavy metal leaching was performed in accordance with two different procedures: the French Standard [31], where TS were leached out using distilled water and the American Toxicity Characteristic Leaching Procedure (TCLP-USEPA 1986) [32], where acetic acid was used as leaching solution. Metals (Al, Ca, Fe, Mg, Ti, Cr, Cu, Mn, Sr, V, Pb, and Zn) in the leaching solutions were determined with a Varian Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES). The results are given in Tables 2 and 3 together with the limits given by the Commission of the European Communities [33] and the TCLP limits.

From Tables 2 and 3 we can observe that leached metal concentrations of the TCLP test are higher than those of the French Standard procedure, because of the higher pH values of the latter. The values of the five selected metal species (Cd, Cu, Ni, Pb, and Zn) are largely below the regulatory limits for both tests, thus treated sediments can be considered as non-hazardous waste. As an example for Zn, the metal of most concern in terms of leaching test, values of 0.18 and 13.8 mg/kg were found for the two tests respectively. However, these values are largely inside the regulated limits (4 and 25 mg/kg respectively).

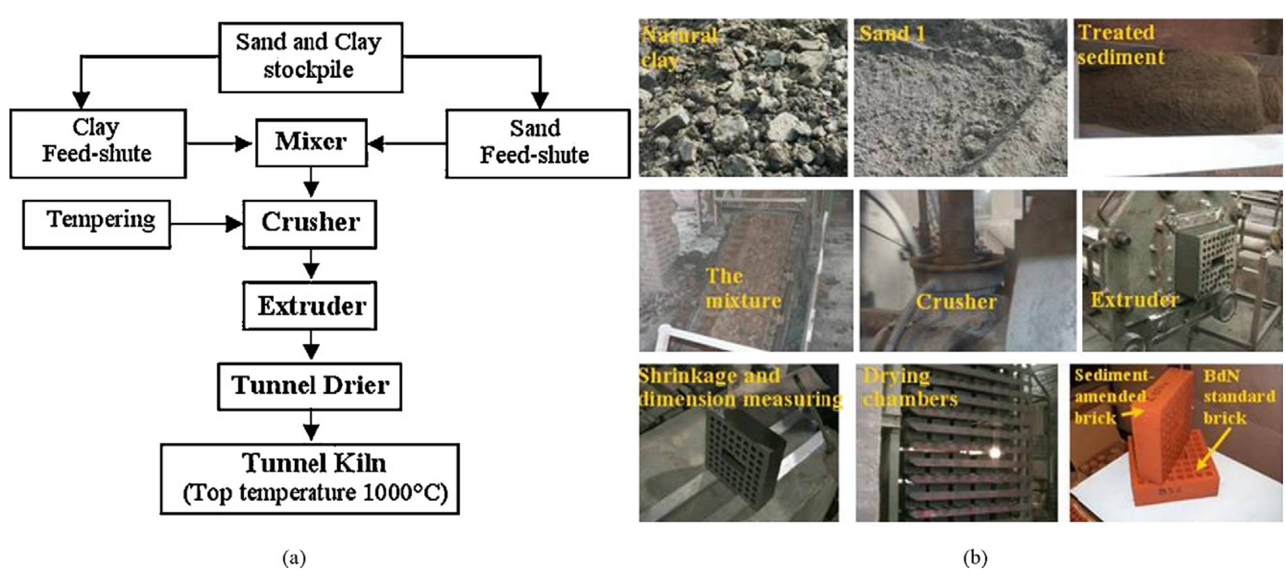

Fig. 2. Process line at the brickwork: (a) flow diagram; (b) photos.

Mineralogical analysis was undertaken using a Philips PW 1730 diffractometer. The results of the X-ray diffraction (XRD) are given in Table 4. They show that TS are composed mainly of quartz which gives them the possibility to fulfil the same role as the quartz sand. They also reveal a marked presence of iron oxide, calcite with some traces of feldspar, mica and sulphates. The presence of hematite gives an encouraging support for the beneficial use of these sediments in brick making, as iron is recognised to possess good fluxing properties [18,19].

The influence of TS addition on the plastic properties of the mixture has been presented in earlier publication [25], where it has been stated that the plasticity index is inversely proportional to the amount of added sediments, thus the addition of treated sediments lowers the plastic nature of the mixture and decreases its bonding ability.

2.2. Factory brick-making process

The Briqueterie du Nord is a participating partner in this study. It operates a brick factory in the Nord-Pas-de-Calais region (North of France). Its annual production reaches 45,000 t of bricks per year. In its brick-making operation the BdN uses tow types of sand (1, 2) and natural clay. The XRD (Table 4) shows that these sands are composed mainly of quartz; while for natural clay a marked presence of quartz, mica and feldspar was detected and the clay fraction (<2 μm) is composed mainly of Smectite (66%). The progressive stages of brick manufacture are illustrated in Fig. 2. Sand and clay are removed from the stockpile and discharged into a feed-shute, where they are mixed. The mixture then is carried by conveyor-belt to a crusher consisting of a rigid base over which tow circular crushing rolls continually revolve. The tempering process takes place at this stage, where additional water is added to transform the mixture into an adequate plastic state after which it passes into a vacuum extruder. The rectangular column that emerges from the extruder is subsequently separated into individual bricks by a wire-cutter that slices vertically through the column. The formed bricks are then stacked into cars and passed through a tunnel drier. The dried bricks then pass through a tunnel kiln where they reach a maximum temperature of 1000 °C. The drying and firing programs are given in Figs. 3 and 4.

