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Particles size distribution of quartz sand for AAC production

ABSTRACT: Quartz sand is generally used as a siliceous raw material for AAC production. The growth rate of tobermorite during autoclaving is controlled by dissolution of silica from quartz sand. And also, the theoretical proportion of dissolution of silica from quartz sand depends on Ca/Si ratio of raw materials. Therefore particle size distribution of quartz sand is very important for the production efficiency as well as the mechanical properties of AAC. In this paper, the optimal particle size distribution of quartz sand will be proposed.

KEY WORDS: quartz sand, particle size, crystalline, autoclave

1. Introduction

In AAC production, siliceous material such as quartz sand, slag or fly ash, calcic material such as cement and lime, water and foaming agent such as aluminum powder are used as raw materials. Quartz sand is generally used as a siliceous raw material after fine-grinding. The raw materials are mixed and cast in the moulds. After foaming and hardening, the green cake is cut in the desired products, followed by autoclaving process. During autoclaving, the growth rate of tobermorite-1.1 nm ($5CaO \cdot 6SiO_2 \cdot 5H_2O$) is controlled by dissolution of silica from quartz sand, resulting in the appearance of mechanical properties [1]. That is why quartz sand needs fine grinding and Ca/Si molar ratio of a bulk composition determined by raw materials mixture composition is usually lower than that of tobermorite (0.83). However, the mixture composition of containing calcium materials such as cement and lime is generally determined by production efficiency, in other word they govern the hardening period from casting to cutting. Therefore, the relationship of Ca/Si molar ratio of a bulk composition and particle size distribution of quartz sand is very important for AAC production.

Influences of particle size of quartz sand on the tobermorite formation and chemical and mechanical properties of AAC were reported [2, 3]. These papers reported that finer quartz sand reduces the autoclaving time, and on the contrary, coarser quartz sand showed higher mechanical properties. However, desirable particle size distribution of quartz sand for AAC production has not been reported yet. In this paper, the relationship of Ca/Si molar ratio of a bulk composition and particle size distribution of quartz sand is investigated.

2. Experimental procedure

2.1. Quartz sand and its particle size distribution

Three types of quartz sand produced by Sanei Silica Co., Ltd, Japan, SP-10, SP-35 and SP-80, whose brain surface areas are 1000, 3500 and 8000 cm²/g, respectively, were used.

They were mined from the same quartz mine and selected by grinding and sieving. Their particle size distribution and chemical composition are shown in Fig. 1 and in Table 1, respectively. The particle size distribution is measured by laser particle size analyzer, Nikkiso Co., Ltd, Japan, Microtrac MT-3300EXII. We prepared 12 different particle size distributions of quartz sand, whose 50% particle size ranges from 8 to 82 μm, by blending of SP-10, SP-35 and SP-80. The blending ratios and the particle size distributions are shown in Table 2 and Fig. 2, respectively.
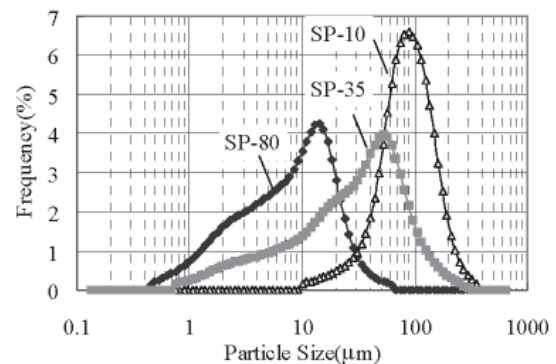

Fig. 1. Particle size distribution of SP-10, SP-35 and SP-80.

Table 1

CHEMICAL COMPOSITIONS (wt%) OF QUARTZ SAND AND CEMENT

	SiO_2	CaO	Al_2O_3	Fe_2O_3	Ig-loss
SP-10	97.1	-	1.09	0.75	0.30
SP-35	96.2	-	1.22	0.90	0.25
SP-80	95.7	-	1.63	1.08	0.39
Cement	20.8	64.4	5.7	2.7	1.9

