

Invited Review

Mechanical, expansion and rheological properties of circulating fluidized bed fly ash based ecological cement: A critical review

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Abstract: Circulating fluidized bed fly ash (CFBFA) is a solid waste product from circulating fluidized bed (CFB) boilers in power plants, and the storage of CFBFA is increasingly become an environmental problem. Previous scholars have made contributions to improve the resource utilization of CFBFA. Especially, ecological cement is prepared by CFBFA, which is more conducive to its large-scale utilization. In recent years, a lot of effort has been paid to improve the properties of ecological cement containing CFBFA. In this work, the physicochemical properties of CFBFA are introduced, and recent research progress on the mechanical, expansion, and rheological properties of CFBFA based ecological cement (CEC) is extensively reviewed. The problem of over-expansion of f-CaO is summarized, which limits the scale application of CFBFA in ecological cement. Hence, the challenge for f-CaO in CFBFA to compensate for cement volume shrinkage is proposed, which is beneficial to the utilization of CFBFA in ecological cement, and the reduction of CO₂ emissions from the cement industry. In addition, the environmental performance, durability, and economy of CEC should be valued in future research, especially the environmental performance, because the CFBFA contains heavy metals, such as Cr, As, which may pollute groundwater.

Keywords: circulating fluidized bed fly ash; ecological cement; expansion; durability; compensation shrinkage

1. Introduction

Circulating fluidized bed fly ash (CFBFA) is a by-product of coal burning produced by circulating fluidized bed (CFB) boilers in power plants [1–2]. The accumulation of CFBFA has gradually increased with the rapid development of CFB technology [3–6]. Moreover, CFB technology efficiently burns coal with clean desulfurization in furnaces, which has garnered worldwide attention [7–9]. Table 1 shows the global production of CFBFA, and these data indicate that CFB boilers are in development around the world [10–14]. Additionally, it clear that the production of CFBFA is much higher in China than in other countries. According to statistics, the production of CFBFA in China accounts for 10% of the total amount of fly ash in Chinese power plants [15].

Fig. 1 presents the estimated production of CFBFA in China from 2011 to 2019 [15–16]. In recent years, the production of CFBFA gradually increased to reach 65 million tons in 2019 due to the rapid development of CFB technology [10]. Up to now, the total accumulation of CFBFA is about 630 million tons in China, but its comprehensive utilization rate is less than 20% [17]. Fig. 2 shows the amount of CFBFA in China's power plants in 9 major provinces in 2018. The top three provinces are Shandong, Jiangsu, and Inner Mongolia, which account for 10.90%, 9.09%, and 8.41%

of CFBFA production, respectively.

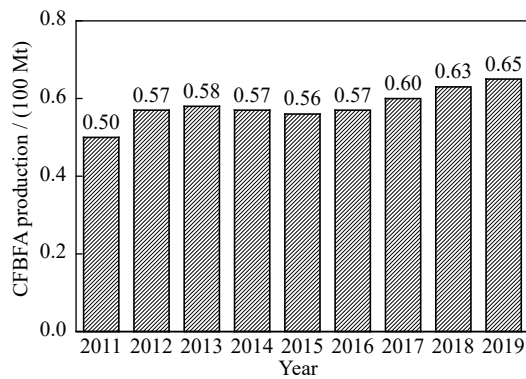
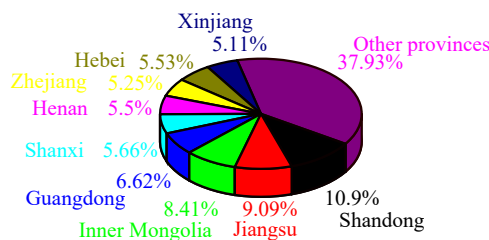
The first study that focused on CFB technology dates back to 1984 by Yu and Schügerl [18]. The operating principle of a CFB boiler is different from that of a pulverized coal boiler [19–21]. First, the CFB combustion temperature is 850–900°C, which is lower than that of pulverized coal furnaces (1200–1400°C) [22]. Second, limestone or dolomite is added to CFB boilers as an adsorbent. CaO is formed due to the decomposition of the adsorbent, while anhydrite (CaSO₄-II) is formed from the reaction of CaO, SO₂, and O₂ [23–24]. The adsorption rate of SO₂ can reach 90% when the molar ratio of calcium to sulfur is 2–2.5 [25]. Therefore, the emission of SO₂ from low-grade coal combustion can be significantly reduced using CFB technology [26]. Currently, there are more than 8000 CFB boilers in the world in power plants, which indicates that the production of CFBFA will significantly increase [27]. Moreover, it is reported that the total accumulation of CFBFA is about 630 million tons in China, which occupies a large amount of land and pollutes groundwater [28–30]. Therefore, it is necessary to treat CFBFA safely from environmental conservation, recycling, and storage cost reduction aspects.

The harmless treatment of CFBFA in different fields has been widely researched and reviewed. For example, valuable components, e.g., silicon and aluminum, are effectively ex-

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Table 1. Annual output of CFBFA in some countries around the world

Nation	Annual output / Mt	Bibliography
China	65.00(2019)	[10]
United States	16.00 (2019)	[11]
Czech Republic	0.60 (2018)	[12]
Korea	2.00 (2017)	[13]
Philippines	7.30 (2016)	[14]

**Fig. 1.** China's annual output of CFBFA from 2011 to 2019.**Fig. 2.** Percentage of CFBFA production in China's provinces in 2018.

tracted from CFBFA, and CFBFA is used to prepare catalysts in the chemical and biological fields [31–32]. CFBFA can also be synthesized as an adsorbent to adsorb hazardous ions in aqueous solutions [33–35]. CFBFA is used to make fertilizers for plants because it contains silicon, phosphorus, potassium, and other valuable elements [36]. In addition, some reviews have been reported on the resource utilization of CFBFA. For example, Ohenoja *et al.* [37] discussed the products prepared for resource utilization of CFBFA, e.g., fertilizer and stabilization materials. He *et al.* [38] summarized some methods to reuse CFBFA, especially the pre-removal methods of unstable components before utilizing CFBFA and proposed that pre-hydration treatments could reduce the f-CaO content in CFBFA. While CFBFA has been treated and applied in these fields, CFBFA has not been utilized on a larger scale.

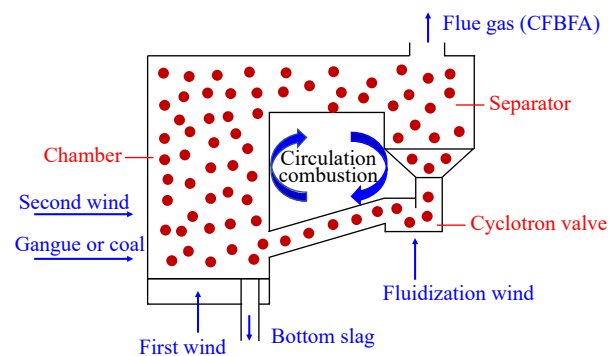
As a consequence, the utilization of CFBFA in cement was proposed, which was first discovered by Drottner and Havlica [39] in 1997. This method not only reduces CO₂ emissions by the cement industry [40–41] but also has the potential to be used as a cement component to conserve the traditional cement [42–45]. Additionally, CFBFA contains

more amorphous clay minerals because the temperature of CFB boilers is lower than that of pulverized coal furnaces. Hence, the activity of CFBFA is higher than that of ordinary fly ash (OFA) [46–48]. Alkali compounds, sulfates, and carbonates can be used as activators of CFBFA [49–51]. The strength of CECs can be enhanced by hydration products, e.g., C–S–H gels and C–A–S–H gels [52–54]. Therefore, the utilization and prospects of active CFBFA components in cement have been widely researched [55–57]. According to the literature, CFBFA content should be less than 30wt%. Otherwise, the performance of the CEC will deteriorate [12].