Details of the BdN-standard and the sediment-amended mix-designs are given in Table 5. We note that sand 1 in the sediment-amended mix-design was substituted by treated sediment introduced with a ratio of 15%. However the results of this experiment will be used to guide other trials where a higher quantity of treated sediment will be valorized.

A granular distribution was carried out on TS and on the BdN feedstock (sand and natural clay). The results are shown in Table 6 together with the granular distribution of the BdN-standard mix-design and the sediment-amended one.

In particle size TS can be considered as silty sand. The BdN-standard mix-design is seen to be somewhat coarser when compared to the sediment-amended one, where 78.15% of particles are <62.5 μm).

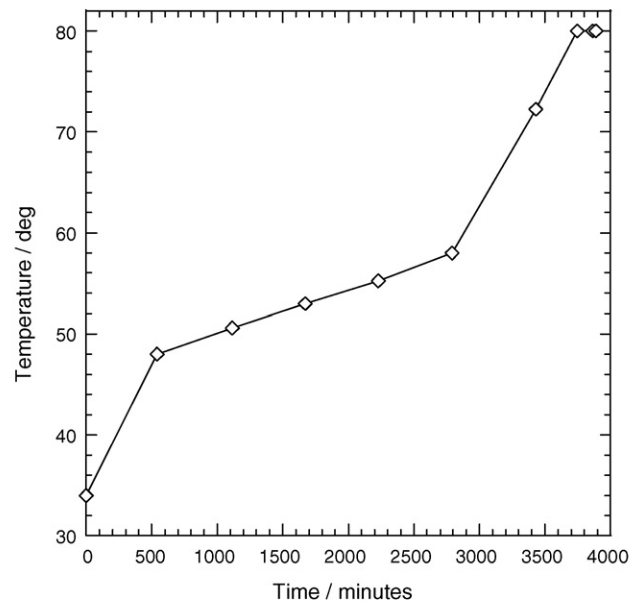

Fig. 3. Drying program of bricks.

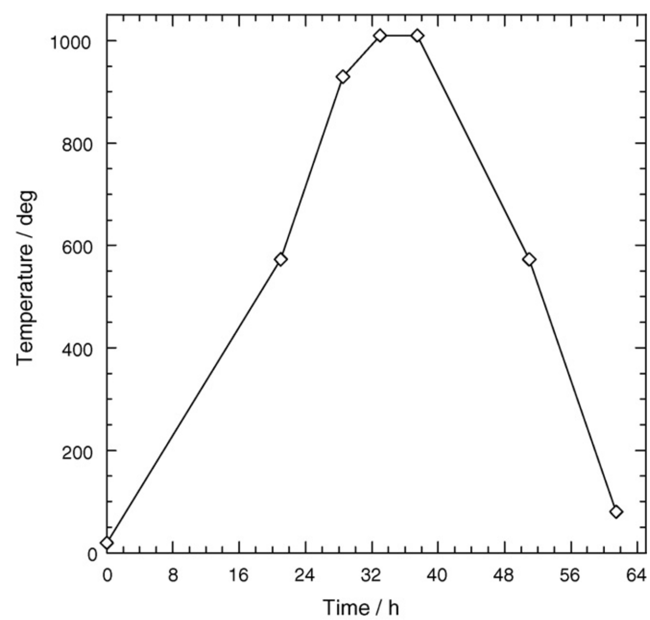

Fig. 4. Heating program of bricks.

3. Results and discussion

Treated sediment was introduced into the manufacturing process without any alteration to the existing process lines. The sediment-amended mix-design required approximately 2% more water to achieve a level of plasticity comparable to that of the stan-

Table 6
Comparative particle size analysis of TS, sand 1 and 2, natural clay, BdN-standard Brick and Factory Trial mix-designs (w%)

Granular distribution	Treated sediment	Sand 1	Sand 2	Natural clay	BdN-standard mix-design	Sediment-amended mix-design
Coarse sand: >500 μm	15	5	15	–	1.7	3
Medium sand: 250–500 μm	13	10	40	–	3.9	3.95
Fine sand: 62.5–250 μm	32	77.8	44.5	9.8	25.8	14.9
Silt: 2.5–62.5 μm	38	7.2	0.5	82	62.3	71.3
Clay <2.5 μm	2	–	–	8.2	6.3	6.85

Table 7
Firing shrinkage of tested bricks

Product	% Fired shrinkage
BdN-standard brick	7
Sediment-amended brick	10

Table 8
Compressive strength (MPa)

Mix-design	BdN brick	Sediment-amended brick
Average compressive strength	22	36

Table 9
Permeability values of the BdN-standard and Factory Trial bricks

Product	Permeability (m/s)	Intrinsic permeability (m^2)
BdN-standard brick	9.5×10^{-7}	8.75×10^{-14}
Sediment-amended brick	7×10^{-8}	6.5×10^{-15}

Table 10
Weight loss in brick specimens (%)

Mix-design	BdN-standard brick (25 cycles)	Sediment-amended brick (50 cycles)
Average weight loss	0.13	0.36

dard brick mix-design. No difficulties were reported either at the mixer or at the extrusion stage of the process line. Approximately 15,000 perforated bricks (6 cm × 22 cm × 22 cm) were fired through the factory tunnel kiln during this trial. The choice of this type of brick was based on their high selling rate.

