Prediction of the Maximum Compressive Strength Geopolymers Using the Method of Weighted Chemical Compositions of Binders

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Abstract: Geopolymers are composite hard materials made by mixing solid binders such as fly ash and slag and alkaline liquid activators such as NaOH and sodium silicate. Geopolymers have recently been developed to be used as a replacement for Portland cement concrete. Industrial by-products such as fly ash, steel making slags and garbage melting furnace slags can be used to create geopolymers in a process that emits less carbon dioxide than in the cement making process. This reduction in CO_2 emission is important because CO_2 is one of the substances known to contribute to global warming. In the future, further uses of these fly ash and slags must be explored. The development of high compressive strength geopolymers using fly ash and slags will strongly contribute to the fields of construction, geotechnical engineering, and architecture. The compressive strength of geopolymers, q_u, is generally considered to be a function of the weight ratio of activator to binder, w, and the weight ratio of NaOH to sodium silicate, η. The maximum compressive strength, q_{umax} were determined as the maximum value among the peak values of q_u which were obtained for various values of w and η. The values of w and η which yield the maximum compressive strength, q_{umax} are defined as the optimum values w_{opt} and η_{opt} respectively. When designing the geopolymer works, it is essential to establish the method to predict the q_{umax} using the chemical compositions of binders only. This research is firstly to clarify the chemical and physical properties of geopolymer materials by using SEM and XRD observation, and

secondly the mechanical properties of the q_{umax}, and lastly to find out the method to predict the q_{umax} by combining three chemical compositions of SiO_2, Al_2O_3 and CaO in binders.

Introduction

More than 60 billion kgs of industrial by-products such as fly ash, steelmaking slags, and garbage melting furnace slags are generated every year in Japan (Koumoto, 2019). Fly ash and slags can be used to create geopolymers in a process that emits less carbon dioxide than in the cement making process. This reduction in CO_2 emission is important because CO_2 is one of the substances known to contribute to global warming. In the future, further uses of these fly ash and slags must be explored.

The chemical mechanism for hardening composite materials by mixing aluminosilicate binders, such as fly ash, and slags with alkaline activators, such as liquid NaOH and sodium silicate, is known as a geopolymer reaction. The hardened composite material is called geopolymer (Davidovits, 1979). Geopolymers are produced by mixing two components of solid binders like fly ash, slags, etc. and liquid activators like NaOH, KOH, sodium silicate and so on (Buchwald, 2006). Geopolymers have recently been developed to be used as a replacement for Portland cement concrete. The development of high compressive strength geopolymer using fly ash and slags will strongly contribute to the fields of construction, geotechnical engineering, and architecture.

Koumoto, 2019 has proposed the method to produce the high compressive strength geopolymers for the above-mentioned binders with an alkaline liquid mixture composed of NaOH and sodium silicate. He has clarified that the w_{opt} which is the optimum value of w (= weight ratio of liquid and binder) yielding the maximum compressive strength q_{umax} becomes a constant value of w_{opt} = 0.40 regardless of kinds of materials (see Fig. 1) and the η_{opt} which is the optimum value of η (= weight ratio of liquid NaOH and sodium silicate) yielding q_{umax} is expressed as a function of $C_{as}(= Al_2O_3 + SiO_2)$.

This paper describes firstly the chemical compositions, the morphological states, the crystallographic structures and the physical properties for 8 starting geopolymer materials. The mechanical characteristics of the high compressive strength geopolymers were studied to establish the method to predict the q_{umax} using the weighted chemical compositions of SiO_2, Al_2O_3 and CaO in binders. To correlate between q_{umax} and these three chemical compositions, the following weighted chemical composition method was introduced simply as: $q_{umax} = f(\chi) = Aexp(B \times \chi)$, where A and B are constants and $\chi = {SiO_2}^{\alpha} \times {Al_2O_3}^{\beta} \times CaO^{\gamma}$ in which α and β are smaller than 1.0, while γ is set as 1. The values of α and β that gives the highest correlation ratio were determined using trial and error.