Although there have been many advances in CECs, a systematic review of CECs has not yet been reported. In this work, we summarized many studies on the preparation of CECs, and based on these, we proposed the synergy of unstable components in CFBFA and discussed how CFBFA can provide a positive effect on the volume stability of newly prepared CECs. Especially, the challenges in using f-CaO to compensate for cement volume shrinkage are discussed, because the durability of CECs can be reduced by over-expansion of f-CaO in CFBFA, which results in cracking of the cement. Besides, the mechanical, expansion, and rheological properties and their action-mechanisms of CECs are summarized and discussed, which can serve as a point of reference for future research. Finally, problems in CEC development are discussed, and suggestions for future CEC research are provided.

2. Physicochemical properties of CFBFA

The physicochemical properties of CFBFA are mainly determined by the combustion process of the CFB boiler [58]. Low-grade fuels are used in CFB technology and waste gases are greatly reduced, which provides an effective way to save energy with cleaner production in power plants [59]. Fig. 3 shows the working principle of CFB technology. The CFB boiler is mainly composed of a combustion chamber, separator, and cyclotron valve. Clean and efficient production is accomplished via the coordinated operation of the three core components.

**Fig. 3.** Schematic illustration of the working principle of a CFB Boiler.

2.1. Particle size

Particle size is an important physical factor in the hydra-

tion reaction of CFBFA. The particle size distribution of untreated CFBFA is shown in Fig. 4 [60]. The median diameter and specific surface area of CFBFA are $36.73 \mu\text{m}$ and $412 \text{ m}^2/\text{kg}$, respectively, with a $45 \mu\text{m}$ sieve residue of 26.50%. These results indicate that CFBFA has a smaller particle size and larger specific surface area compared with OFA [61]. Therefore, CFBFA can increase the contact area between particles in hydration reactions, and the reaction rate of the whole system is improved. The rheological properties of CECs are also determined by the particle size of the CFBFA.

2.2. Chemical composition

The chemical composition of CFBFA varies based on different regions and factories. Table 2 shows a chemical composition comparison of CFBFA from different provinces of China [10,17,22,26,28,32,46,60,62]. There are many kinds of CFBFA in China, especially in Shanxi Province, and although the oxide types in CFBFA from different power plants are similar, the content is different. The combustion temperature of CFBs is low ($850\text{--}900^\circ\text{C}$); thus, clay minerals cannot be melted, which results in many active silicon–aluminum components in CFBFA. Additionally, limestone is used as a desulfurizer in CFB, which leads to high CaO content. Low-grade coal is consumed by CFB boiler

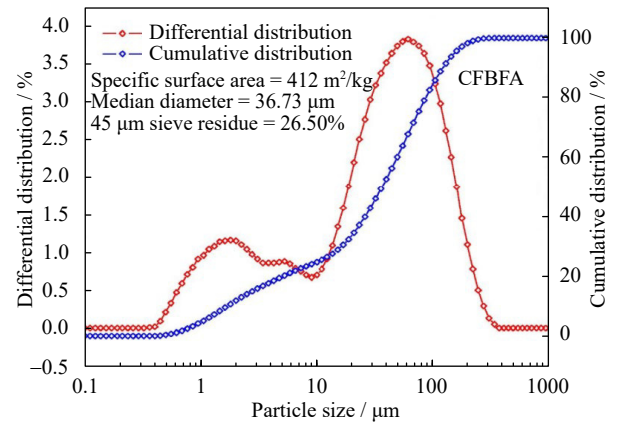


Fig. 4. Particle size distribution curve of CFBFA [60]. Reprinted from *Constr. Build. Mater.*, 101, J.H. Zhao, D.M. Wang, and S.C. Liao, Effect of mechanical grinding on physical and chemical characteristics of circulating fluidized bed fly ash from coal gangue power plant, 851–860, Copyright 2015, with permission from Elsevier.

ers to produce a large amount of SO_2 , which is consolidated by lime and results in high SO_3 content in CFBFA [63]. The combustion loss of CFBFA is large due to its high carbon content. Therefore, different chemical compositions between CFBFA and OFA lead to differences in mineral composition.

Table 2. Chemical composition comparison of CFBFA from provinces in China

Place	SiO_2	Al_2O_3	CaO	SO_3	Fe_2O_3	MgO	Na_2O	LOI	Total*
Shanxi [46]	41.17	30.43	11.48	3.00	2.41	0.31	0.16	6.84	95.80
Sichuan [28]	34.56	13.49	18.36	12.90	11.22	—	—	5.80	96.33
Hubei [62]	32.46	20.06	16.49	8.91	6.05	2.68	0.32	10.03	97.00
Shanxi [60]	37.78	30.76	10.13	4.35	5.42	1.73	0.21	5.01	95.39
Inner Mongolia [32]	35.14	45.35	2.87	0.54	2.61	0.23	0.08	10.90	97.72
Shanxi [22]	40.24	32.83	7.46	2.67	5.62	0.83	—	7.95	97.60
Shanxi [10]	44.90	18.30	17.80	3.30	6.90	2.80	0.90	2.80	97.70
Shanxi [26]	17.43	9.95	41.19	10.13	1.19	1.13	0.29	15.78	97.09
Shanxi [26]	29.37	17.87	22.00	4.21	2.32	1.51	0.39	18.64	96.31
Shanxi [17]	28.51	28.06	16.51	10.26	8.00	—	—	5.23	96.57

Notes: LOI—Loss on ignition (the percentage of lost mass (H_2O , CO_2 , organic matter, etc.) in the mass of the original sample at a certain high temperature and time). *—Only some oxides are listed in the Table 2, so the total is less than 100%.

2.3. Mineral composition

Mineral composition analysis is a method for determining the components in CFBFA. The mineral compositions of OFA and CFBFA are presented in Fig. 5. The primary mineral composition of CFBFA is anhydrite (CaSO_4), calcite (CaCO_3), lime (f-CaO), and quartz (SiO_2), but it does not contain mullite found in OFA [46,64].

This difference can be explained as CaSO_4 is mainly formed from SO_2 , and CaCO_3 and f-CaO are produced by adding an excess of the sulfur fixing agent to the CFB boiler. The hydration reaction of anhydrite with calcium aluminate to produce ettringite can improve the early strength of the CEC. Moreover, the production of $\text{Ca}(\text{OH})_2$ by f-CaO hydration leads to volume expansion. Therefore, the properties of cement containing CFBFA or OFA are quite different be-

cause the mineral compositions of CFBFA and OFA are different.