Once out of the kiln white spots and grains were observed on their surface (Fig. 5). To study the nature of these spots a scanning electron microscopy (SEM) was carried out on brick samples (Fig. 6). It revealed the presence of gypsum ($CaSO_4·2H_2O$) and barite ($BaSO_4$). This occurs due to soluble salts (especially sulphates) contained in TS, which migrate to the brick surface during drying and subsequently persisting as white spots on the fired product. The presence of white grains is linked to the marked presence of calcite in treated sediments (Fig. 7). In fact, calcite with grain size <1 mm reacts with silica and alumina, deriving from already dehydroxylated phyllosilicates (grain-boundary reaction between CaO, Al_2O_3 and SiO_2) to form new high-temperature phases such as gehlenite and anorthite. The XRD analysis, carried out on sediment-amended brick samples, revealed the presence of anorthite within these samples [34]. When calcite grain size is >1 mm, it transforms into lime (CaO) and remains within the fired body [35–37].

A representative sample batch of this trial underwent a series of tests including compressive strength, freeze and thaw resistance, water absorption, permeability, porosity and heavy metal leaching to determine the properties of the new material. The results are summarised in Tables 7–12 compared with the BdN-standard brick.

Table 12
Concentrations of heavy metals in the leachates of brick samples leached with acetic acid in mg/kg on dry material

Element	Sediment-amended brick pH 4.92	BdN-standard brick pH 4.97	Regulated TCLP limit
Cd	<0.04	<0.04	1.00
Cu	0.1	0.2	15
Zn	3.7	3.3	25.00
Ni	<0.14	0.67	–
Pb	<0.4	<0.4	5.00

3.1. Firing shrinkage

The firing shrinkage was measured for 10 bricks of each mix-design. The results are given in Table 7. We can observe that the firing shrinkage of the sediment-amended brick is higher than that of the BdN-standard one. This is due to the fact that the BdN-standard mix-design is coarser than that of the sediment-amended one (Table 6), where 20% of sand 1 (composed mainly of quartz) was used which increases the expansion coefficient of the body and thus decreasing the linear shrinkage [38]. Since the sintering rate is proportional to the particle size [39,40]; the trial mix-design is expected to undergo more important sintering than the BdN-standard one resulting in more shrinkage. On the other hand the sediment-amended mix-design required approximately 2% more water to achieve a level of plasticity comparable to that of the BdN-standard one. This is due to the porous microstructure of TS which gives them a high specific surface area. Thus, they tend to soak up and hold a proportion of the water added at the mixing stage, which cause more shrinkage during drying.

3.2. Porosity and water absorption

Porosity and water absorption were measured according to the procedure proposed by Khalaf and DeVenny [41]. Test sample (at least 100 g of brick lumps) was obtained by smashing 5 full-size bricks. The lumps of crushed bricks were mixed and sieved on 20 and 14-mm sieves. The fraction passing the 20-mm sieve and being retained on the 14-mm sieve was kept. Brick lumps were oven-dried (105 °C) for 24 h. The dry mass was determined to an accuracy of 0.1%. The test sample was placed in a desiccator and air was removed using a vacuum pump, operating at a pressure of 0.07 bar, for 0.5 h. Porosity and water absorption by 24 h submersion in cold water, were determined using Eqs. (1) and (2) respectively:

$$\text{Porosity}\,(\%) = \frac{B-A}{B-C} \times 100 \tag{1}$$

$$\text{Water absorption}\,(\%) = \frac{B-A}{A} \times 100 \tag{2}$$

where A, B and C are: the dried mass, the saturated mass and the mass in water.

The results of porosity and water absorption are given in Figs. 8 and 9. They indicate that the BdN-standard bricks are more porous than sediment-amended ones. Indeed, at lower temperature (up to 800 °C), porosity closing and pore elimination is due

Table 11
Results of the leaching test undertaken on brick specimens in accordance with the French Standard in mg/kg on dry material

Element	Sediment-amended brick pH 8.9	BdN-standard brick pH 7.6	Limit values for waste acceptable as inert $L/S = 10$ (l/kg)	Limit values for waste acceptable as non-hazardous $L/S = 10$ (l/kg)
Cd	<0.02	<0.02	0.04	1
Cu	<0.03	<0.03	2	50
Zn	0.053	0.177	4	50
Ni	<0.07	<0.07	0.4	10
Pb	<0.2	<0.2	0.5	10

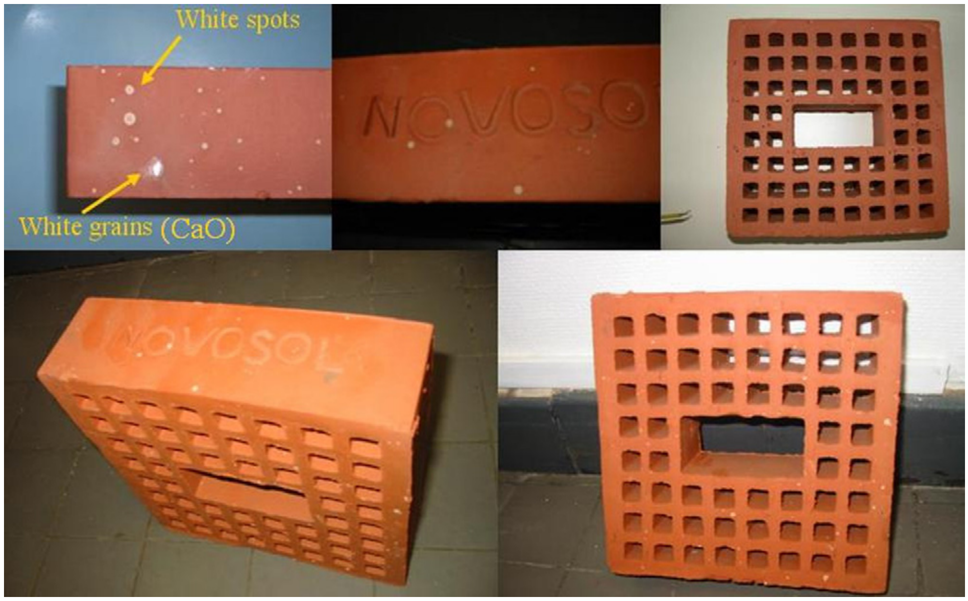

Fig. 5. Some examples of produced sediment-amended bricks.

