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Direct-utilization of sewage sludge to prepare split tiles

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Abstract

As a type of solid waste with large quantities of pollutants, municipal sewage sludge (MSS) itself and its reclamation are becoming a growing concern of governments. In this paper, the direct-utilization of MSS to prepare split tiles is proposed and tested. Without any pretreatments such as thermokinetic drying, the crude MSS from domestic wastewater treatment plant is directly incorporated into the batch mixtures, and then wet ball-milled, filter-pressed, pug-milled, extrusion-formed, dried and fired to obtain split tiles. A series of formulation experiments and physical and chemical characterizations were carried out; the results show that the feasible maximum content of the crude MSS is as high as 60 wt%, and corresponding bending strength and water absorption of split tile samples fired at 1210 °C are 25.5 MPa and 1.14 wt% respectively. TCLP test reveals that the samples are environmentally compatible. The prospective industrial application of MSS to produce split tile will help to significantly reduce its environmental impacts.
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Keywords: Sewage sludge; Split tile; Waste reclamation; Ceramic; Building material

1. Introduction

Municipal sewage sludge (MSS) is a kind of solid waste discharged by domestic wastewater treatment plants, and contains a great amount of pollutants such as organic contaminants, heavy metals, pathogenic microorganisms and so on, which easily lead to serious secondary environmental pollutions [1–4]. Therefore, much attention has been paid to the pollution controls of MSS worldwide, due to the growing social and environmental pressure [5–8]. Moreover, it is more significant to develop the recycling technologies of MSS while solving its environmental pollution. Several investigations in this field have been carried out, and the main technologies include composting [9,10], anaerobic digestion [11–13], combustion [14,15], thermolysis [16,17], producing building materials [18–20], and so on.

In terms of building materials, MSS is mainly utilized to produce cements [21,22], bricks [23,24], ceramic pellets [25,26] and ceramic tiles, of which the ceramic tile industry is the most potential technological activity sector to absorb solid wastes, due to large quantities of raw materials and final products used.

A large number of works on preparing ceramic tiles from MSS have been performed. Li et al. [27] applied dried MSS for the fabrication of ceramic tiles, and concluded that, with the increase of sludge content, the compressive strength of the ceramic tiles decreases rapidly and the bending strength increases slightly. Jordán et al. [28] utilized dried MSS in the manufacture of ceramic tiles and confirmed that the addition of MSS decreases the bending strength and increases the water absorption of the products, and further suggested that the amount of MSS added must be controlled. Monteiro et al. [19] assessed the utilization of MSS dried at 110 °C in the preparation of red ceramics and concluded that the incorporation of MSS up to 10 wt% will impact the technological properties of the products. Qi et al. [29] used MSS dried at 105 °C to prepare ultra-lightweight ceramics tiles, and revealed that the optimum addition of dried MSS is 20–30 wt% and the resulting samples are light, waterproof and

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nontoxic. Besides, the residues from combusting sewage sludge were also reused to prepare ceramic tiles [30,31].

It can be noted from the above literatures that, owing to its high water content, MSS is firstly pre-dried and then reused and recycled, which would lead to high energy consumption and thus limit the industrial utilization of MSS in a large scale.

In this paper, a novel proposal on direct-utilization of crude MSS as the main raw material to prepare split tiles was designed and tested by the authors. Split tile is a kind of ceramic tile, generally used as a decorative building material mainly for the wall surface, which is produced generally by procedures such as wet ball-milling, filter-pressing, vacuum pug-milling, aging, extrusion-forming, drying and sintering [32,33].

In the manufacture of split tile, the batch mixture generally needs addition of fresh water for subsequent wet ball-milling. Therefore, the water from MSS can partially substitute for the fresh water, implying that the crude MSS with high water content may be directly incorporated into the mixture to produce the split tile. In this case, the dehydrating and/or drying pretreatment of MSS can be bypassed, so as to reduce the energy consumption and to save the corresponding cost, which is the key point and technological significance of this paper.

Besides, during firing the split tile, pathogenic bacteria and organic matters can be decomposed and detoxified [15,34], and the released bioenergy can be recovered [34,35]. Furthermore, the heavy metals from MSS can be effectively solidified in split tile bodies, like other ceramic materials [36–38].