2.2. Mixing ratio of raw materials

To simplify the relation of particle size distribution of quartz sand and Ca/Si molar ratio of raw materials, quartz sand, Portland cement, aluminum powder and water is only used for the experiment. Ordinary Portland cement provided by Japan Cement Association,
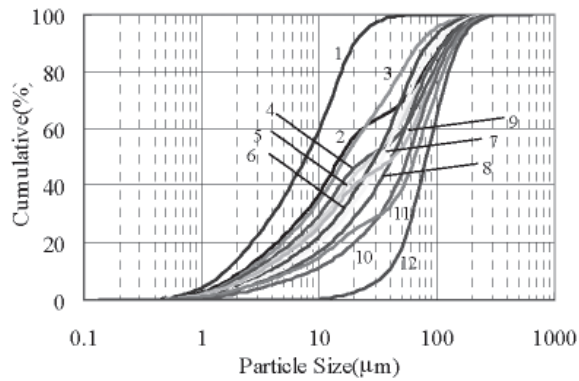
Fig. 2. Particle size distribution of blended quartz sand No.1-12 as shown in Table 1.

Table 2

BLENDING RATIOS OF SP-10, SP-35 AND SP-80, PARTICLE SIZE AND SiO_2(%).

No.	1	2	3	4	5	6	7	8	9	10	11	12
SP-10		3/8		1/2	1/3		1/10	1/4	4/7	1/2	3/4	1/1
SP-35			1/2		1/3	1/1	9/10	3/4		1/2		
SP-80	1/1	4/8	1/2	1/2	1/3				3/7		1/4	
10% (μm)	1.6	2.2	2.4	2.6	2.9	3.8	4.2	5.2	3.1	8.6	5.8	38.6
30% (μm)	4.0	7.4	8.2	10.2	11.4	14.8	16.9	21.1	12.4	33.0	33.3	62.4
40% (μm)	7.9	15.3	16.9	26.1	28.6	30.5	34.9	42.5	43.7	56.9	65.6	82.3
70% (μm)	12.7	52.9	38.0	72.0	62.7	50.2	56.3	66.3	79.6	83.2	93.9	108.1
90% (μm)	20.3	113.4	72.3	126.5	116.0	86.2	99.3	115.4	132.5	134.1	144.3	156.8
SiO_2 (%)	95.7	96.2	96.0	96.4	96.3	96.2	96.3	96.4	96.5	96.7	96.8	97.1

Table 3

THE MIXING QUANTITY OF RAW MATERIALS WITH WATER/SOLID AND CA/SI MOLAR RATIOS.

No.	Mixing quantity (g)				Water/Solid	Ca/Si mol
	Quartz	Cement	Water	Al powder		
Ca/Si = 0.35	118	63	120	0.09	0.66	0.343
Ca/Si = 0.50	100	81	120	0.09	0.66	0.494
Ca/Si = 0.65	86	95	120	0.09	0.66	0.638

aluminum powder produced by Yamato Metal Powder Co., Ltd and deionised water were used. Chemical composition of cement is shown in Table 1.

The mixing ratios of raw materials with water/solid and average Ca/Si molar ratios of No.1-12 are shown in Table 3.

2.3. Casting and curing conditions

The mixed slurry of raw material at a temperature of 41-42°C was cast in the 500 ml disposable mould. The curing condition was 50°C and 95% R.H. for 16 hours in a constant temperature and humidity chamber. Autoclaving condition was 180°C, 10 atm for 6 hours.

2.4. Analysis

To investigate the crystallization of tobermorite-1.1 nm, X-ray diffraction were measured by Rigaku Co., Ltd, Japan, RINT1000 under the conditions of 40 kV and 30 mA. The sum total of the 5-strongest XRD peak (hkl = 002, 004, 200, 220, 222) intensities of tobermorite-1.1 nm was analyzed quantitatively.

To investigate the durability, carbonation degree was measured. After cutting into φ12 mm x 5 mm, the specimens were carbonated under the conditions of 20°C, 75% R.H. and 3% CO_2 for 1 week. Carbonation degree was determined as shown in the previous study [4].