to diffusion mechanism during solid state sintering. Since sintering rate is proportional to particle size, sediment-amended bricks are expected to undergo a higher sintering than the standard ones resulting in a higher reduction in porosity. At high temperature (>900 °C) the additional reduction in porosity is due to vitrification, where smaller pores between clay particles disappear as a result of melting and coalescence of particles and the phyllosilicate surfaces become smoother and the pores become ellipsoid with smooth edges (Fig. 10).

BdN-standard brick exhibit a higher water absorption coefficient than sediment-amended one since this parameter is directly related to porosity.

3.3. Compressive strength

The compressive strength test was undertaken on 10 full-size bricks of both mix-designs in accordance with the procedure as described in the French Standards [42,43]. The average results are given in Table 8.

From Table 8 one can observe that the average compressive strength of the sediment-amended brick is 63% higher than that of the BdN-standard one. This is due to the substitution of coarse quartz sand (sand 1) by TS, resulting in a finer mix-design. Thus, sediment-amended bricks are better sintered than the BdN-standard ones. This resulted in a denser microstructure, as confirmed by the density measurements, where values of 1.99 and 2.05 g cm^{-3} were obtained for BdN-standard bricks and sediment-amended ones respectively. In addition, the compressive strength is directly related to porosity and it has been stated that the porosity of the standard brick is higher than that of the sediment-amended one, which induces a decrease in its compressive strength. On the other hand, quartz with particle size in the range of (10–30 μm) improves the mechanical strength, while large-size quartz particles tend to decrease it. This is due to the susceptibility to micro-cracks formation due to the quartz allotropic transformation at a temperature around 573 °C, which induces in a volumetric change (TV = 1%) that exerts a tensile stress on the surrounding matrix prior to full densification [44–46]. This results in debonding between the quartz grains and the matrix generating a decrease in mechanical strength. A laser granular distribution undertaken on the fraction <62.5 μm of TS (Fig. 11) showed that the volume of particles with a diameter <30 μm was equal to 50%, while in sand 1 the silt fraction was equal to 7%.

3.4. Permeability test

This test is not required for the qualification of bricks. Nevertheless, permeability is an important factor of durability. High permeability facilitates the ingress of water into pore structure and accelerates the deterioration of the brick due to repeated freeze and thaw cycles. The test sample was placed in a cell and was subjected to a confining pressure of 6 MPa. Then a liquid (distilled water) was injected with a constant pressure of 1.5 MPa using a pump. The flow rate of the injected fluid was measured and the permeability was determined using the Darcy's law. The results are given in Table 9.

We can note that the BdN-standard bricks are more permeable than the Factory Trial bricks. This is due to the presence of quartz with a particle size >30 μm which make the brick sample susceptible to micro-cracks formation as a consequence of expansion accompanying quartz transformation (573 °C). Thus the quartz, as a non-plastic material, decreases the plasticity, facilitates the deflocculation and increases the permeability of the brick. On the other hand there is a correlation between the compressive strength and the permeability and porosity results, thus the higher permeability of the BdN-standard bricks generates a decrease in its mechanical strength.

3.5. Freeze and thaw resistance

The frost resistance test was undertaken on 10 specimens of each mix-design. The BdN-standard Brick samples were subjected to 25 cycles of freezing and thawing according to the procedure as described in the French Standard [47], while the Factory Trial ones were first subjected to 25 cycles and then to another 25 cycles to determine their limit resistance. After the completion of the test, specimens were placed in open air for 24 h and then oven-dried at 105 °C, so that the specimen's weight loss could be determined. The results are shown in Table 10.

We can observe that the average weight losses for all brick samples are less than 1% (French Standard limit). In addition, neither

