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SHORT RESEARCH AND DISCUSSION ARTICLE

An effective treatment method for shale gas drilling cuttings solidified body

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Abstract

The exploration and production of shale gas technology provides a way for utilization of clean fuels. However, during the exploration process of shale gas, enormous amount of drilling cutting was generated and had to be solidified and landfilled. So the accumulation of shale gas drilling cutting solidified body (SGDS)causes severe land resource misuse and environmental complications. This study focuses on the utilization of SGDS as a raw material for the production of cement clinker, and the phase composition, microstructure, and environmental performance of the cement clinker was investigated by X-ray powder diffraction (XRD), scanning electronic microscopy (SEM), energy-dispersive X-ray spectrum analysis (EDX), and soaking test, respectively. The results show that the cement clinker obtained mainly constitutes of typical Portland cement mineral (C_3S, C_2S, C_3A, and C_4AF). The leaching test indicated that the concentration of heavy metal ions in leachate is within the limits allowed by the state "Technical specification for co-processing of solid wastes in cement kiln" (GB 30760-2014). This study therefore provides a benchmark on environmental effects resulting from drilling cuttings and utilization of resources.

Keywords Shale gas · Solidified body · Treatment · Utilization

Introduction

With the rapid development of China's economy, demand for energy is constantly on the rise. This has made shale gas leading as the new energy source and predominantly the main motivation in China's development economically. The straight hole and horizontal well technology for shale gas exploration is commonly used, and it is inevitable that enormous amount of drilling cuttings mixed with drilling fluid generated. Hardening agents, such as Portland cement and fly ash are added to act as a binder for purposes of stabilizing and solidifying it to reduce the contamination caused by the drilling cuttings within a shorter period (Leonard and Stegemann 2010; Kogbara et al. 2016; Kogbara 2014; Chao-qiang et al. 2017). The Shale gas drilling cuttings solidified body (SGDS) was generated and had to been landfilled. Since the SGDS are situated underground, the solid blocks undergo a serious of physical-chemical reaction, resulting in secondary-contamination, especially in heavy metal release and organic pollution (Antemir et al. 2010; Leonard and Stegemann 2010). And thus the need for safe disposal and recycling of SGDS is in urgent.

Previous studies of the physicochemical properties of drilling cuttings have indicated that SGDS can be used in place of sand and mineral addition for the production of building materials, such as the preparation of non-fired brick (Acchar and Marques 2016; Liu et al. 2018; Dmitriy V. Oreshkin et al. 2015), concrete block (Ablieieva and Leonid 2016; Mahmoud Kassem et al. 2018), road material (Tuncan et al. 2000; Chang-Seon Shon et al. 2016), concrete (N.M. Wasiuddin et al. 2002; Ehsan et al. 2015), cement (Wang et al. 2017; Bernardo et al. 2007; Al-Otoom 2006), sintered brick (Xiang-Guo Li et al. 2011), and other materials, such as lightweight ceramsite and cementitious material (Bamdad Ayati et al. 2019; M. Aboutabikh et al. 2016). Although, the research on resource utilization of drilling cuttings has a lot of aspects, but few of them could be put into practice. So, based

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Table 1 Lixivium heavy metal pollutants of raw materials (mg/kg)

Samples	Hg	Tl	As	Pb	Cd	Cr	Cr^{6+}	Cu	Ni	Mn	Be	Sn	Sb	Co	V	Zn	Mo
SGDS	0.713	2.07	6.91	24.03	0.505	46.7	ND	36.9	16.7	119	1.10	ND	1.36	5.19	46.7	131	6.8
Raw meal	0.117	ND	4.37	21.1	1.19	ND	ND	80.7	6.34	140	ND	ND	0.56	5.8	8.9	236	41.2
Pulverized coal	0.121	ND	2.45	1.67	0.196	10.2	ND	11.9	9.1	198	0.17	ND	1.03	3.9	21	47.1	ND
Slag	0.319	ND	183	30.9	1.14	154	ND	103	87.4	796	ND	ND	11.39	41	232	253	16.8
Fly-ash	0.108	ND	7.98	ND	2.92	132	0.27	116	90.3	209	ND	ND	2.58	38.7	149	179	6.76

ND means the item was undetected

on the results of above research, and according to the actual situations, the aim of this study is to evaluate the potential use of SGDS as a raw material for the production of Portland cement clinker on an industry scale, so as to make a resource utilization of SGDS and eliminate the potentially secondary-contamination caused by the landfill of SGDS. This study provides a way for the security and environmental disposal of SGDS.