Materials

Preparation of Geopolymer Materials

The finer the particle size, the greater the compressive strength of geopolymers (Koumoto, 2019). In the present tests, all slags which were provided in granular were ground after being air dried to a maximum particle size of 0.106 mm for effective chemical reaction.

Chemical compositions of Geopolymer materials

The chemical compositions of the geopolymer materials tested are obtained by the fluorometric analysis and listed in Table 1. The eight kinds of geopolymer materials (three Fly ash, two each of Slag1 (steel factory slags) and Slag2 (garbage melting furnace slags), and one Acidproof cement were the starting geopolymer materials, and the twenty three mixture materials which consist of the mixture of one Acidproof cement and Slag1, three Fly ash and Fly ash, eight Fly ash and Slag1, two Fly ash and Slag2, five Slag1 and Slag1 and four Slag1+Slag2) were prepared as geopolymer materials with a wide range of chemical compositions. In Table 1, geopolymer samples which are marked with * are data cited from Koumoto, 2019.

Physical properties of starting geopolymer materials

The physical properties such as density of particle ρ_s, loss of ignition and brain value for 8 starting geopolymer materials are listed in Table 2.

SEM observation

Preparation of 8 starting geopolymer materials for scanning electron microscopy observation were as follow. Materials were loaded onto carbon tape and sealed on the cylinder stub (Cat. No. 1255B-100, EM Japan Co., Ltd.). 5 nm-thickness of platinum coating was carried out (Dual Head Sputter Coater Cressington 208DS, Cressington Scientific Instruments Ltd.) before being transferred to the SEM. Images were obtained by scanning electron microscope (JSM-7900F, JEOL) at accelerating voltages of 5 kV.

Fig. 2 shows SEM pictures of 8 starting geopolymer materials (2000x magnification) listed in Table 1. Typical cenospheres were observed in the Fly ash Matsuura and Reihoku, however, not in the Fly ash Karita which showed similar morphology to other slag materials (even though chemical compositions (SiO_2/Al_2O_3) of Karita showed similar ratio to those in Fly ash Matsuura and Reihoku). Notable composition difference on Karita contain 500% more CaO and contain only half to 1/3 of Fe_2O_3 to those in Matsuura and Reihoku. Karita is the pressurized fluidized bed coal ash (PFBC ash) and accordingly Karita contains large amount of Ca component comparing to other Fly ash such as Matsuura and Reihoku (JIS ash). Fly ash Karita and other slag materials exhibit different morphology depending on their origin. Fig. 3 shows SEM pictures of Fly ash Matsuura and Reihoku (Top: x2000 & Bottom: x5000). Not only cenosphere but also pleroshpere (sphere in sphere) were clearly observed in Matsuura and Reihoku, but not in Karita. Addition to this, cenosphere found in Matsuura were not only smooth surface, but also rough and often perforated compared with Reihoku indicates that Matsuura is a mixture of magnetic cenosphere and plerosphore.

XRD observation

Fig.4 shows XRD pattern of different fly ash and slag samples. Fly ash Matsuura, Reihoku and Karita exhibit definite crystalline pattern. However, slag such as Koro, Kazusa, Narashino and Selament exhibit non-crystalline patterns which is typical of quenched slag samples and are therefore comparatively reactive in nature. Compared to this, the slag Stainless exhibits crystalline patterned and is considered comparatively less reactive (Das *et al*, 2020). Amorphous region can be observed in Fly ash Matsuura and Reihoku clearly. On the other hand, slag materials all showed very complex pattern with several overlapping peaks presumably resulting from mixture of many different minerals in the samples as previously reported

(Yildirim and Prezzi, 2011).