2.4. Microscopic characterization

Microscopic characterization of CFBFA and OFA are shown in Fig. 6. OFA particles are spherical with smooth and compact surfaces, with a few irregular particles. However, CFBFA consists mainly of irregular particles, which exhibit a loose porous surface structure with many pores. The CFB boiler combustion temperature is $850\text{--}900^\circ\text{C}$, resulting in clay minerals that are difficult to melt, and thus the observed irregular morphology [65–66]. Moreover, these irregular particles are composed of unburnt carbon particles, anhydrite, and calcite [28,60]. This difference in microstructure results in the different effects of CFBFA and OFA on the rhe-

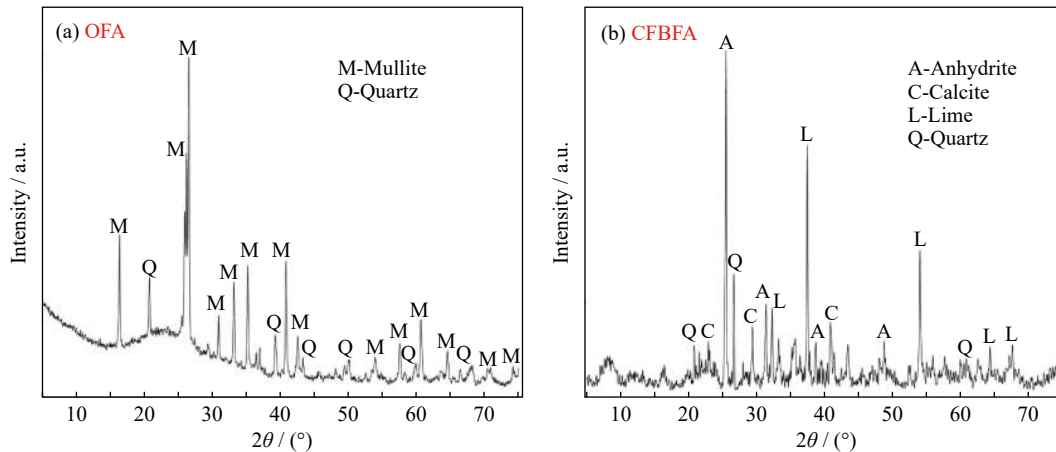


Fig. 5. XRD patterns of OFA (a) and CFBFA (b) [46]. Reprinted from *Constr. Build. Mater.*, 251, M.K. Zhou, P. Chen, X. Chen, X.X. Ge, and Y.Q. Wang, Study on hydration characteristics of circulating fluidized bed combustion fly ash (CFBCA), art. No. 118993, Copyright 2020, with permission from Elsevier.

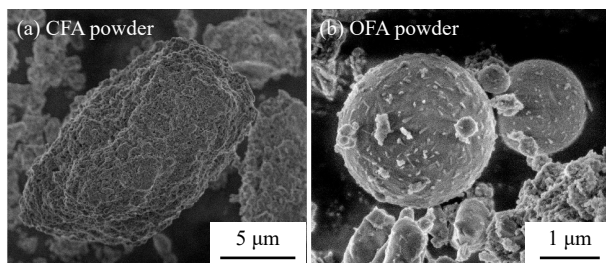


Fig. 6. Microstructures of (a) CFBFA and (b) OFA [12]. Reprinted from *Cem. Concr. Compos.*, 90, P. Hlaváček, R. Šulc, V. Šmilauer, C. Rößler, and R. Snop, Ternary binder made of CF-BC fly ash, conventional fly ash, and calcium hydroxide: Phase and strength evolution, 100-107, Copyright 2018, with permission from Elsevier.

ological properties of cement.

2.5. Self-hardening property

CFBFA has self-hardening properties because of its mineral composition. Fig. 7 shows the self-hardening compressive strength of CFBFA paste at two different water–binder ratios. CFBFA contains f-CaO and CaSO₄ (a mineral con-

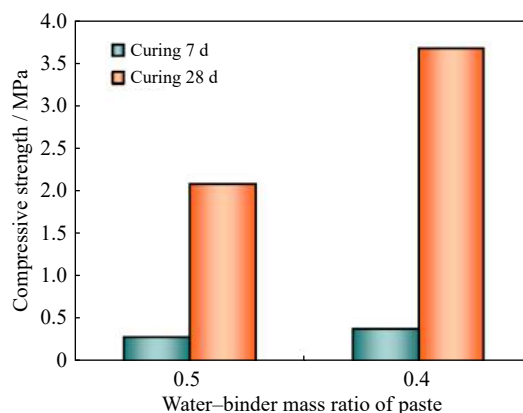
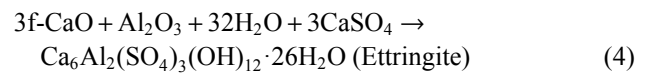
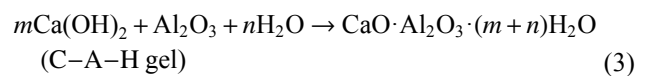
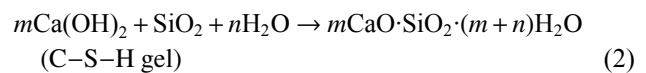


Fig. 7. Self-hardening compressive strength of CFBFA paste [60]. Reprinted from *Constr. Build. Mater.*, 101, J.H. Zhao, D.M. Wang, and S.C. Liao, Effect of mechanical grinding on physical and chemical characteristics of circulating fluidized bed fly ash from coal gangue power plant, 851-860, Copyright 2015, with permission from Elsevier.

taining SO₃), and f-CaO hydration generates Ca(OH)₂ [65]. Meanwhile, the active silica–alumina in CFBFA reacts with Ca(OH)₂ to form C–S–H and C–A–H gels. Additionally, the direct hydration reaction of CaSO₄, f-CaO, and Al₂O₃ produces ettringite. The reactions can be shown as follows:



Therefore, the presence of these hydration products causes CFBFA to have certain self-hardening properties (Fig. 7); however, the self-hardening strength is low [60]. The compressive strength of CFBFA paste is 3.75 MPa after hydration 28 d when the water–binder mass ratio is 0.4.