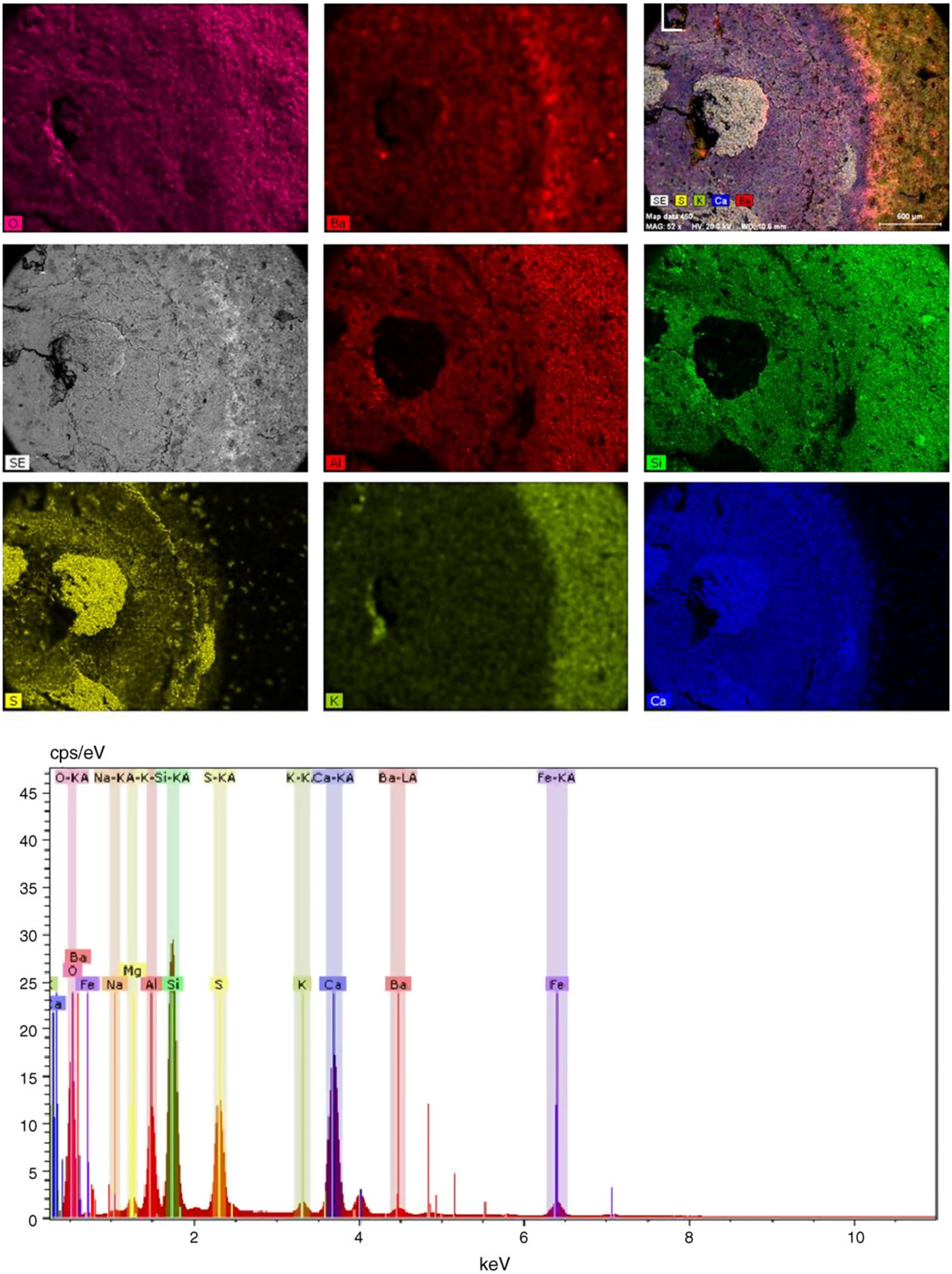

Fig. 6. The nature of white spots and white grains on the surface of sediment-amended bricks.

cracking, nor breakage occurred on all the specimens tested. Thus we can conclude that all specimens have successfully passed the test of qualification of bricks.

3.6. Heavy metal leaching

Treated sediment contains significant amounts of heavy metals. To evaluate the subsequent risk of their leaching out of the final product, two different leaching tests were performed.

3.6.1. French Standard

In masonry, the pH value of the run-off water is between neutral and alkaline due to the presence of mortar. Thus the French procedure is expected to give a good simulation of these conditions. Table 11 presents the average values of a leaching test carried

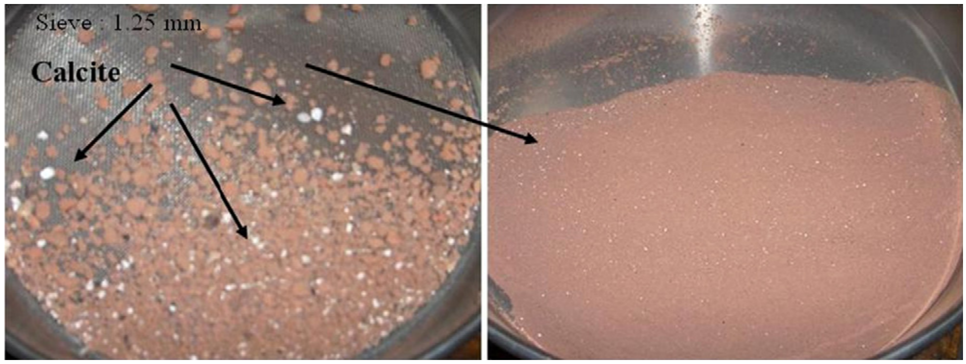

Fig. 7. The presence of calcite in TS.

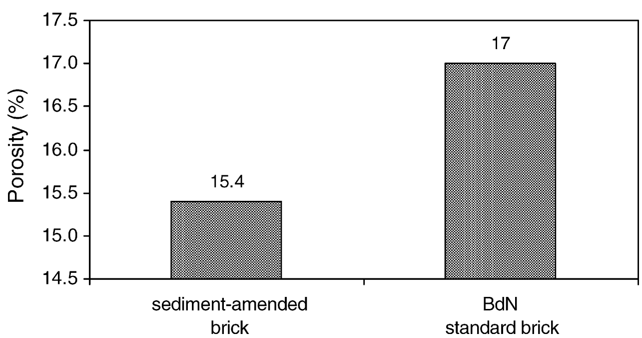

Fig. 8. Porosity values of sediment-amended and BdN-standard bricks.

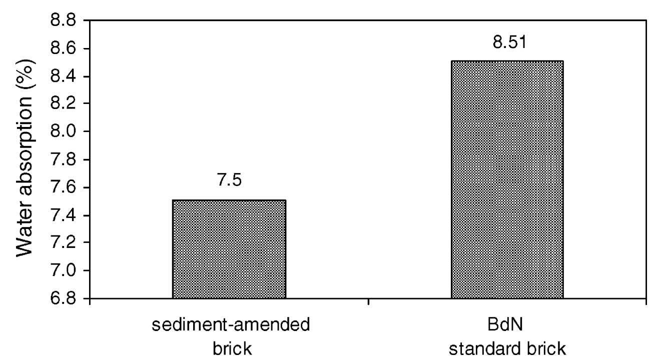

Fig. 9. Water absorption values of sediment-amended and BdN-standard bricks.