Fig. 5 shows the relationship between density of particle ρ_s for starting geopolymer materials (in Table 2) and each chemical composition: (a) SiO_2, (b) Al_2O_3 and (c) CaO (in Table 1). In Fig. 5 ρ_s largely decrease with an increase in SiO_2 and Al_2O_3 as well. As in Fig. 3 there are seen a lot of pleroshpere which is created from SiO_2 being origin of glass, ρ_s of Fly ash Matsuura and Reihoku become smaller. On the other hand ρ_s proportionally increases with an increase in CaO, accordingly, it might be said that the amount of CaO decides the densities of geopolymer materials.

Making Geopolymer Samples

According to Koumoto, 2019, the 48% NaOH (18mol/L) and sodium silicate ($Na_2O \cdot nSiO_2$, n=3.2) were used as activators in this research. Geopolymer samples were made for η ranging from 0.0 to 1.0 at a constant value of $w = w_{opt} = 0.40$. After mixing the activator and binder, the geopolymer samples were compacted in plastic molds of diameter D = 50 mm and height H = 100 mm. The geopolymer samples were removed from the molds after 1 or 2 days and cured at room temperature under dry condition for 28 days.

Test and Results

Compression tests

Before compression tests, diameter d, height h and weight W of all geopolymer samples were measured.

Compression tests of geopolymer samples were carried out at Saga Construction

Technology Support Organization (SCTSO) using the concrete testing apparatus in the same test method used for concrete samples.

The volume shrinkage ratio $\Delta V/V$ ($=(V-V')/V$) in which $V=\frac{1}{4}\pi D^2 H$ and $V'=\frac{1}{4}\pi d^2 h$ are volume of plastic mold and geopolymer sample at q_{umax}, respectively and the density of geopolymer sample at q_{umax} ρ_t (=W/V') were calculated.

Compression test results

Compression test results are shown in Fig. 6A for Fly ash, Acidproof cement, Fly ash+Fly and Fly ash+Slag1 and Fig. 6B for Fly ash+Slag1, Slag1+Slag1 and Slag1+Slag2.

As shown in Fig. 6A and Fig. 6B, q_u generally increase as η increase, and reach the maximum value q_{umax} at a certain η value, which is defined as the optimum value η_{opt}. The q_{umax} is defined as the peak value among q_u values obtained for several η values. The values of η_{opt} and q_{umax} from Fig. 6A and Fig. 6B are summarized in Table 3. Table 3 also includes the value of w_{opt} and both values of the density of geopolymer sample at q_{umax}.

Discussions

Correlation between η_{opt} and C_{as}(=Al_2O_3+SiO_2)

Because geoporimarization is a chemical reaction specially an aluminosilicate reaction, the amount of NaOH which contributes to ionize metals contained in the binder increases with the increase of the amount of Al_2O_3 and SiO_2. Accordingly, η, which is the weight ratio of NaOH to sodium silicate, is considered as the function of C_{as} (=Al_2O_3+SiO_2) (Koumoto, 2019).

Fig. 7 shows the correlation between η_{opt} from Table 3 and chemical compositions of binders C_{as} (=Al_2O_3+SiO_2) calculated from Table 1. In Fig. 7 the relationship between η_{opt} and C_{as} is well correlated by the straight line with a high correlation coefficient of r = 0.949 as

$$\eta_{opt} = 0.0160C_{as} - 0.453 \qquad (r = 0.949) \qquad (1a)$$

Equation (1a) is almost same to the equation (1b) which was proposed by Koumoto, 2019 as

$$\eta_{opt} = 0.0157C_{as} - 0.414 \qquad (r = 0.949) \qquad (1b)$$

From Fig. 7, the newly added fifteen compression test data points also support the equation (1b). The equation (1b) is newly proposed to calculate the η_{opt} value from the value of C_{as} of binders to very effectively produce high compressive strength geopolymers.