In this research, the objective of our study is to probe the possibility of directly preparing split tile from crude MSS and the impacts of MSS on technological conditions, to investigate the feasible maximum proportion of MSS in split tile formulations, and to measure and evaluate the physical properties and environmental compatibility of the resulting split tile. The final purpose is to develop a novel technology of directly producing split tile from MSS.

2. Materials and methods

2.1. Raw materials

MSS from a domestic wastewater treatment plant in Wuhan City of Hubei Province, PR China, was used in this study. The MSS specimens were obtained in January, April, July and October, and their chemical compositions and water contents are given in Table 1. The ignition losses (IL) of MSS-April and MSS-October are comparable and close to the annual mean value. On the other hand, after deducting IL, the contents of inorganic matters of all specimens are similar to each other. So, the MSS-October specimens were used to prepare split tiles in this paper. In addition, the chemical compositions of supplementary raw materials are also given in Table 1.

In terms of the preparation of ceramic tiles, it can be seen in Table 1 that the chemical compositions of MSS are mainly SiO_2 and Al_2O_3, which are consistent with those of the traditional clayey raw materials for ceramic tiles. Besides, such melting constituents as MgO, K_2O and Na_2O present in MSS are beneficial to the sintering of split tile. However, some harmful impurities, for instance, Fe_2O_3, TiO_2 and CaO, could affect the firing behavior of split tile bodies, and especially large quantities of organics indicated by ignition loss (IL) could lead to relatively high shrinkage and porosity.

2.2. Preparation of split tiles

The flowchart of preparing split tile from crude MSS is shown in Fig. 1. The detailed procedures are as follows: (1) weighing the proportioned raw materials into batch mixtures according to the designed formulations shown in Table 2 and wet ball-milling them into slurries; (2) filter-pressing the slurries into filter cakes with a moisture content of 18–30 wt%; (3) vacuum pug-milling the filter cakes into mud-strips at the pressure of −0.09 MPa and then aging them for 24 h to homogenize their moistures; (4) extrusion-forming the mud-strips into wet green bodies with the size of 150 mm × 65 mm × 7 mm; (5) drying the wet green bodies at 110 °C in an oven, to obtain dry ones with a moisture content of 1–2 wt%; (6) firing the dry green bodies in an electric furnace following the heating procedure below: heating the bodies at a rate of 20 °C/min to 850 °C for 10 min to fully decompose organic matter, continually heating them at the same rate to different sintering temperatures varying from 1150 °C to 1230 °C for 20 min with a temperature interval of 20 °C, and finally obtaining MSS split tiles after cooling naturally.

Table 1
Chemical compositions and water contents of MSS samples and other raw materials (wt%).

Materials	Chemical compositions[a]									Water content
	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	IL[b]	
MSS-January	34.95	11.17	4.26	0.88	5.07	1.55	1.65	1.13	37.24	90.96
MSS-April	36.19	12.20	5.87	0.31	4.13	0.98	2.37	0.58	34.52	86.32
MSS-July	38.01	13.75	4.73	0.26	4.53	2.13	1.39	0.67	32.09	85.47
MSS-October	36.40	12.05	5.16	0.69	3.97	1.31	2.01	0.49	35.40	87.01
Quartz	98.23	0.21	0.62	0.13	0.24	0.22	0.10	0.12	0.05	/
Feldspar	72.18	15.17	0.50	0.50	0.50	0.13	3.50	7.11	0.27	/
Kaolin	44.72	36.91	0.35	0.20	0.11	0.08	0.13	0.15	17.12	/

[a]All measured specimens were dried at 110 °C.
[b]IL: ignition loss.

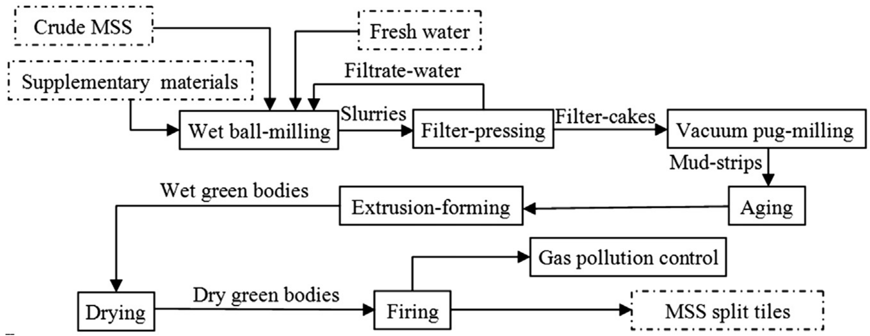

Fig. 1. Flowchart of direct preparation of split tile from crude MSS.