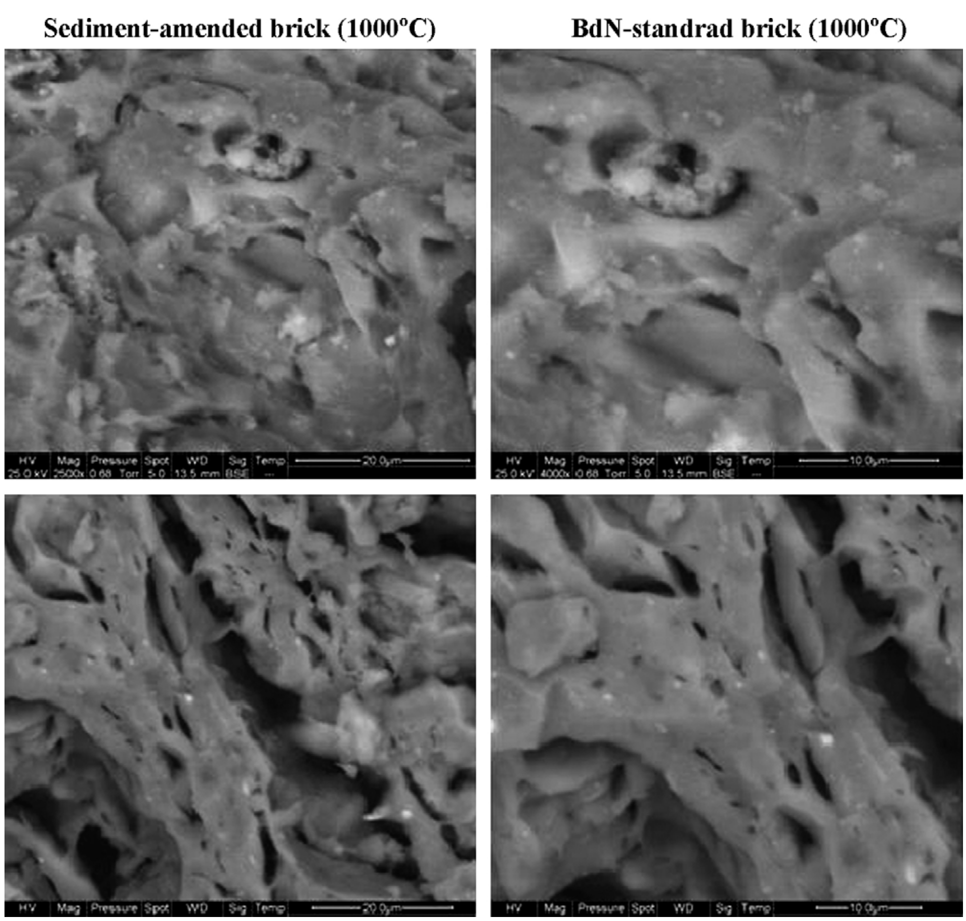

Fig. 10. SEM photomicrographs of sediment-amended and BdN-standard brick samples fired at 1000 °C.

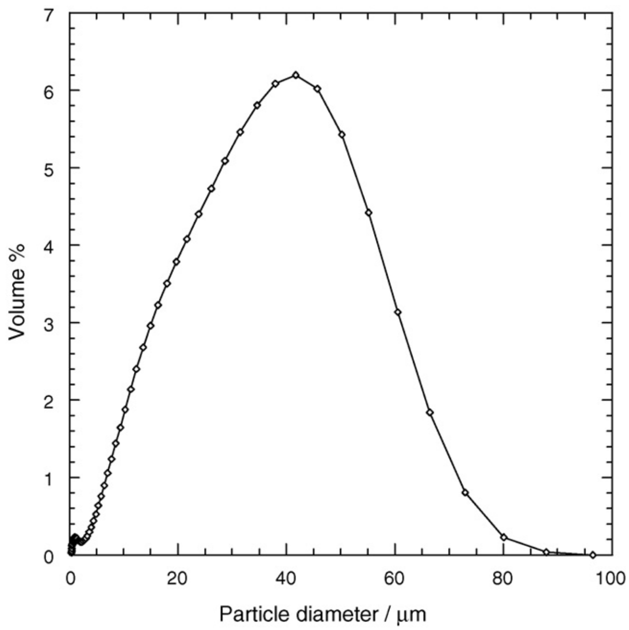

Fig. 11. TS: particle size distribution in the part <62.5 μm.

out according to the French Standard. Three different brick samples of each mix-design were crushed, sieved <4 mm and then leached with distilled water.

The higher pH value obtained for sediment-amended bricks (8.9) may be explained by the transformation of calcite ($CaCO_3$) into lime (CaO) during the firing process. From Table 11 we can observe that the concentrations of the selected metal species (Cd, Cu, Ni, Pb, Zn), for the two mix-designs, are below the regulatory limits. We can also state that the quantities of metals leached out of the fired bodies are less than those of treated sediments, thus metals were either being immobilised within the glassy melt phase or converted to low solubility metal oxides during the firing process. For Zn, values of 0.053 and 0.177 mg/kg were obtained for BdN-standard and sediment-amended bricks respectively. Nevertheless, these values were less than the regulated limit (4 mg/kg).