Chemical characteristic of q_{umax}

Fig. 8 shows the relationship between the q_{umax} and each chemical composition SiO_2, Al_2O_3 and CaO for 8 starting geopolymer materials. In the Fig. 2, plerosphere (sphere in sphere) were found in the Matsuura and Reihoku fly ash but not in Karita, in which support the lightweight of the materials (Fig. 3 & Table 3). This supports the fact that incorporation of high volumes of cenosphere reduces the density of composites but also reduce the mechanical properties of hardened composites (Xu et al, 2015). This was indeed the case that Matsuura and Reihoku fly ash showed significantly lower compression strength (q_{umax}) compared with Karita fly ash.

In Fig. 8 the q_{umax} decreases with increase of both SiO_2 and Al_2O_3, however increases with increase of CaO except the case of Slag Stainless. The case of Stainless might be by the reason which is judged from XRD observation being less reactive. According to ZHAO et al, 2010, in the case of Fly ash Matsuura the addition of CaO proportionally increased the q_{umax} to some content of CaO of 8-10 % in weight after that q_{umax} decreased with increase of CaO. It can be said that an increase of CaO contents in the geopolymer material contributes the increase of q_{umax} to some content.

As a result, it may be effective to correlate the factor C_{cas} (= CaO/C_{as}; C_{as}=SiO_2+Al_2O_3) with the mechanical properties of the high compressive strength geopolymers as Koumoto, 2019 has pointed out. Fig. 9 shows the relationship between the q_{umax} and C_{cas}. The q_{umax} increases with an increase in C_{cas} until C_{cas} reaches about 0.8 and after that the q_{umax} decreases with an increase in C_{cas}. There seems to have the q_{umax} being greater than about 150 N/mm^2 in the range of C_{cas} in between 0.6 and 0.9.

Volume shrinkage of geopolymer samples

Fig. 10 shows the relationship between the volume shrinkage ratio of geopolymer sample, $\Delta V/V$ at q_{umax} and C_{cas}. The values of $\Delta V/V$ were generally less than 2% when C_{cas} is less than 0.95 and increased sharply with increase in the factor C_{cas}. These results will be helpful for the design of geopolymer construction work.

Prediction of q_{umax} using the method of weighted chemical compositions of binders

When designing the geopolymer works, it is essential to predict the q_{umax} using the chemical compositions of binders only. Das, et al, 2000 has proposed to clarify the behavior of the q_{umax} of industrial by-products based geopolymer samples as dependent on Al/Si, Ca/Si and Ca/(Si+Al) ratios. In the research the values of SiO_2, Al_2O_3 and CaO are directly used to develop the q_{umax} equation.

Now although Fig. 9 shows the chemical characteristics of q_{umax}, there is no linear relationship to predict the value of q_{umax}. This section is to consider to combine the weighted three chemical compositions of SiO_2^{α}, $Al_2O_3^{\beta}$ and CaO^{γ} , where α, β and γ are powers and γ is set to be 1.0 as in binders and to correlate q_{umax} and $\chi = SiO_2{}^{\alpha} \times Al_2O_3{}^{\beta} \times CaO^{1.0}$ as the equation to decide the values of α and β by trial and error as:

$$q_{umax} = f(\chi) = Aexp(B \times \chi) \tag{2}$$

where $\chi = SiO_2{}^{\alpha} \times Al_2O_3{}^{\beta} \times CaO^{1.0}$ (3)

Fig. 11 shows the relationship between q_{umax} and χ in the case of $\alpha = \beta = \gamma = 1.0$. In the Fig. 11, the relationship between q_{umax} and χ is assumed to express as:

$$q_{umax} = Aexp(B \times \chi) \tag{4}$$

According to the least square method, the equation (4) is expressed with a rather high correlation coefficient as:

$$q_{umax} = 17.74\exp\,(9.48 \times 10^{-5} \times \chi) \qquad (r = 0.848) \qquad (5)$$

where $\chi = SiO_2 \times Al_2O_3 \times CaO$.

Now the values of α and β were determined by trial and error to yield the highest correlation factor r to the q_{umax} equation. The obtained values of α and β were 0.59 and 0.69, respectively.