Table 2
Formulations of batch mixtures for MSS split tiles and traditional one (wt%).

Formulations	Raw materials			
	Crude MSS	Quartz	Feldspar	Kaolin
S1	50	6.0	25.2	18.8
S2	55	5.6	22.9	16.5
S3	60	5.2	20.6	14.2
S4	65	4.8	18.4	11.8
Traditional split tile	/	5–15	30–50	40–65[a]

[a]Except kaolin, other kinds of clay minerals may also be used to produce split tile.

Table 3
Chemical composition range of traditional split tile products (wt%).[a]

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O
60–65	20–28	1–3	< 1	< 1	1–3	2–4

[a]Data extracted from literatures [39,40].

It is noted that, when the process is industrially applied in future, combustion kilns will be used for firing with fast heating procedures [39]. In this case, in order to completely burn up the organic matter and further to minimize or even eliminate the organic exhaust gas pollutants, air–oxygen–fuel combustion or oxygen–fuel combustion must be adopted, and it will be also necessary to hold some time at 800–1000 °C. However, in this paper, attention was paid to the preparation process of the split tile, and the exhaust gas issue was not studied and will be a key point in the future work.

In this technology, a great amount of moisture from crude MSS can be firstly used for wet ball-milling, and then be removed by filter-pressing. Subsequently, the filtrate-water is recycled as the new round of ball-milling water, which is favorable to cost-effectively solve the high water-containing problem of crude MSS.

In order to probe the feasible maximum content of crude MSS and the effects on the production of split tile, a series of formulations were designed with the crude MSS contents: 50, 55, 60 and 65 (wt%) (as shown in Table 2). The contents of supplementary materials in each formulation were calculated and determined according to the chemical composition range of traditional split tile products [39,40] as shown in Table 3, and so the chemical compositions of all formulations were approximately constant.

2.3. Characterization of split tiles

According to the standard of "Ceramic tiles—Definitions, classification, characteristics and marking, MOD (ISO 13006:1998)", such properties as linear sintering shrinkage, bending strength, water absorption of the MSS split tile samples were respectively measured as follows: the lengths of split tile specimens before and after firing were measured and used to estimate their linear shrinkages; bending strengths were tested by the three-point method with 60 mm span at a crosshead speed of 1 mm/min; the fired split tile specimens were boiled for 2 h in distilled water, then immersed continually for 4 h, and their weights before and after the immersion were measured to obtain the mass increase rate due to water absorption. For quality assurance and reproducibility, five specimens for each sample were tested and the data averaged.

Differential scanning calorimetry/thermogravimetric analysis (DSC/TGA) of MSS dried at 110 °C was recorded by a differential thermal analysis meter (STA409 PG/PC, Netzsch) at a heating rate of 10 °C/min in air atmosphere in the range between room temperature and 1100 °C. The crystalline phase composition of the samples was identified using an X-ray powder diffractometer (XRD; D/Max-3B, Rigaku) with CuKα radiation, at 35 kV and 40 mA with 10 s scanning time. The microstructure of the split tile after coating with gold was observed by scanning electron microscopy (SEM; Quanta-200FEG, FEI) at 30 kV and 2 μA in the vacuum environment.

Leaching behavior of heavy metals of MSS and split tile samples was parallel when evaluated according to the US EPA Test Method 1311-TCLP, Toxicity Characteristic Leaching Procedure. The specimens were dried at 110 °C to constant weight and milled into powder with the particle size of less than 50 μm, and then were immersed into CH_3COOH water solution (pH 4.93 ± 0.05) at 22 ± 3 °C for 18 ± 2 h. The concentrations of heavy metals in the leachates were tested by inductively coupled plasma mass spectrometry (ICP-MS; AF1 × 7500, America).