3.6.2. TCLP-USEPA 1986

Leaching with acidic solution was performed to simulate the leaching process that could be induced by acid rain when bricks are stored unsheltered or used in pedestrian walkways. Table 12 gives the average values of TCLP test undertaken on three different bricks of each mix-design. As shown in Table 12, metal concentrations for the two mix-designs are higher than those obtained by the French procedure (pH 8.9), but they are still far below the regulated TCLP limits. We can also observe, as for the French Standard test, that the quantities of metal leached out of the fired bodies are less than those of treated sediments, confirming that the firing process led to better stabilization of heavy metals. For Zn, values of 3.7 and 3.3 mg/kg were obtained for sediment-amended and BdN-standard bricks respectively. These values are largely inside the TCLP limit, which is equal to 25 mg/kg. The results in Tables 11 and 12 indicate that sediment-amended brick can be considered as non-hazardous material.

4. Conclusion

This industrial experiment demonstrates the practical incorporation of treated river sediments into brick production as a suitable alternative to their current disposal paths. Treated sediments were introduced into the brick-manufacturing process as a replacement of sand 1 (quartz sand) with a substitution ratio of 15%. This material was introduced without any alteration of the existing process lines. Approximately 15,000 of sediment-amended perforated bricks (6 cm × 22 cm × 22 cm) were produced. The produced bricks were subjected to several qualification tests (compressive strength, freeze and thaw resistance, water absorption). The results obtained have demonstrated that sediment-amended bricks meet the "clay bricks" specifications and environmental requirements given by the existing standards. The substitution of sand by treated sediments resulted in a compressive strength increase of 63%, a decrease in porosity of 10%, a decrease in water absorption coefficient of 13% and an increase in firing shrinkage of 40%. Moreover, leaching tests undertaken on sediment-amended bricks showed that the concentrations of heavy metals in the leachates were largely inside the regulatory limits thus, sediment-amended bricks can be regarded as non-hazardous material. White spots and grains were observed on the surface of sediment-amended bricks. This is linked to the presence of calcite with a particle size >1 mm in treated sediments. The presence of these spots does not affect the mechanical properties of produced bricks. We currently work on the introduction of more important percentages of treated sediments into brick production which will improve the valorization path environmentally and economically.

References

[1] Agence de l'Eau, La qualité des sédiments des cours d'eau, 1991–1996.
[2] F. Marot, Caractérisation et traitement de sédiments de dragage contenant des polluants métalliques, BRGM Eds, 1998.
[3] Y. Krieger, R.T. Barber, Effects of waste dumping in New York bight on the growth of natural populations of phytoplankton, Environ. Pollut. 5 (4) (1970) 237–252.
[4] R. Rosenberg, Effects of dredging operations on estuarine benthic macrofauna, Marine Pollut. Bull. 8 (5) (1977) 102–104.
[5] I. Mannino, S. Soriani, G. Zanetto, Management of port dredged material: an environmental-political issue, in: The Changing Coast, Littoral 2002, EUROCOAST/EUCC, Porto, Portugal, EUROCOAST, Portugal, 2002.
[6] A. Andrés, M. Carmen Días, A. Coz, J.R. Viguri, A. Irabien, Proceedings 2004. Global Symposium on Recycling Waste Treatment and Clean Technology, Madrid, Spain, 2004, pp. 171–181.
[7] A.M. Segadães, C. Kniess, W. Acchar, N.C. Kuhnen, D. Hotza, Proceedings 2004. Global Symposium on Recycling Waste Treatment and Clean Technology, Madrid, Spain, 2004, pp. 503–511.
[8] K. Hamer, C. Waschkowitz, M. Isenbeck-Schröter, H.D. Schulz, Harbour sediments for brick production, in: Ressourcen-Umwelt-Management, Schriftenreihe der Gesellschaft für Umwelt Geowissenschaften (GUG), Köln, 1999, pp. 223–240.
[9] K. Hamer, V. Karius, Brick production with dredged harbour sediments. An industrial-scale experiment, Pergamon, Waste Manage. 22 (5) (2002) 521–530.
[10] V. Karius, K. Hamer, PH and grain-size variation in leaching tests with bricks made of harbour sediments compared to commercial bricks, Sci. Total Environ. 278 (3) (2001) 73–85.
[11] I. Demir, M. Orhan, Reuse of waste bricks in the production line, Build. Environ. 38 (2003) 1451–1455.
[12] P. Turgut, H. Murat Algin, Limestone dust and wood sawdust as brick material, Build. Environ. 42 (2007) 3399–3403.
[13] I. Demir, An investigation on the production of construction brick with processed waste tea, Build. Environ. 41 (2006) 1274–1278.
[14] Y.S. Hsu, B.J. Lee, H. Liu, Mixing reservoir sediment with fly ash to make bricks and other products, in: International Ash Utilisation Symposium, Paper #89, Center for Applied Energy Research, University of Kentucky, 2003.
[15] A.G. Liew, A. Idris, A.A. Samad, C.H.K. Wong, M.S. Jaafar, A.M. Baki, Reusability of sewage sludge in clay bricks, Springer-Verlag, J. Mater. Cycles Waste Manage. 6 (2004) 41–47.
[16] D.F. Lin, C.H. Wenig, Use of sewage sludge ash as brick material, J. Environ. Eng. 127 (October (10)) (2001).
[17] C.H. Weng, D.F. Lin, P.C. Chiang, Utilization of sludge as brick materials, Adv. Environ. Res. 7 (2003) 679–685.
[18] M. Anderson, R.G. Skerratt, J.P. Thomas, S.D. Clay, Case study involving using fluidised bed incinerator sludge ash as a partial clay substitute in brick manufacture, Water Sci. Technol. 34 (3–4) (1996) 195–205.
[19] M. Anderson, M. Elliott, C. Hickson, Factory scale trials using combined mixtures of three by-product wastes (including incinerated sewage sludge ash) in clay building bricks, J. Chem. Technol. Biotechnol. 77 (2002) 345–351.