Materials and methods

Raw materials

SGDS was obtained from a drilling plant in Chongqing. Slag and fly-ash were procured from a local coal-fired power plant. Cement raw meal used was obtained from a cement plant in Chongqing. The raw materials' lixivium heavy metal pollutants were presented in Table 1.

Field experimentation and testing methods

Field experimentation

Figure 1 shows the process flow of the production of the Portland cement clinker using SGDS.

In reference to the preliminary experiments' results, for preparing the cement raw meal, 83.6% limestone, 10.1% sandstone, 2.5% SGDS, 2.8% coal ash, and 1.0% sulfuric acid residue were used (in mass). All the raw materials were crushed, mixed, and ground to the required fineness, typically 20% of retained on an 80 micro sieve. The homogenized raw

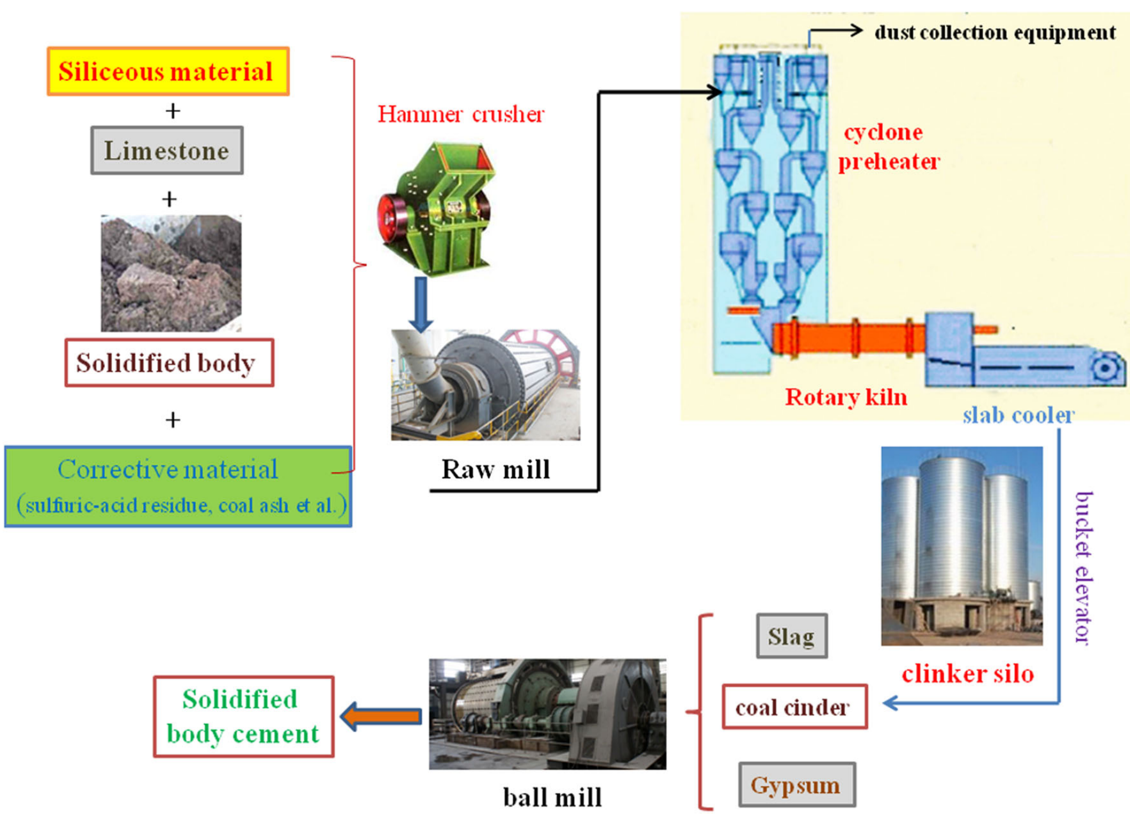


Fig. 1 Process flow sheet of the solidified body cement production

Table 2 Limiting value of SGDS cement clinker heavy metal

Heavy metal	Unit	Result	Maximum allowable value
Hg	mg/kg cli	0.195	0.23
Tl+Cd+Pb+15×As		136.3	230
Be+Cr+10 Sn+50Sb+Cu+Mn+Ni+V		433.4	1150
Cr	mg/kg cem	61.2	320
Cr^{6+}		0.06	10
Zn		374.5	37,760
Mn		398.2	3350
Ni		45.86	640
Mo		53.53	310
As		45.8	4280
Cd		2.28	40
Pb		31.72	1590
Cu		141.53	7920
Hg		0.24	4

meal is introduced into the top of the cyclone preheater and it was heated and decarbonated. After decomposition of the raw meal, it was fed into a rotary kiln in which the raw meal is heated to 1500 °C. After firing, the clinker exits the kiln and is cooled from 1200 °C to 60 °C in a cooler. And then the clinker is ground together with slag, cinder, and desulfurization gypsum in a mill to produce Portland cement.

Test methods