3. Macro-properties of CEC

3.1. Compressive strength

Compressive strength is an important index to evaluate the macro-performance of CECs [67–68]. Various mechanical properties of CECs, such as elastic modulus, wear resistance, fatigue, bending strength, and splitting tensile strength, are related to compressive strength [69]. There has been extensive research on the preparation of cement using CFBFA, and different CFBFA dosage and water–binder mass ratio was tried to improve the compressive strength of CECs to meet Portland fly-ash cement performance standards [70]. The compressive strengths of different formulations of CECs are summarized in Table 3 [11,25–26,46,71–74]. As shown in the Table 3, the compressive strength of CECs is similar to or even higher than that of traditional cement materials, which satisfy Portland fly-ash cement for compressive strength at curing 28 d (≥ 32.5 MPa (32.5#), ≥ 42.5 MPa (42.5#), ≥ 52.5 MPa (52.5#), and ≥ 62.5 MPa (62.5#)). The main reason is that CFBFA contains many clay minerals, and dehydration

Table 3. Mechanical properties of various CECs reported after curing 28 d

Reference	Water–binder mass ratio	Compressive strength / MPa	CEC composition (mass ratio)
Zahedi <i>et al.</i> [11]	0.50	32.00–33.00	CFBFA : cement = 1:4
Zhou <i>et al.</i> [46]	0.50	47.00–48.00	CFBFA : cement = 1:3
Lee <i>et al.</i> [71]	0.30	61.00–62.00	CFBFA : cement = 1:4
Chen <i>et al.</i> [72]	0.50	42.60–45.30	CFBFA : cement = 3:2
Sheng [26]	0.50	49.50–49.80	CFBFA : cement = 4:1
Chen <i>et al.</i> [25]	0.30	48.00–49.00	CFBFA : quicklime : cement = 35:9:6
Nguyen <i>et al.</i> [73]	0.30	60.00–61.00	CFBFA : OFA : cement = 3:20:15
Chi [74]	0.50	67.00–67.10	CFBFA : slag : cement = 1:3:6

produces amorphous silica–alumina materials. In further, first, the pozzolanic reaction indexes of CFBFA with different particle sizes all satisfy GB/T 1596-2005 standards [72]. The active SiO₂ and Al₂O₃ in CFBFA take part in the hydration reaction to form useful gel products. Second, CFBFA contains SO₃ in the form of anhydrite, which reacts with calcium aluminate in cement to form ettringite.

In addition, the strength of CEC is affected by the water–binder mass ratio and dosage of CFBFA, and the specific study is shown below. The compressive strength of CEC samples prepared by Zahedi *et al.* [11] was 32.00–33.00 MPa when the water–binder mass ratio was 0.50 and 20wt% CFBFA was added in the CEC. Lee *et al.* [71] used CFBFA to replace 20wt% of the cement with a water–binder mass ratio of 0.30, and the compressive strength of the CEC reached 61.00–62.00 MPa. This indicates that a low water–binder ratio is beneficial to the development of the compressive strength of CECs.

Compared with the results from Ref. [11], when the content of CFBFA was increased to 25wt% with the same water–binder mass ratio of 0.50, the compressive strength of the CEC was increased from 32.00–33.00 MPa to 47.00–48.00 MPa [46]. However, when the CFBFA content of CEC continuously increased to 60wt% [72] and 80wt% [26], respectively, the compressive strength of the resulting CECs only reached 45.30 and 49.80 MPa, respectively. This indicates that the compressive strength of CEC samples cannot be continuously improved only by increasing the mass ratio of CFBFA in CECs. Besides, Researchers [25,73–74] found a synergistic effect between two solid wastes, and the compressive strength of the resulting CEC samples can reach 67.10 MPa. These studies demonstrate that the synergistic effect of CFBFA with other solid waste is beneficial to improve the compressive strength of CECs. In conclusion, controlling the CFBFA mass ratio, water–cement ratio, and multi-solid waste synergy can help to improve the compressive strength of CECs.

3.2. Expansion performance

The expansion performance of CFBFA is a major characteristic that distinguishes it from OFA. The expansion performance of CFBFA is closely related to the unstable SO₃ and f-CaO components. Chen *et al.* [72] conducted an in-depth study on the expansion performance of CFBFA in 2017, and CECs were prepared using CFBFA and cement. As shown in Fig. 8(a), the expansion degree of CECs gradu-

ally increases with an increasing of CFBFA mass ratio. This phenomenon indicates that the SO₃ in the CFBFA formed into a large amount of ettringite, leading to volume expansion in the early hydration stage (7 d). Additionally, the formation of Ca(OH)₂ by f-CaO hydration also leads to a continuous increase in CEC volume in the later period (7–90 d). The expansion of CECs is also related to the particle size of the CFBFA. These results (Fig. 8(b)) indicate that the expansion rate of CECs increases with decreasing CFBFA particle size (Fig. 8(c)). This trend demonstrates that CFBFA particle size decreases, the hydration area increases, which is beneficial to improve CEC reaction rates.

Lee *et al.* [71] applied CFBFA to cement to study the expansion performance of CECs. The results indicated that f-CaO and SO₃ in CFBFA have significant effects on the expansion performance of CECs. Fig. 8(c) shows the expansion deformation of CECs under water curing. The results demonstrated that the cement paste expands slightly and with a linear deformation of less than 1000 μm/m with increasing curing time when without CFBFA and contain 10wt% CFBFA. However, the expansion of the paste gradually increases with increasing curing time when CFBFA content increased to 20wt%. The linear deformation of the paste is greater than 4000 μm/m after curing 100 d. These phenomena indicate that CFBFA plays a large role in the expansion of CECs. First, f-CaO hydration generates Ca(OH)₂, which results in volume expansion in CECs. Second, the formation of ettringite is promoted by CFBFA containing SO₃, which also results in volume expansion in the early hydration reaction. This CEC expansion mechanism analysis is consistent with results from Chen *et al.* [72].

CFBFA composite slags also influence CEC expansion. Length variation is an important index to judge CEC durability. A length change will lead to stress imbalance in the CEC, and an excessive length change can lead to CEC cracking, which significantly affects CEC durability. The expansion of CECs is influenced by CFBFA and aggregate content, hydration time, and experimental methods. Chi *et al.* [74] studied the expansion performance of CECs compounded by CFBFA and slags. Twelve groups of CEC samples were prepared according to type and binder amount. The length changes of cement mortars (OPM) and CECs with slags after curing 14, 28, 56, 91, and 180 d are shown in Fig. 8(d), and corresponding raw material compositions are shown in the table inset in Fig. 8(d). The expansion rate of CEC samples