3. Results and discussion

3.1. Characterization of MSS

The XRD pattern of MSS sample is shown in Fig. 2. It can be seen that the crystalline phase of MSS is composed mainly of quartz (PDF 03-0427) and kaolinite (PDF 74-1784) and scarcely of illite (PDF 43-0685) and albite (PDF 70-3752), indicating that the as-received MSS is a typical clayey material. All crystal minerals from MSS are available for ceramic production, implying that the substitution of MSS for traditional minerals has the potential to prepare split tiles.

To assess the firing behavior of MSS, DSC/TGA test was carried out and the result is shown in Fig. 3. The weight loss is about 3.6% at 20–200 °C, mainly resulting from the release of hygroscopic water [19]. A large weight loss of 26.0% and a sharp exothermic peak are observed at 200–650 °C, attributed to the combustion and decomposition of biodegradable materials, undigested organics and dead bacteria, as well as to the emission of semivolatile compounds [14,15,41]. Over 650 °C, only a little weight loss occurs, owing to the decomposition of calcium carbonate and the dehydroxylation of kaolinite [42]. Considering that MSS has a relatively high firing sensitivity below 650 °C, it is necessary to lower the heating rate and/or to hold for some time at 650 °C or higher constant temperature, in order to fully destroy the organic matter from MSS and to lower and even eliminate their impact on the sintering of split tile bodies.

3.2. Filter-pressing performance

The wet ball-milled slurries were filter-pressed into filter cakes at the pressure of 4.0 ± 0.5 MPa, and the moisture contents of filter cakes and their changes with the incorporation amount of crude MSS are shown in Fig. 4. The result shows that the moisture contents of filter cakes considerably increase with the increase of the incorporated amount of MSS in split tile formulations, signifying that filter-pressing performance of the slurries becomes more and more difficult and seriously hampers the following extrusion-forming operation. The reason lies in the fact that MSS contains a large amount of organic components and is essentially a colloidal system with high specific resistance, thus giving rise to the difficulty of particles co-aggregating and the blinding of the filter medium, and finally leading to the difficulty in filter-pressing operation [43].

In order to improve the mechanical dewatering, lower the moisture contents of filter cakes, and further to coordinate subsequent extrusion-forming, acetic acid, as a modifier, was doped into batch mixtures at a ratio of 9.0 g acetic acid/kg MSS [44]. The hydrogen ions from acetic acid change the zero zeta potential of the particles in the ball-milled slurry to make them co-agglomerate easily and to release interstitial water [45]. Therefore, it is observed from Fig. 4 that, with the addition of acetic acid, the moisture contents of the modified filter cakes could be considerably reduced, favoring the following extrusion-forming operation.

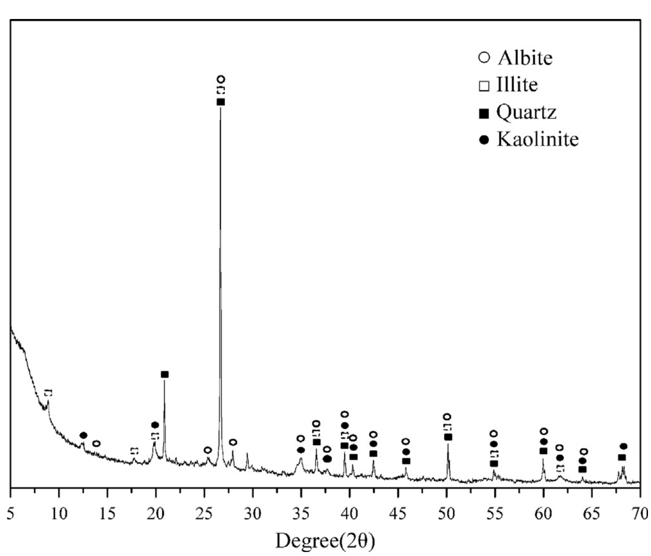

Fig. 2. XRD pattern of MSS.