Fig. 12 shows the relationship between q_{umax} and $\chi\ (= {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0})$. The q_{umax} equation according to least square method is expressed as:

$$q_{umax} = 15.71\exp\,(1.06 \times 10^{-3} \times \chi) \qquad (r = 0.940) \qquad (6)$$

where $\chi = {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0}$.

In Fig. 12, Eq.6 is drawn by the solid line together with break lines of error for $\pm 25\%$. Almost all the values of q_{umax} are plotted along the equation (6) in between the break lines of error for $\pm 25\%$..

From the design of geopolymer construction works point of view, the following equation (7) is recommended to estimate the values of q_{umax} for designing on safe side as:

$$q_{umax} = 11.71\exp\left(1.06 \times 10^{-3} \times \chi\right) \tag{7}$$

where $\chi = {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0}$.

Conclusions

This paper describes the chemical and mechanical characteristics of geopolymers by mixing fly ash and slags as binders with 48% NaOH (18 mol/L) and sodium silicate ($Na_2 \cdot nSiO_2$, n=3.2) as activators in which geopolymer samples were made for values of η from 0.0 to 1.0 at a constant value of w = 0.40.

The research results are summarized as:

(a) There is an optimum value, η_{opt} (the weight ratio of NaOH to sodium silicate) yielding q_{umax} for each binder,

(b) The η_{opt} is well correlated with the factor C_{as} (=Al_2O_3 + SiO_2) as:

$\eta_{opt} = 0.0160C_{as} - 0.453$. The value η_{opt} for an arbitrary binder necessary to manufacture the high compressive strength geopolymer is calculated by the above equation.

(c) The q_{umax} increases with an increase in C_{cas} until C_{cas} value of about 0.8 and after that q_{umax} decreases with an increase in C_{cas}. There seems to have the q_{umax} being greater than about 150 N/mm^2 in the range of C_{cas} in between 0.6 and 0.9.

(d) The density of the fly ash sample displayed the Matsuura and Reihoku showed significantly lower (~23%) than that of Karita. In the SEM analysis, plerospheres were found in the Matsuura and Reihoku fly ash but not in Karita, in which support the lightweight of the materials (Fig. 3 & Table 2). This supports the fact that incorporation of high volumes of cenosphere reduces the density of composites but also reduce the mechanical properties of hardened composites (Xu *et al*, 2015). This was indeed the case that Matsuura and Reihoku fly ash showed significantly lower compression strength (q_{umax}) compared with Karita fly ash.

(e) q_{umax} for Matsuura exhibited significantly low compared to not only other Fly ash samples but also to all the geopolymer materials. One interesting point is the chemical compositions of Matasuura that contain lower content of FeO and CaO which are similar percentile to those found in other Fly ash. This is contradicted to previously reported that magnetic cenosphere generally contain higher amounts of FeO, CaO (Sokol *et al*, 2000) indicating the influence of additional factors from this coal burning power plant.

(f) The volume shrinkage ratio $\Delta V/V$ is generally less than 2% when C_{cas} is smaller than 0.95.

(g) The q_{umax} equation is expressed with high correlation index r = 0.940 by using weighted chemical compositions of $\chi = {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0}$ as:

$$q_{umax} = 15.71 \exp\left(1.06 \times 10^{-3} \times \chi\right) \qquad \text{where } \chi = {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0}.$$

(h) For designing of geopolymer construction works the following equation is recommended to estimate the values of q_{umax} on safe side as:

$$q_{umax} = 11.71 \exp\left(1.06 \times 10^{-3} \times \chi\right) \qquad \text{where } \chi = {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0}.$$

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Data Availability

All data, models, and code generated or used during the study appear in the submitted article.