The clear crystalline minerals of the clinker were identified by an X-rd (X-ray powder diffraction), conducted in a Panalytical (X'Pert PRO diffractometer) with a speed current of 60 mA and the voltage of 35 kV. The acquired spectrum was scrutinized in X'Pert High Score and software Plus MDI Jade 5.0.

The use of scanning electronic microscopy (SEM) and energy-dispersive X-ray spectrum analysis (EDX), the morphologies and elements of clinker were analyzed by scanning photo-electron microscope (ASTEREO SCAN440, Leica Cambridge Ltd).

According to the requirement standards for the production of lixivium which detects its admissible indexes and refers to Chinese national standard "Technical specification for co-processing of solid wastes in cement kiln" (GB 30760-2014) to determine the environmental performance of the product. The heavy metal content in leachates were analyzed by ICP-MS methods,

Results and discussion

Heavy metal analysis

According to the requirements of national quality, "Environmental protection technical specification for co-processing of solid wastes in cement kiln" (HJ 662-2013), the limiting value of heavy metal kiln feeding has specific requirements. Therefore, according to the article, the heavy metal content contained in solidified body cement clinkers is presented in Table 2. It is indicated that the heavy metal content contained in the clinker produced from SGDS is within the limits set by national standard.

XRF analysis

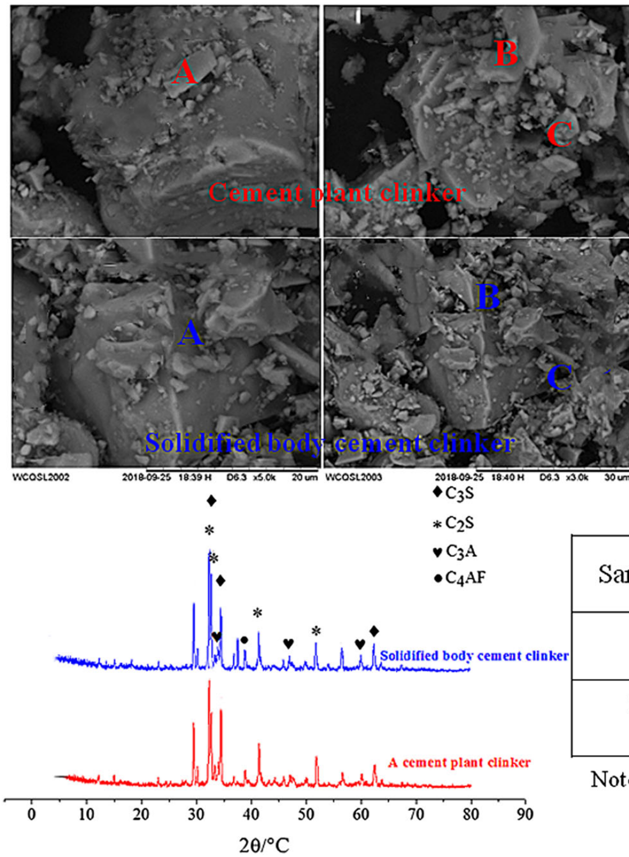
It is clear from the Table 3 that the chemical composition of the SGDS is similar with the composition of commercially available cement. However, the Cl, SO_3, and K_2O contained in SGDS were lower than commercially available cement, making the SGDS an idea raw material for the production of the Portland cement clinker.