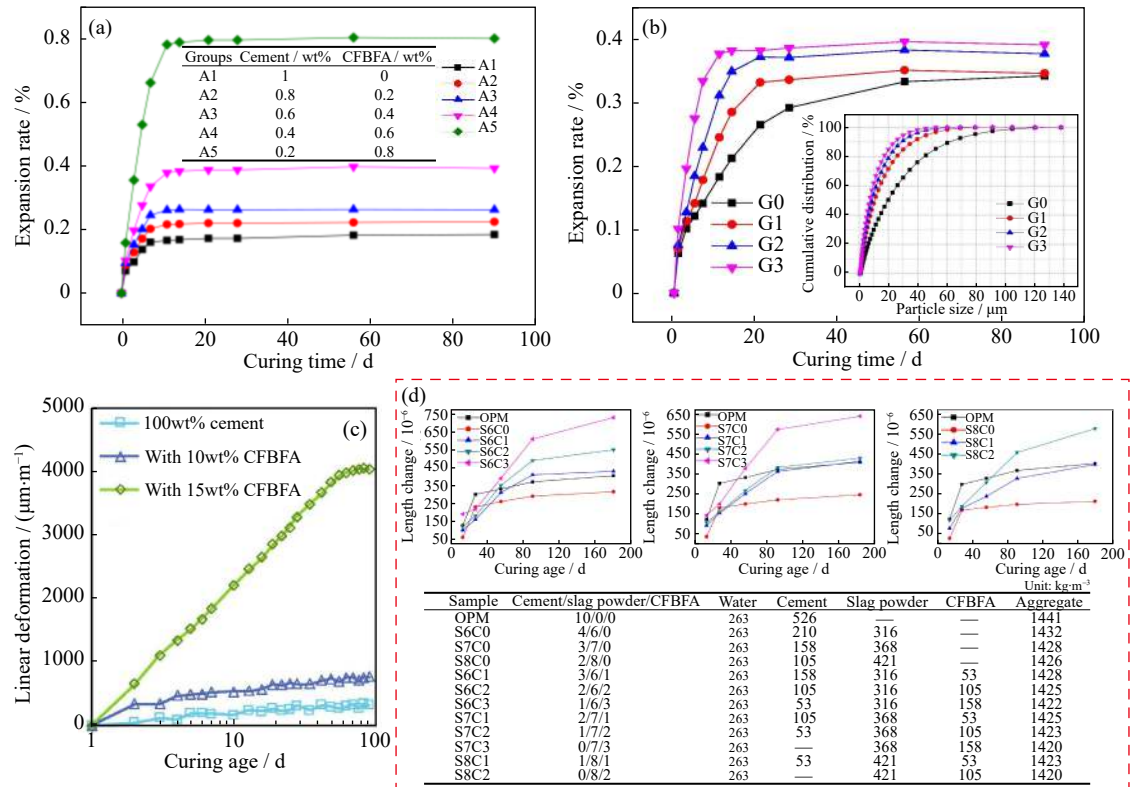


Fig. 8. Effect of particle size and dosage of CFBFA on the expansion of CECs [71–72,74]: (a) change of expansion degree in CECs with curing time and dosage of CFBFA; (b) change of expansion degree in CECs with curing time and CFBFA particle size, and the insert is the corresponding cumulative particle size distribution of CFBFA; (c) expansion deformation of cement and CECs under different water curing times; (d) length changes of cement mortars (OPM) and CECs with slags after curing different days. (a) and (b) Reprinted from *Constr. Build. Mater.*, 157, X.M. Chen, J.M. Gao, Y. Yan, and Y.Z. Liu, Investigation of expansion properties of cement paste with circulating fluidized bed fly ash, 1154–1162, Copyright 2017, with permission from Elsevier. (c) Reprinted from *Constr. Build. Mater.*, 234, H.K. Lee, S.M. Jeon, B.Y. Lee, and H.K. Kim, Use of circulating fluidized bed combustion bottom ash as a secondary activator in high-volume slag cement, art. No. 117240, Copyright 2020, with permission from Elsevier. (d) Reprinted from *Constr. Build. Mater.*, 123, M. Chi, Synthesis and characterization of mortars with circulating fluidized bed combustion fly ash and ground granulated blast-furnace slag, 565–573, Copyright 2016, with permission from Elsevier.

increases with the increase of CFBFA when the mass ratio of the slag remained unchanged. The resulting expansion rate is consistent with that of Lee *et al.* [71] and Chen *et al.* [72]. This phenomenon is mainly caused by expansion from f-CaO and SO₃ in CFBFA.

In summary, the volume expansion of CECs caused by f-CaO and SO₃ in CFBFA has been demonstrated by several researchers. The CaSO₄ (a mineral containing SO₃) reacts quickly in the early hydration stage. Thus, the expansion of CECs is mainly due to the reaction of f-CaO.

3.3. Rheological properties

The relationship between the internal structure and flow behavior of cement is described by its rheological properties, and rheology is reflected by yield stress, viscosity, torque, and other indicators. Researchers have made many contributions to study cement rheological properties. For example, Zheng *et al.* [22] researched the rheological properties of CEC pastes by rheometry. Fig. 9(a) shows that the yield stress of CECs gradually decreases when the CFBFA content increase from 0 to 30wt%, increases when the CFBFA content from 30wt% to 50wt%, and decreases when the CFBFA content is more than 50wt%. The phenomenon is ra-

tionalized as a shape effect of the CFBFA. More pore water is released, i.e., CEC fluidity is enhanced when the CFBFA content is lower than 30wt%. Cement paste mixed with FA1 (OFA), FA2 (CFBFA), and FA3 (Ultra-fine CFBFA) requires more water when the OFA/CFBFA content is increased to 50wt%, resulting in less free water and increased friction between particles, thus increasing the yield stress of the paste. As shown in Fig. 9(a), the yield stress of CECs is significantly reduced when the amount of FA1 and FA2 increases from 50wt% to 70wt%. However, the yield stress of CEC is only slightly reduced when the amount of FA3 increases from 50wt% to 70wt%. This phenomenon can be explained by the poor compacting of the large particles of the raw material, which leads to a sharp decrease in the CEC yield stress.

Fig. 9(b) shows a slight decrease in the plastic viscosity of cement with OFA (FA1). However, the plastic viscosity of the CEC increases with the addition of FA2, and the plastic viscosity of the CEC increases rapidly when the addition of FA2 exceeds 50wt%. The plastic viscosity of the CEC increases to 1.95 Pa·s when the content of FA2 is 70wt%. These increases may have been influenced by particle shape. There may be additional energy dissipation when particles

are non-spherical, which increases the viscosity of the CEC paste [75]. However, the plastic viscosity of the CEC decreases with the addition of FA3. The plastic viscosity decreases by 0.13 Pa·s when the FA3 content increases to 70wt%. The particle size of FA3 is smaller; thus, the intergranular plastic viscosity is reduced. Genovese [75] and Liu *et al.* [76] demonstrated that the flow of large particles is lubricated by small particles, which reduces the viscosity of the paste. Fig.9(b) shows that the rheological properties of cement pastes can be improved by OFA and fine particle CFBFA. The improvement by FA3 on the rheological properties of the cement paste is better than that of FA1.

According to Zheng *et al.* [22], the particle size of CFBFA is the main factor that affects the rheological properties of CECs. Therefore, Xia *et al.* [28] and Liu *et al.* [77] conducted an in-depth study on the influence of CFBFA particle size on CEC rheological properties and developed a concrete produced from CFBFA. They explored the influence of CFBFA on the rheological properties of cement concrete. Fig. 9(c) shows the change in torque of the samples (No. 1, No. 2, No. 3, No. 4) with different speeds and CFBFA particle sizes. Samples No.2, No.3, and No.4 show relatively lower yield

stresses and higher rheological behavior indices compared with No.1. The intergranular viscosity value decreases with decreasing CFBFA particle size, which improves the rheological properties of the CEC.

The effects of OFA, CFBFA, and ultra-fine CFBFA (UCFA) on the rheological properties of cement were compared and discussed. For example, Li *et al.* [78] researched the effect of the concentration of UCFA on the rheological properties of CECs. Fig. 9(d) shows rheological changes of CEC pastes with different UCFA content. The rheology of the CEC pastes first increases and then decreases as the UCFA content increases from 10wt% to 30wt%. The liquidity of the CEC reaches a maximum (312 mm) when the UCFA content is 10wt%. These trends occur because the gap between cement particles is filled by UCFA particles, which releases water and increases CEC fluidity. However, the fluidity of the CEC paste decreases when the UCFA content increases from 30wt% to 70wt% because the UCFA particles adsorbed more free water due to larger specific surface areas. In summary, the particle size and dosage of the CFBFA are the most important physical factors that affect the rheological properties of CEC.