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Table 1. Chemical composition of tested geopolymer materials

Geopolymer material	Geopolymer sample	Chemical composition (%)						
		SiO_2	Al_2O_3	CaO	Fe_2O_3	MgO	SO_3	Mixture ratio of geopolymer samples:
Fly ash	Matsuura	63.3	22.8	3.8	3.7	0.8	0.5	
	Reihoku *	55.0	21.1	9.1	5.3	1.1	0.9	
	Karita *	38.8	24.3	19.5	1.5	0.5	6.6	
Slag1	Koro *	34.6	14.8	42.7	0.4	5.7	0.0	
	Stainless *	26.7	5.3	48.2	1.0	5.5	0.4	
Slag2	Kazusa *	34.2	13.2	42.0	2.6	1.9	0.7	
	Narashino *	34.2	13.9	39.3	3.7	1.8	0.6	
Acidproof cement	Selament	49.8	13.6	26.7	2.6	--	--	
A.cement and Slag1	SelaKoro	42.2	14.2	34.7	1.5	--	--	SelaKoro: Selament:Koro=1:1 by weight
Fly ash and Fly ash	KariRei *	46.9	22.7	14.3	3.5	0.8	3.8	
	KariRei2 *	49.6	22.2	12.6	4.1	0.9	2.8	
	MatRei	59.2	22.0	6.5	4.5	1.0	0.7	MatRei: Matsuura:Reihoku=1:1 by weight
Fly ash and Slag1	ReiKoro *	44.8	18.0	25.9	2.9	3.4	0.5	
	KoroRei5	51.6	20.1	14.7	4.5	1.9	0.8	KoroRei5: Koro:Reihoku=1:5 by weight;
	ReiKoro2 *	41.4	16.9	31.5	2.0	4.2	0.3	
	ReiKoro5	38.2	15.9	37.1	1.2	4.9	0.2	ReiKoro5: Reihoku:Koro=1:5 by weight
	KariKoro *	36.7	19.5	31.1	1.0	3.1	3.3	
	KoroKari5	38.1	22.7	23.4	1.4	1.4	5.5	KoroKari5: Koro:Karita=1:5 by weight
	KariKoro2 *	36.0	18.0	35.0	0.8	4.0	2.1	
	KariKoro5	35.3	16.4	38.8	0.6	4.8	1.1	KariKoro5: Karita:Koro=1:5 by weight
	KariKoro7	35.1	16.0	39.8	0.6	5.1	0.8	KariKoro7: Karita:Koro=1:7 by weight
Fly ash and Slag2	ReiKazu *	44.6	17.2	25.6	4.0	1.5	0.8	
	ReiNara *	44.6	17.5	25.6	4.5	1.4	0.8	
Slag1 and Slag1	StaKoro5	33.3	13.2	43.6	0.5	5.7	0.1	StaKoro5: Stainless:Koro=1:5 by weight
	StaKoro2	32.0	11.6	44.5	0.8	5.6	0.1	StaKoro2: Stainless:Koro=1:2 by weight
	StaKoro	30.7	10.1	45.4	0.7	5.6	0.1	StaKoro: Stainless:Koro=1:1 by weight
	KoroSta2	29.3	8.5	46.3	0.8	5.5	0.3	KoroSta2; Stainless:Koro=1:2 by weight
	KoroSta5	28.0	6.9	47.3	0.9	5.5	0.1	KoroSta5: Koro:Stainless=1:5 by weight
Slag1 and Slag2	KazuKoro5	34.5	14.5	42.6	0.8	5.1	0.1	KazuKoro5: Kazusa;Koro=1:5 by weight
	KazuKoro2 *	34.5	14.3	42.5	1.1	4.4	0.2	
	KazuKoro *	34.4	14.0	42.4	1.5	3.8	0.4	
	KoroKazu3	34.3	13.6	42.2	2.1	2.9	0.5	KoroKazu3: Koro:Kazusa=1:3 by weight

Note: *= Data from Koumoto (2019).