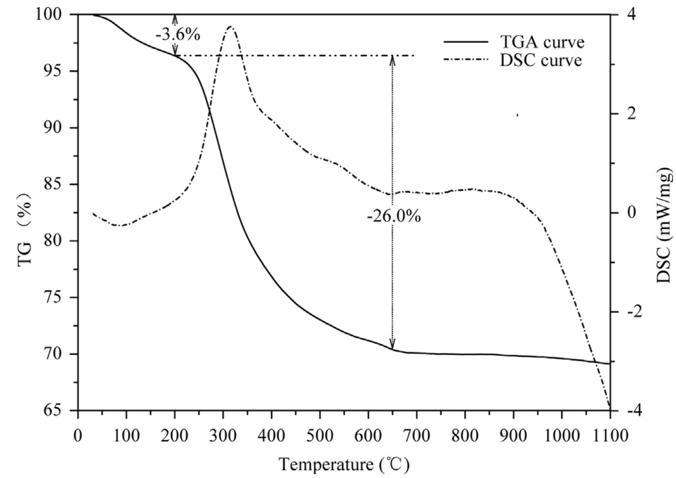

Fig. 3. DSC/TGA curves of MSS dried at 110 °C.

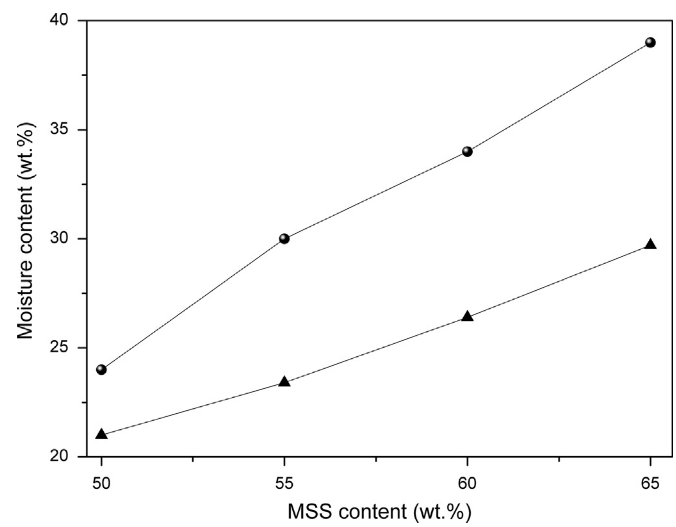

Fig. 4. Changes of the moisture contents of filter cakes with the incorporation amount of crude MSS ((●) not modified by acetic acid and (■) modified by acetic acid).

3.3. Firing performance and physical and chemical properties

The main objective of this research is to estimate the firing performance of MSS split tiles and to determine the feasible maximum incorporation amount of crude MSS and the optimum firing technological conditions.

The surface appearances of the fired samples are shown in Table 4. The bending strength, water absorption and linear sintering shrinkage of the fired samples are exhibited in Fig. 5. However, the specimens of S4 fired at 1210 °C and 1230 °C possess very serious defects that the corresponding physical properties cannot be measured. It is observed that, when the content of crude MSS is lower than 60 wt% (i.e. formulations S1–S3), the water absorption and bending strength of the split tile specimens sintered at 1190 °C and 1210 °C are lower than 3 wt% and higher than 23 MPa respectively, meeting the quality requirement of the fine-grade split tile according to the standard of “Ceramic tiles—Definitions, classification, characteristics and marking, MOD (ISO 13006:1998)”. However, for S3 and S4 samples, when the firing temperature rises up to 1230 °C, over-firing phenomenon arises obviously, leading to a slight increase of water absorption, rapid decrease of bending strength and the emergence of some serious defects.

On the other hand, with the increase of the content of crude MSS, a large amount of organic matter from MSS is also introduced inevitably into batch mixtures, resulting in the increase of ignition losses, linear shrinkage and carbon residue, and thus easily giving rise to such defects as cracks, deformation and bloating [29]. In particular, when the content of crude MSS rises up to 65 wt% (i.e. formulation S4), the corresponding split tile samples present serious defects.

To sum up the above experimental results, for the production of MSS split tile, the feasible maximum content of crude MSS is 60 wt% (i.e. formulation S3), and the optimal firing temperature is 1210 °C. The split tile prepared on the optimal conditions (marked as S3-4) has the bending strength of 25.5 MPa and the water absorption of 1.14 wt% and its appearance is shown in Fig. 6. According to Table 5, S3-4 meets the quality requirement of the fine-grade split tile regulated by ISO 13006:1998 and is comparable to the commercial ones.