Microstructure analysis

As shown in Fig. 2, that the mineral phases of the cement clinker produced from SGDS mainly consist of C_3S, C_2S,

Table 3 Chemical composition % by mass

Samples	CaO	SiO_2	Al_2O_3	SO_3	Fe_2O_3	MgO	K_2O	Na_2O	Cl
Commercially available cement	69.68	21.03	3.91	1.17	3.42	1.01	0.61	0.25	0.03
SGDS	70.79	20.11	3.75	0.84	2.69	1.04	–	0.19	–



Atom	Spot A/%	Spot B/%	Spot C/%
Ca	61.8	62.3	60.9
Si	35.7	33.6	33.8
Al	1.87	1.90	1.63
Fe	1.47	1.53	1.50

Atom	Spot A/%	Spot B/%	Spot C/%
Ca	64.1	62.5	63.7
Si	34.4	34.3	35.2
Al	2.01	1.99	1.84
Fe	1.26	1.22	1.35

Table 4 Molar ratio of calcium to silicon

Samples		Spot A/%	Spot B/%	Spot C/%
1#	Ca/Si	1.731	1.853	1.804
2#		1.863	1.822	1.811

Note : 1# is the cement plant clinker
2# is the solidified body cement clinker

Fig. 2 Mineral phases of the cement clinker produced from SGDS

C_3A, and C_4AF, and the diffraction peak intensity and position were brought into correspondence with typical commercially available cement. In addition, Fig. 2 indicated that the major crystalline state of the cement clinker produced from SGDS is arranged in a lamellar fashion C_3S (spot B and spot C) and near-spherical particles C_2S (spot A) around the mineral phases. This may owe the addition of SGDS which can promote the formation of liquid phase in the raw meal. The early appearance of liquid phase reduces the burning temperature of cement clinker prepared with SGDS. The burnability was improved of the raw meal which contains SGDS, and is help for the synthesis of Belite. In addition, from Table 3, we can conclude that the calcium to silicon ratio of C_2S and C_3S unblended raw meal are 1.731 and 1.829, respectively; the cement clinkers produced from SGDS are 1.863 and 1.817, respectively, which is in accordance with the proposed theoretical analysis. Another cause could be the presence of alkaline liquid phase and valence bond strong AlO_4^{5-} and FeO_4^{5-} tetrahedron which are not conducive to the formation and growth of C_3S crystals, so that the C_3S mineral of clinker is close to C_2S.

Table 4 Lixivium content of heavy metal pollutants Unit: mg/kg

Item	Cu	Zn	Cd	Ni	As	Cr	Pb	Mn
Standard value	100	500	1.5	100	40	150	100	600
Detection value	87.9	279.4	0.51	31.9	6.38	47.6	54.7	316.9

Environmental safety analysis

The results of leaching property of the cement clinker produced from SGDS are presented in the Table 4.

The dates in Table 4 clearly indicate all indexes within the required limit for the China standard "Technical specification for co-processing of solid wastes in cement kiln" (GB 30760-2014) (Liu et al. 2018; Shu et al. 2016). In conclusion, drilling cuttings solidified body-based cement clinker cannot pollute the environment.

Conclusions

This paper presents a case study of resource utilization of the SGDS in industrial scale. And it is indicated that the SGDS could be used as a raw material for the production of Portland cement clinker in a cement production line with a daily output of 10,000 tons using the ingredient ratio: 83.6% limestone, 10.1% sandstone, 2.5% solidified body, 2.8% coal ash, and 1.0% sulfuric-acid residue, resulting a daily consumption of about 250 tons of SGDS. The cement clinker obtained mainly constitutes of typical Portland cement minerals (C_3S, C_2S, C_3A, and C_4AF) and the leaching tests showed that the concentration of heavy metal ions in leachate of the cement clinker is within the limits allowed by the state national standard. Our strategy has provided a convenient reference for the controlling of the pollution induced by the SGDS and the resource utilization of it in a practical way.

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