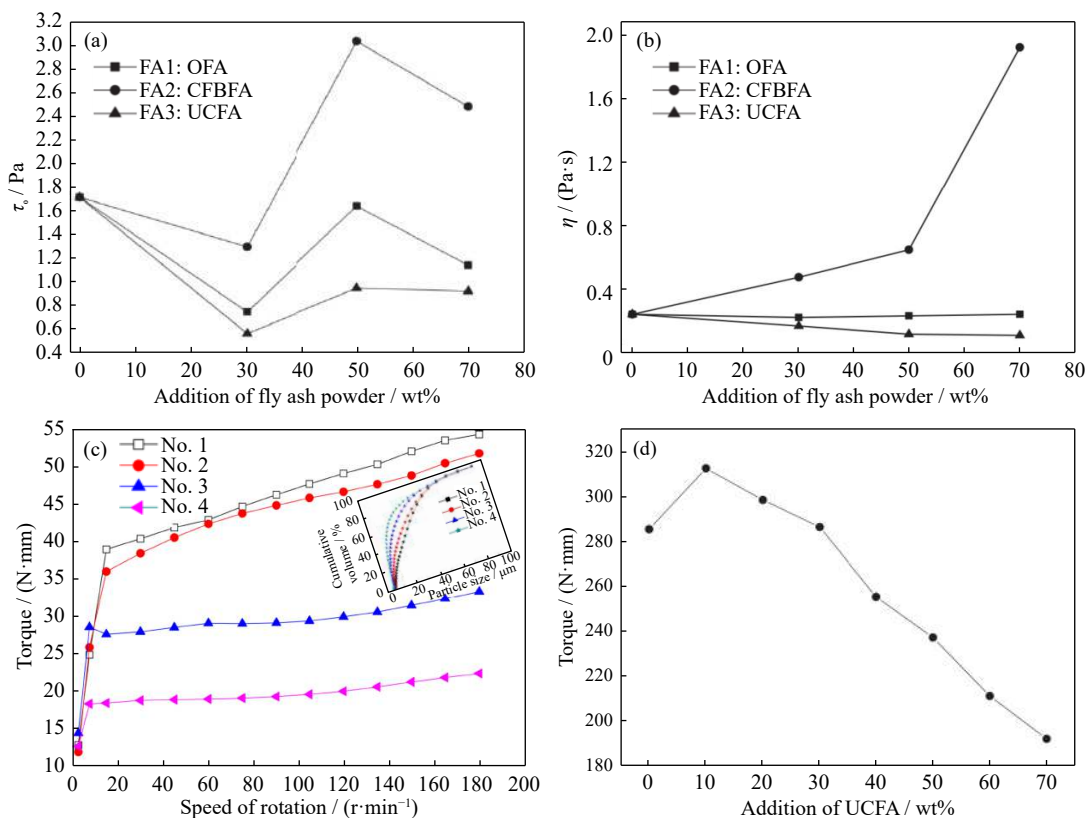


Fig. 9. Effects of fly ashes on the rheology properties of cements [22,28,78]: (a) effect of OFA, CFBFA, and UCFA content on the yield stress (τ_0) of paste; (b) effect of OFA, CFBFA, and UCFA dosage on the plastic viscosity (η) of paste; (c) influence of OFA, CFBFA, and UCFA fineness on rheological properties of pastes, with the insert figure of cumulative particle size distribution of CFBFA; (d) fluidity of cement paste with different content of UCFA. (a) and (b) Reprinted from *Constr. Build. Mater.*, 135, D.P. Zheng, D.M. Wang, D.L. Li, C.F. Ren, and W.C. Tang, Study of high volume circulating fluidized bed fly ash on rheological properties of the resulting cement paste, 86-93, Copyright 2017, with permission from Elsevier. (c) Reprinted from *Constr. Build. Mater.*, 47, Y.Q. Xia, Y. Yan, and Z.H. Hu, Utilization of circulating fluidized bed fly ash in preparing non-autoclaved aerated concrete production, 1461-1467, Copyright 2013, with permission from Elsevier. (d) Reprinted from *Constr. Build. Mater.*, 188, D.L. Li, D.M. Wang, C.F. Ren, and Y.F. Rui, Investigation of rheological properties of fresh cement paste containing ultrafine circulating fluidized bed fly ash, 1007-1013, Copyright 2018, with permission from Elsevier.

4. Microstructural characteristics of CECs

4.1. Microstructural evolution affects compressive strength

The microstructural evolution of CECs was analyzed to study further the enhancement mechanism of the compressive strength of CECs [79]. Various minerals in CECs can be quantitatively analyzed by backscattering scanning electron microscopy (SEM). The phase state and strength evolution of pastes containing CFBFA were analyzed by Hlaváček *et al.* [12] using SEM–energy dispersive X-ray detector (EDX).

Fig. 10(a) shows an SEM image of the CEC (total scanning area: 1.5 mm²) and the grey levels of the various minerals. In the grey image, the left peak is from the visible pores, the intermediate peak is from the OFA and CFBFA mixed matrix, and the right peak is mainly from the OFA and CFBFA particles. CECs produced more hydration products after 60 d of hydration, resulting in a relatively dense microstructure with small porosity. However, the C–S–H gel matrix and ettringite could not be quantified. Therefore, the CEC miner-

als were analyzed by EDX. Fig. 10(b) shows the EDX maps of the CEC after 7 d and 60 d. Fig. 10(b) shows that the main minerals of the CEC are unreacted CFBFA, C–S–H gel, Ca(OH)₂/CaCO₃, and ettringite/Single sulfur calcium sulphoaluminate hydrate (AFm). The formation of ettringite is the main factor for the improvement of the early compressive strength of CECs based on the microstructure analysis of the CEC samples and is facilitated by CFBFA containing SO₃. C–S–H gel and Ca(OH)₂ are generated by cement hydration in the CEC. CFBFA differs from OFA because it contains f-CaO and SO₃. Additional Ca(OH)₂ and ettringite/AFm are produced by the reaction of f-CaO and SO₃ in the CEC. Therefore, the hydration mechanism of CECs prepared by CFBFA is different from that of OFA cement. However, SO₃ exists in CFBFA as CaSO₄. Although ettringite can be formed by CaSO₄, CaSO₄ can also be directly hydrated to form gypsum (CaSO₄·2H₂O), resulting in the volumetric expansion of CECs. Therefore, CaSO₄ (SO₃) in CFBFA should be used to promote the formation of ettringite to increase the early strength of CECs rather than CaSO₄·2H₂O, which can lead to harmful expansion [80].