To further investigate the crystalline composition and microstructure of MSS split tiles, XRD and SEM analyses of

Table 4
Surface appearances of the split tile samples fired at different temperatures.

Samples	Firing temperature				
	1 1150 °C	2 1170 °C	3 1190 °C	4 1210 °C	5 1230 °C
S1	F	F	F	F	F
S2	F	F	F	F	F
S3	F	F	F	F	B and D
S4	F	F	C	B, C, and D	B, C, and D

F: fine; B: bloating; C: cracked; D: deformed.

Fig. 6. Photograph of the MSS split tile S3-4.

Table 5
Physical properties of the MSS split tile S3-4 and a commercial split tile.

Samples	Physical properties	
	Water absorption (wt%)	Bending strength (MPa)
S3-4	1.14	25.5
Commercial split tile	0.96	26.4
ISO 13006:1998 (AI-grade)	3.0	23

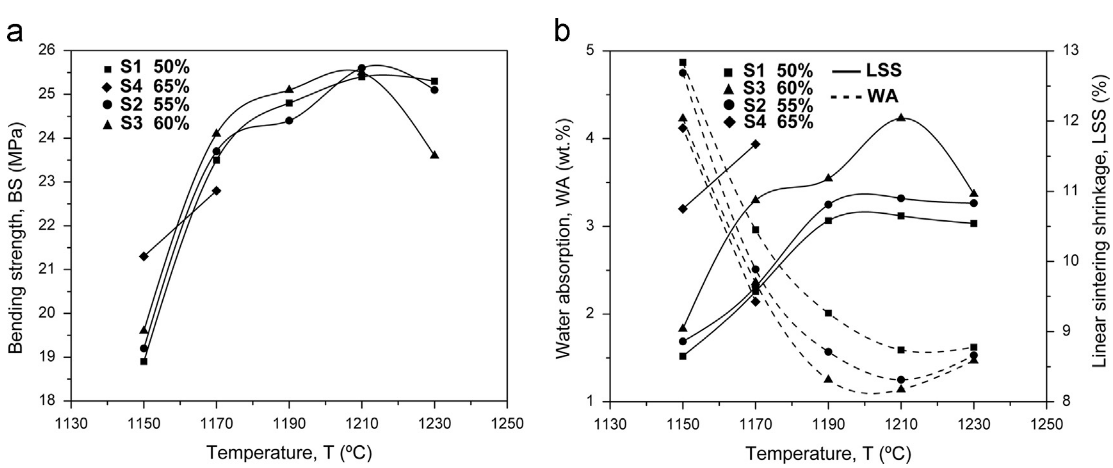

Fig. 5. Bending strength (a) and water absorption and linear sintering shrinkage (b) of the sintered MSS split tile samples versus firing temperature.

S3-4 samples were carried out, and the results are shown in Figs. 7 and 8, respectively. Fig. 7a reveals that the crystalline composition of S3-4 split tile consists mainly of quartz (PDF 86-1692) and mullite (PDF 74-2419). Meanwhile, Fig. 8a shows that the microstructure of S3-4 is constructed by crystal phase and vitreous phase, the former embedding into the latter. The results of XRD and SEM of MSS split tiles are in accordance with those of the commercial split tile as shown in Figs. 7b and 8b, indicating the MSS split tile as being a typical ceramic sintering body [46–48].

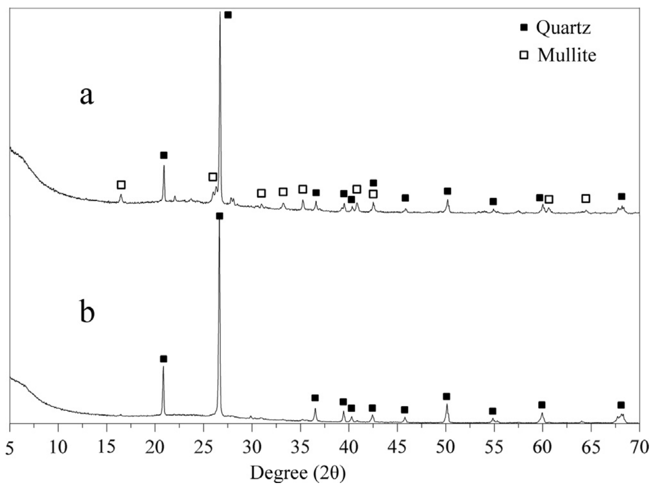

Fig. 7. XRD patterns of (a) the S3-4 split tile with 60 wt% of crude MSS and (b) the commercial split tile.