3. Results and discussion

3.1. XRD peak intensities and carbonation degree

The sum of the 5-strongest XRD peaks intensities of tobermorite-1.1 nm for each specimen were shown in Fig. 3. XRD peak intensities were higher in the samples with coarser quartz sand (the highest in No.12) at Ca/Si molar ratio of 0.35, were the highest at comparatively coarser quartz sand of No. 10 or 11 at Ca/Si molar ratio of 0.50 and were the highest at the middle quartz sand of 60 No. 4 to 9 at Ca/Si molar ratio of 0.65. Besides, No. 9 with
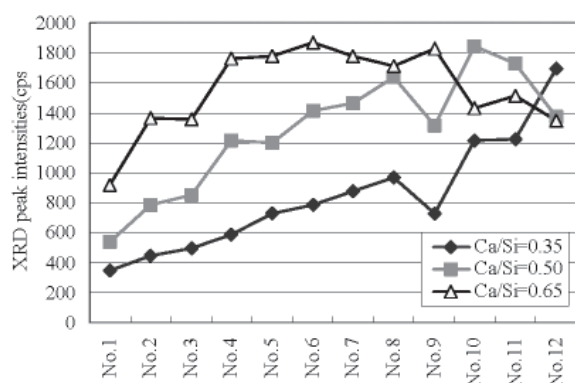
Fig. 3. The sum of the 5-strongest XRD peaks intensities of tobermorite-1.1 nm.
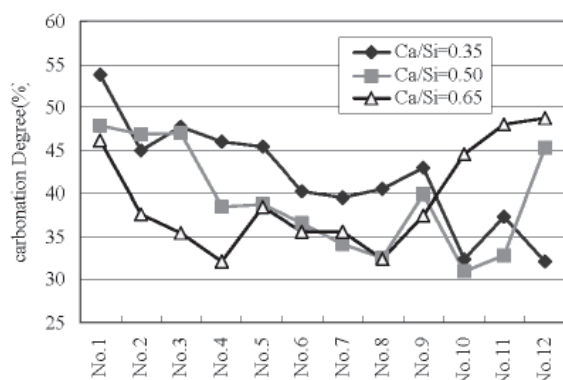

Fig. 4. Carbonation degree.

Ca/Si molar ratio of 0.35 and 0.50 was comparatively lower than adjacent to No. 8 and 10.

Carbonation degree for each specimen was shown in Fig. 4. Carbonation degrees were lower with the coarser quartz sand (lowest in No.12) at Ca/Si molar ratio of 0.35, were lowest at comparatively coarser quartz sand of No. 10 or 11 at Ca/Si molar ratio of 0.50 and were lower at the middle quartz sand of No. 2 to 9 at Ca/Si molar ratio of 0.65. Besides, No.9 with Ca/Si molar ratio of 0.50 was comparatively higher than adjacent of No. 8 and 10.

The results of carbonation had remarkable correlation with that of XRD. The specimen with high crystalline of tobermorite-1.1 nm was considered to have high durability against carbonation.

3.2. Dissolution of silica from quartz sand

The crux of the results above was that the optimal particle size distribution of quartz sand might be different for each Ca/Si molar ratio. For the lower and higher Ca/Si molar ratio, the coarser and finer particle size of quartz sand is suitable, respectively. It was considered that the demand of dissolution of silica from quartz sand during autoclaving depends on Ca/Si molar ratio of raw materials.

It was reported [5] that in the $CaO-SiO_2-H_2O$ system, the rate of dissolution of quartz sand with particle size of 10 to 20 μm under the autoclaving condition of 180°C and 10atm was 0.8 μm/h from the surface to the core, resulting in the formation of tobermorite. By utilizing this data, dissolution of silica from quartz sand during
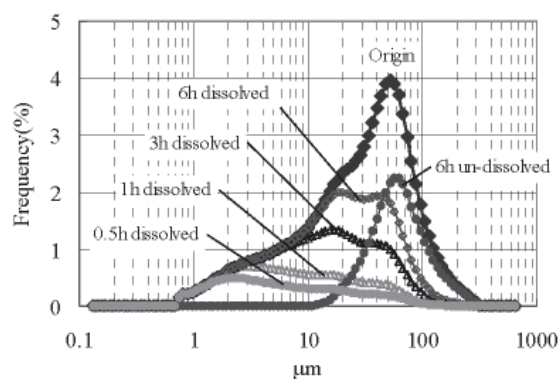

Fig. 5. Particle size distribution of origin and dissolved proportions at 0.5h, 1h, 3h and 6h autoclaving and un-dissolved proportion at 6h autoclaving of SP-35 (No. 6).

autoclaving was investigated with Ca/Si molar ratio of raw materials. The proportion of dissolved quartz sand was calculated by using the rate of dissolution of quartz sand of 0.8 μm/h, regardless of particle size and chemical composition under an assumption that the shape of quartz particles is spherical.