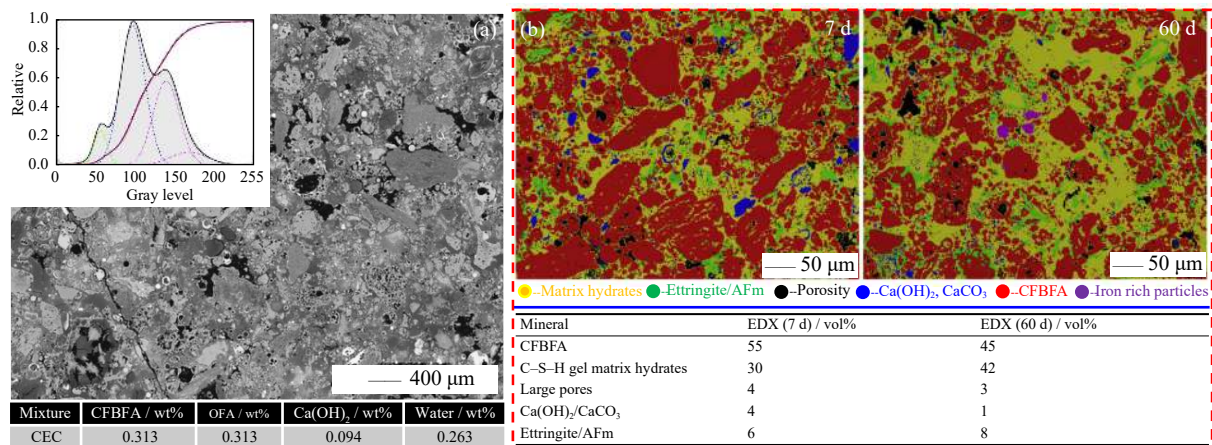


Fig. 10. Microstructural evolution of the compressive strength of CECs [12]: (a) SEM image of the CEC (total scanning area: 1.5 mm²) and the corresponding grey levels of the various minerals; (b) EDX maps of the CEC after curing 7 d and 60 d and corresponding quantitative EDX analysis results. Reprinted from *Cem. Concr. Compos.*, 90, P. Hlaváček, R. Šulc, V. Šmilauer, C. Rößler, and R. Šnop, Ternary binder made of CFBC fly ash, conventional fly ash, and calcium hydroxide: Phase and strength evolution, 100–107, Copyright 2018, with permission from Elsevier.

4.2. Microstructure affects expansion performance

CEC volume expansion is caused by CFBFA. Previous studies have explored the reason for this expansion. Among them, Chen *et al.* [72] studied the expansion mechanism of CEC based on microstructure, and some results are shown in Fig. 11. Fig. 11(a) shows that CEC paste cured in water for 28 d forms a dense microstructure. Many acicular and polygonal ettringite minerals were found in the matrix. However, Fig. 11(b) shows that curing CECs in ambient air for 28 d leads to incomplete hydration. Thus, the microstructure of the CEC is loose, and there are many voids between mineral particles. The lower crystallinity of ettringite in ambient air results in the lower expansion rate of CEC samples cured in ambient air versus water. Fig. 11(c) shows that the size of G0 (the mass ratio of cement to CFBFA is 0.4:0.6) particles is

larger, and the microscopic morphology is irregular, which lead to an uneven distribution of G0 hydration products after 1 d (acicular ettringite is still generated). However, Fig. 11(d) shows more CEC hydration products after 1 d and a uniform microstructure due to finer G3 particles filling the voids. These images indicate that the hydration reaction rate of the G3 sample at 1 d is higher than that of G0, which promotes the formation of ettringite in the CEC. However, the smaller pores in G3 only partially eliminated the expansion, resulting in a higher expansion rate of the CEC system. XRD results of CEC at hydration 180 d are shown in Fig. 11(e). The diffraction peak corresponding to Portlandite (Ca(OH)₂) is found in G0–G3, which is consistent with the formation of Ca(OH)₂ from the hydration of f-CaO, thus leading to the volume expansion of CECs later in the hydration period.

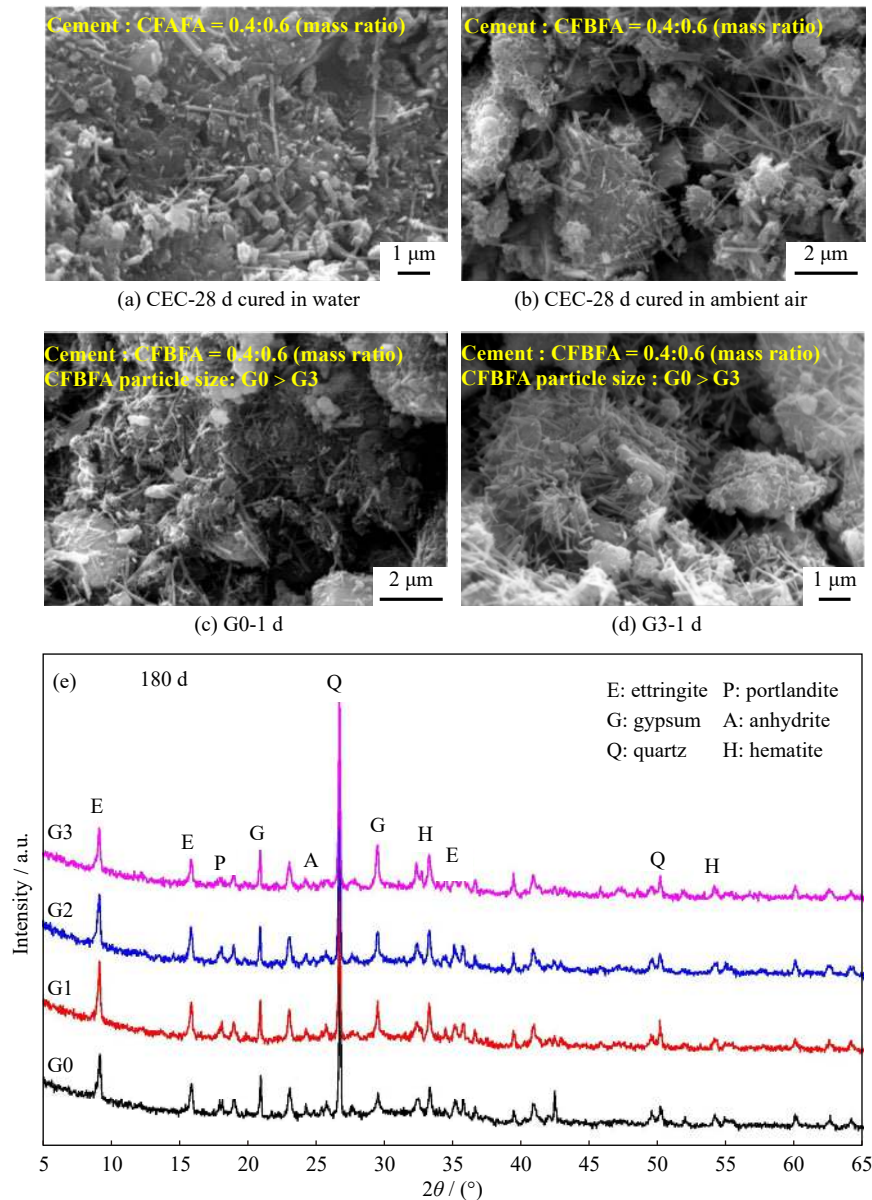


Fig. 11. (a–b) Microstructure of CEC after cured in water or in ambient air for 28 d; (c–d) microscopic morphology of CEC contains CFBFA with different particle sizes; (e) XRD results of CEC at hydration 180 d. Reprinted from *Constr. Build. Mater.*, 157, X.M. Chen, J.M. Gao, Y. Yan, and Y.Z. Liu, Investigation of expansion properties of cement paste with circulating fluidized bed fly ash, 1154–1162, Copyright 2017, with permission from Elsevier.