[20] B. Wiebusch, M. Ozaki, H. Watanabe, C.F. Seyfried, Assessment of leaching tests on construction material made of incinerator ash (sewage sludge). Investigations in Japan and Germany, Water Sci. Technol. 38 (1998) 195–205.
[21] X. Lingling, G. Wei, W. Tao, Y. Nanru, Study on fired clay bricks with replacing clay by fly ash in high volume ratio, Construct. Build. Mater. 19 (2005) 243–247.
[22] R. Menezes, H.S. Ferreira, G.A. Neves, H. Lira de L, H.C. Ferreira, Use of granite sawing wastes in the production of ceramic bricks and tiles, J. Eur. Ceram. Soc. 25 (2005) 1149–1158.
[23] C. Huang, J.R. Pan, Y. Liu, Mixing water treatment residual with excavation waste soil in brick and artificial aggregate making, J. Environ. Eng. 131 (February (2)) (2005) 1.
[24] E.A. Dominguez, R. Ullmann, Ecological bricks made with clays and steel dust pollutants, Appl. Clay Sci. 11 (2) (1996) 237–249.
[25] Z. Lafhaj, M. Samara, F. Agostini, L. Boucard, F. Skoczylas, G. epelsenaire, Polluted river sediments from the North region of France: treatment with Novosol® process and valorization in clay bricks, Construct. Build. Mater. 5 (2008) 755–762.
[26] Official Journal J.O.No. 184 (10 August 2000), 12415 (NOR: ATEE0090254A).
[27] C. Alzieu, Environmental impacts of port dredging, in: Dredging and Marine Environment, Editions Ifremer, 2005.
[28] Publication EP1341728 (19/04/2002), Patent correspondant: FR2815338 (17/10/2000), Procédé d'inertage de boues, SOLVAY.
[29] N. Kahalé, Novosol® process: sludge stabilisation and beneficial reuse, in: Proceeding of the 2nd International Conference on Remediation of Contaminated Sediments, Batelle Press, Venice, Italy, 2003.
[30] J.E. Aubert, Valorisation d'une cendre d'incinérateur d'ordures ménagères, traitée par le procédé REVASOL®, dans le béton hydraulique, Thèse de doctorat, Université Paul Sabatier de Toulouse, 2002.
[31] AFNOR, XP X31-210, May 1998, Déchets - Essai de lixiviation.
[32] SEPA, 986. Test Methods for Evaluating Solid Waste: Physical/Chemical Methods. SW-846. Washington, DC.
[33] Commission of the European Communities, 2002, Bruxelles, le 20.9.2002COM, 512 final.
[34] M. Samara, Valorisation des sediments fluviaux pollués après inertage dans la brique cuite, Thèse de doctorat, Ecole Centrale de Lille, 2007.
[35] K. Elert, G. Cultrone, C.R. Navarro, E.S. Pardo, Durability of historic buildings—influence of composition and microstructure, J. Cult. Heritage 4 (2003) 91–99.
[36] G. Cultrone, E. Sebastián, K. Elert, M.J. de la Torre, O. Cazalla, C.R. Navarro, Influence of mineralogy and firing temperature on the porosity of bricks, J. Eur. Ceram. Soc. 24 (2004) 547–564.
[37] G. Cultrone, C. Rodriguez Navarro, E.E. Sebastian, O. Cazalla, M.J. de la Torre, Carbonate and silicate phase reactions during ceramic firing, Eur. J. Miner. 13 (2001) 621–634.
[38] A. Barba, V. Beltrán, C. Feliu, J. García, F. Ginés, E. Sánchez, V. Sanz, Materias primas para la fabricación de soportes de baldosas cerámicas, Instituto de Tecnologia Cerámica-AICE, Castellón, 1997, 291 pp.
[39] W.D. Kingery, Introduction to Ceramics, John Wiley & Sons, New York, 1960.
[40] W.D. Kingery, M. Berg, Study of the initial stages of sintering solids by viscous flow, evaporation-condensation, and self-diffusion, J. Appl. Phys. 26 (10) (1955) 1205–1212.
[41] F.M. Khalaf, A.S. DeVenny, New tests for porosity and water absorption of fired clay bricks, J. Mater. Civil Eng. 14 (4) (2002).
[42] AFNOR, NF EN 771-1, Specification for masonry units—Part 1: Clay masonry units, February 2004.
[43] AFNOR, NF EN 771-1, Methods of tests for masonry units—Part 1: Determination of compressive strength, January 2001.
[44] Y. Kobayashi, O. Ohira, Y. Ohashi, E. Kato, Effect of firing temperature on bending strength of porcelains for tableware, J. Am. Ceram. Soc. 75 (7) (1992) 1801–1806.
[45] V. Kilikoglou, G. Vekinis, Y. Maniatis, Toughening of ceramic earthenwares by quartz inclusions: an ancient art revisited, Acta Metall. Mater. 43 (8) (1995) 2959–2965.
[46] S. Maity, B.K. Sarkar, Development of high-strength whiteware bodies, J. Eur. Ceram. Soc. 16 (1996) 1083–1088.
[47] AFNOR, NF P 13-304, Essai de résistance au gel, Octobre 1983.