The calculated results of the particle size distributions of origin and dissolved proportion at 0.5h, 1h, 3h and 6h autoclaving as well as undissolved proportion at 6h autoclaving for SP-35 (No.6) were shown in Fig. 5. Until 0.5h or 1h autoclaving, finer particles have the main share of dissolved part. After 6h of autoclaving, the particle size under 15 μm were dissolved entirely and un-dissolved particles size was at around 10 to 100 μm. Fig.6 shows the relation of the proportion of dissolved quartz sand and autoclaving time for SP-35 (No.6). At the initial period of autoclaving, the rate of dissolution of silica is relatively high because of the dissolution of finer particles. After 6 h of autoclaving, the proportion of dissolved silica reaches approximately 65% at least, because the un-dissolved quartz sand still remains as shown in Fig. 5. That is why in efficient AAC production, Ca/Si molar ratio of a bulk composition of raw materials mixture is usually around 0.3 to 0.7, which is lower than that of tobermorite (0.83).
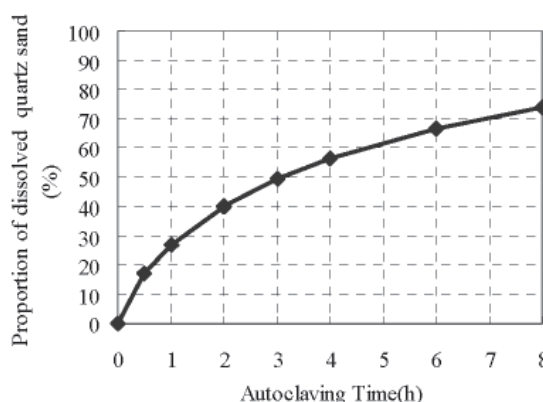

Fig. 6. Relation of dissolved quartz sand of autoclaving time for SP-35 (No. 6).

We calculated the share of dissolved quartz sand, shown in Fig. 7, taking into account SiO_2 content of quartz sand. Also, we calculated the demand of dissolved silica from quartz sand for each Ca/Si

molar ratio by solving the equation (1) of ideal Ca/Si molar ratio of components of raw materials and that of tobermorite.

$$\{(CaO\_cement)/56\}/\{(SiO_2\_cement + SiO_2\_dissolved\ quartz)/60\} = 0.83 \quad (1)$$

From this equation, the demand of silica from quartz sand is approximately 36% for Ca/Si molar ratio of 0.35, approximately 54% for Ca/Si molar ratio of 0.50 and approximately 74% for Ca/Si molar
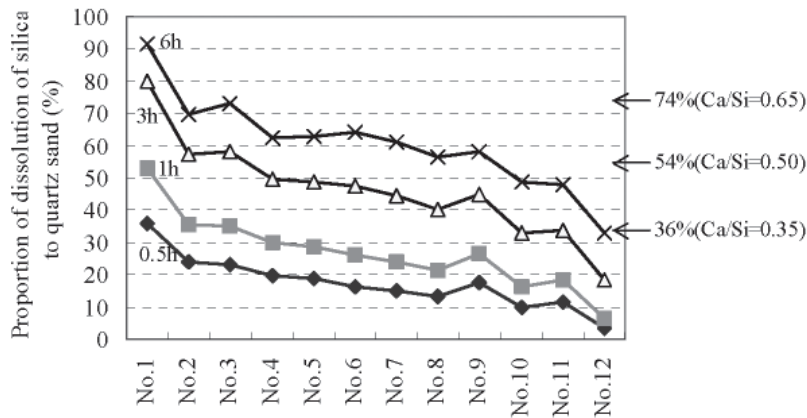

Fig. 7. Proportion of dissolved silica to quartz sand at 0.5h, 1h, 3h and 6h autoclaving.

ratio of 0.65. These values are also shown in Fig. 7.