In conclusion, ettringite formation in CECs is promoted by CFBFA containing SO_3 based on microstructure analysis, which leads to early volume expansion of CECs. Meanwhile, the late volume expansion of CECs is mainly due to the hydration of f-CaO to form $\text{Ca}(\text{OH})_2$.

4.3. Microstructure affects rheological performance

Rheological properties are an important parameter in the cement processes. The particle size, morphology, and roughness of particles are important factors that determine the rheological properties of cement pastes. Internal particles move relative to each other when cement pastes flow. In parallel, the intergranular friction force of cement pastes is determined by the morphology and roughness of the particles. Therefore, the rheological microstructure of CECs has been

studied in detail based on the rheological properties of cement pastes. For example, Li *et al.* [78] conducted an in-depth study on the rheological properties of CEC pastes containing UCFA. Fig. 12 shows SEM images of P. I 42.5 cement, CFBFA, OFA, and UCFA particles. Fig. 12(a) shows an SEM image of P. I 42.5 cement particles. A small amount of spherical particles is in P. I 42.5 cement, and P. I 42.5 cement has a dense microstructure and smooth surface, which may be caused by high calcination. However, the formation temperature of CFBFA is low (850–900°C), and many clay minerals are not melted, which leads to the loose particle structure and rough surface microstructure of CFBFA in Fig. 12(b). Kwan *et al.* [81] reported that OFA had spherical particles (Fig. 12(c)), and the addition of OFA to cement produced lubrication, which resulted in a decrease of the yield

stress and viscosity. CFBFA has no spherical particles (Fig. 12(b)) compared with OFA. Therefore, the addition of CFBFA to the cement produced no lubrication, and the viscosity of the cement paste was increased by the CFBFA particles.

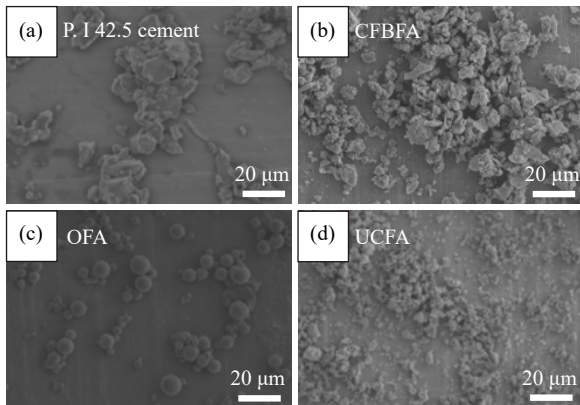


Fig. 12. SEM images of P. I 42.5 cement (a), CFBFA (b), OFA (c), and UCFA (d) particles [22,78]. (a), (c), (d) Reprinted from *Constr. Build. Mater.*, 188, D.L. Li, D.M. Wang, C.F. Ren, and Y.F. Rui, Investigation of rheological properties of fresh cement paste containing ultrafine circulating fluidized bed fly ash, 1007-1013, Copyright 2018, with permission from Elsevier. (b) Reprinted from *Constr. Build. Mater.*, 135, D.P. Zheng, D.M. Wang, D.L. Li, C.F. Ren, and W.C. Tang, Study of high volume circulating fluidized bed fly ash on rheological properties of the resulting cement paste, 86-93, Copyright 2017, with permission from Elsevier.

The frictional resistance between particles was increased by nonspherical particles, which hindered the flow of the particles [76]. Thus, the yield stress and viscosity significantly increased, and the rheological properties of the paste decreased [82–83]. Moreover, free water was absorbed by the loose particle structure of the CFBFA, and free water in the paste was reduced. According to the above analysis, the rheological properties of CECs are negatively affected by the shape and surface roughness of CFBFA. Therefore, the liquidity of CEC pastes was worse than that of P. I 42.5 cement pastes when the CFBFA content was slightly higher. However, Kwan and Li [81] demonstrated that CEC pastes mixed with UCFA (Fig. 12(d)) has significantly higher fluidity compared with OFA-cement pastes. Thus, the rheological properties of cement are affected more by CFBFA particle size than particle shape and roughness.

5. Conclusions and prospects

The mechanism of CFBFA in CECs depends on three characteristics: the active silica–alumina components, particle size, and the CaSO_4 phase. The active silica–alumina components of CFBFA form hydration products in cement, which contribute to the development of CEC compressive strength. The rheological properties of the CEC are improved by regulating the particle size of the CFBFA. The reaction of CaSO_4 in CFBFA forms ettringite, which is benefi-

cial for improving the early compressive strength of CECs.

At present, CFBFA is utilized in ecological cement mostly at the experimental stage. CEC strength is improved mainly by the active silica–alumina in CFBFA. Additionally, the influence of unstable components in CFBFA on the expansion of CECs has been explored in many studies. However, the utilization of the expansion characteristics of CFBFA in cement has not been reported. The rheological properties of CECs are controlled by regulating CFBFA particle size. Nonetheless, the cost of CECs increases after CFBFA particle size pretreatment.

There are remaining problems that need to be solved, and we propose future research directions based on these previous studies and considering comprehensively the utilization of CFBFA in cement. We think these suggestions are beneficial for the application of CFBFA in cement, which are summarized as follows.

(1) The utilization of the unstable SO_3 component in CFBFA needs to be investigated comprehensively. Some achievements have been made in utilizing SO_3 in cement, but the hazardous from the expansion of CaSO_4 into $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ has been ignored. The formation of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ must be inhibited to improve CFBFA utilization in cement.

(2) The long-term volume stability of cement is destroyed by the expansion of f-CaO, which limits the applications of CFBFA in cement. However, the volume shrinkage of cement may be compensated by the expansion of f-CaO. Therefore, the mechanism of shrinkage compensation must be analyzed.

(3) The economic benefits of CECs prepared by CFBFA should be considered. CO_2 emissions from the cement industry and costs can be reduced by reducing traditional cement usage and CFBFA pretreatment.

(4) Cement is a major building material for long-term applications, and its durability cannot be ignored. The impermeability, frost resistance, and erosion resistance of CECs should meet relevant standards. Different kinds of CFBFAs were pre-homogenized before the preparation of CECs. Additionally, the sufficient surplus coefficient of CEC performance enables the component fluctuation of CFBFA to be neglected. The reaction of unstable components in CFBFA could provide compensation to cement volume shrinkage, which can be beneficial to the durability. Thus, an appropriate amount of CFBFA could enhance the performance of cement.

(5) The environmental performance of CECs prepared by CFBFA cannot be neglected. While the mechanical properties, expansion performance, and rheological properties of CECs have been investigated, the evaluation of environmental safety is lacking. The leaching of harmful elements from CECs must satisfy WHO standards for drinking water.

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Conflict of Interest

The authors declare that this manuscript has no competing financial interests.

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