3.4. Leaching behavior

The leaching tests of the dried MSS, the MSS split tile S3-4, and the commercial split tile were carried out according to the US EPA Test Method 1311-TCLP, Toxicity Characteristic Leaching Procedure, and the results are given in Table 6. The concentrations of heavy metals in MSS, especially Cu, Zn, Cr and Ni, are relatively high. When MSS is being transformed into split tiles, the leaching concentrations of all heavy metals are substantially reduced and are far lower than the regulatory level required by 40 CFR 261.24 [49], and are slightly higher than those of the commercial samples. The leaching results confirm that, like other ceramic materials [50,51], the heavy metals from MSS can as well be solidified and stabilized in the split tile body, and the MSS split tiles are environmentally friendly building materials.

4. Conclusions

Directly using the crude MSS as the main raw material, split tile can be prepared. The optimal formulation with maximum MSS content is composed of 60 wt% crude MSS, 15.2 wt%

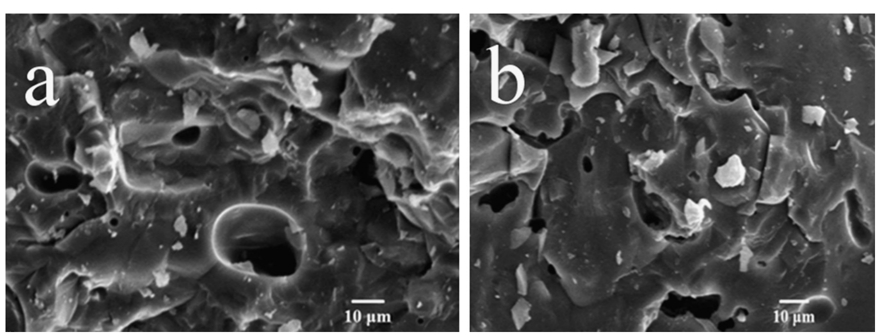

Fig. 8. SEM images of (a) the S3-4 split tile with 60 wt% of crude MSS and (b) the commercial split tile.

Table 6
The leaching concentrations of heavy metals from the dried MSS, the MSS split tile S3-4 and the commercial split tile.

Heavy metals	Dried MSS (mg/L)	S3-4 (mg/L)	Commercial split tile (mg/L)	RL[a] (mg/L)
Cu	3.0243	0.2269	0.0552	NE[b]
Zn	3.4830	0.2845	0.0647	NE
Cd	0.0501	0.0009	0.0001	1.00
Pb	0.0841	0.0136	0.0052	5.00
Be	0.0034	0.0003	ND[c]	NE
Cr	0.1317	0.0009	0.1103	5.00
Ni	0.6434	0.1517	0.0045	NE
Ba	0.7121	0.2204	0.0129	100.00
As	0.8432	0.0520	0.0076	5.00
Se	0.0400	0.0008	ND	1.00
Ag	0.0005	ND	0.0036	5.00
Hg	ND	ND	ND	0.20

[a]RL: regulatory level required by 40 CFR 261.24.
[b]NE: not established.
[c]ND: not detected.

quartz, 20.6 wt% feldspar and 14.2 wt% kaolin, and acetic acid as a modifier. The split tiles fired at the optimum temperature of 1210 °C have the bending strength of 25.5 MPa and the water absorption of 1.14 wt% meeting the property requirement of fine-grade split tiles in ISO 13006:1998. The result of TCLP test reveals that the leaching values of heavy metals are far lower than the regulatory levels required by the US Code of Federal Regulation (40 CFR 261.24), indicating that the MSS split tiles are environmentally compatible. Moreover, without being pretreated, the crude MSS is directly utilized to prepare split tiles, which is favorable to save energy and reduce cost of recycling MSS.

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