From the results presented in Fig. 3 and 4, crystalline tobermorite and durability were the highest for coarser quartz sand of No. 12 for Ca/Si molar ratio of 0.35, for comparatively coarser quartz sand of No. 10 or 11 for Ca/Si molar ratio of 0.50 and for the middle quartz sand of No. 4 to 9 for Ca/Si molar ratio of 0.65. These results are considered to have high correlation with the share of dissolved silica from quartz sand at 6 h autoclaving as shown in Fig. 7. It is considered that higher content of crystalline of tobermorite-1.1 nm, resulting in higher durability, is obtained by fulfilling the demand of silica dissolved by selecting the proper particle size distribution of quartz sand. Although, it is considered that for Ca/Si molar ratio of 0.50 and 0.65 the rate of dissolution of quartz sand could vary depending on particles size, chemical composition, the shape of quartz grains and so on.
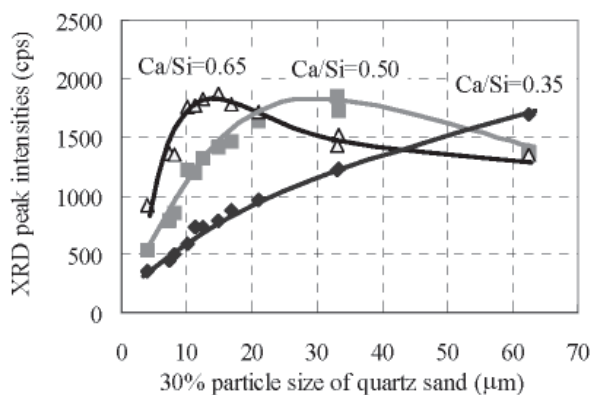

Fig. 8. Sum totals of the 5-strongest XRD peak intensities of tobermorite-1.1 nm and 30% particle size of quartz sand.

It is evident from Fig. 3 and 4 that crystalline and durability of No. 9 with Ca/Si molar ratio of 0.35 and 0.50 were comparatively lower than for the adjacent. It was distinctive for No.9 that the share of dissolved silica after 0.5 and 1 h autoclaving is much higher than for the adjacent. It is caused because No. 9 have high content of fine particles as shown in Table 2 and Fig. 2. It was reported [6] that the tobermorite formation using finer quartz sand of approximately 4 μm resulted in the smaller crystals with lower crystallinity. Therefore, it is suggested that too much content of fine quartz sand has to be avoided for AAC production.

3.3. A simple index for particle size of quartz sand

Although optimal particles size distribution of quartz sand is proposed above, it is complex and complicated to calculate for AAC production. However, widely used average or 50% of particles size is larger than the important particles size dissolved during autoclaving. Therefore, we would like to suggest 30% particles size of quartz sand to a simple and useful index for AAC production because the proportion of dissolved of silica after 6 h autoclaving was approximately 60% on the average as shown in Fig. 7.

Fig. 8 shows the relation of in total of the 5-strongest XRD peaks intensities of tobermorite-1.1 nm and 30% particles size of quartz sand. In this figure, the three Ca/Si molar ratios of 0.35, 0.50 and 0.65 have the relative maximum, which might mean optimal 30% particle size of quartz sand for each Ca/Si molar ratio. Fig. 9 shows the relation of Ca/Si molar ratio and optimal 30% particle size of quartz sand. By using this figure, it is easy to know a useful estimation of optimal particles size of quartz sand.
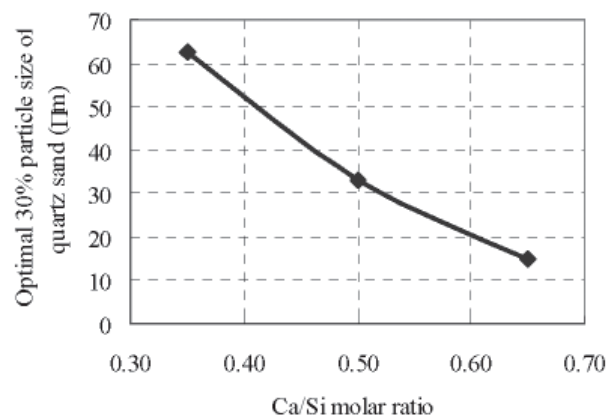

Fig. 9. Relation of Ca/Si molar ratio and optimal 30% particles size of quartz sand.

4. Summary

(1) Optimal particles size distribution of quartz sand was different for each Ca/Si molar ratio. For the lower and higher Ca/Si molar ratio, the coarser and finer particles size of quartz sand is suitable.

(2) Higher crystallization and durability could be obtained by the fulfilling of the demand of dissolved silica and by selecting the particles size distribution of quartz sand.

(3) Too much content of fine quartz sand has to be avoided in AAC production.

(4) 30% particle size of quartz sand is proposed to a simple index for AAC production.

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