Table 2 Physical properties of starting geopolymer materials

Geopolymer materials	Fly ash			Slag1		Slag2		Acid-proof cement
Geopolymer sample	Matsuura	Reihoku	Karita	Koro	Stainless	Kazusa	Narashino	Selament
Density of particle(g/cm^3)	2.22	2.40	2.66	2.91	3.05	2.84	2.88	2.82
Loss of Ignition (%)	2.6	1.3	6.6	0.09	5.0	0.28	0.23	1.39
Brain value (cm^2/g)	4130	4020	5160	4180	5868	N/A	N/A	N/A
Specific surface (m^2/g)	N/A	N/A	N/A	N/A	N/A	1.55	0.83	3.33

Note: Data for Karita are from Kondo *et al*, 2014, Data for Stainless are from Takami *et al*, 2016.

Brain value and Specific surface were obtained by Brain method and BET method, respectively.

Table 3 Compression test results of geopolymer samples

Geopolymer material	Geopolymer sample	Compression test results				
		q_{umax}(N/mm^2)	w_{opt}	η_{opt}	ρ_t (kg/m^3)	ΔV/V (%)
Fly ash	Matsuura	15.1	0.40	1.00	1522	0.40
	Reihoku*	45.2	0.40	0.80	1868	0.20
	Karita*	104.3	0.40	0.50	1985	0.20
Slag1	Koro*	168.0	0.40	0.40	2227	1.20
	Stainless*	48.1	0.40	0.00	2269	11.34
Slag2	Kazusa*	110.0	0.40	0.30	2202	1.49
	Narashino*	86.4	0.40	0.50	2043	0.80
Acidproof cement	Selament	108.0	0.40	0.45	2160	1.10
A.cement and Slag1	SelaKoro	137.0	0.40	0.44	2218	1.49
Fly ash and Fly ash	KariRei*	70.1	0.40	0.65	1971	0.80
	KariRei2*	40.9	0.40	0.70	1986	1.00
	MatRei	38.5	0.40	0.87	1780	1.40
Fly ash and Slag1	ReiKoro*	79.3	0.40	0.60	2098	0.70
	KoroRei5	54.6	0.40	0.70	2174	2.00
	ReiKoro2*	118.0	0.40	0.60	2144	1.20
	ReiKoro5	130.0	0.40	0.50	2129	2.28
	KariKoro*	117.0	0.40	0.45	2122	0.90
	KoroKari5	83.4	0.40	0.40	2113	0.99
	KariKoro2*	159.0	0.40	0.43	2149	0.70
	KariKoro5	177.0	0.40	0.39	2183	1.09
Fly ash and Slag2	ReiKazu*	102.0	0.40	0.55	2081	1.39
	ReiNara*	94.3	0.40	0.65	2024	1.89
Slag1 and Slag1	StaKoro5	135.0	0.40	0.30	2208	0.80
	StaKoro	126.0	0.40	0.25	2243	3.26
	StaKoro2	136.0	0.40	0.10	2261	2.68
	KoroSta2	86.6	0.40	0.15	2296	6.17
	KoroSta5	45.8	0.40	0.20	2349	5.69
Slag1 and Slag2	KazuKoro*	132.0	0.40	0.35	2219	1.29
	KazuKoro2*	166.0	0.40	0.36	2218	1.49
	KazuKoro5	155.0	0.40	0.36	2249	1.49
	KoroKazu3	153.0	0.40	0.40	2180	2.08

Note: * Data from Koumoto, 2019; q_{umax}=maximum value of compressive strength q_u; w_{opt}=optimum value of w yielding q_{umax}; η_{opt}=optimum value of η yielding q_{umax}; ρ_t=density of geopolymer sample at q_{umax}; and ΔV/V=volume shrinkage ratio of geopolymer sample.

Appendix Notation

The following symbols are used in this paper:

A = constant,

B = constant,

C_{as} = Al_2O_3+SiO_2 (%),

C_{cas} = CaO/C_{as},

D = diameter of mold (mm),

d = diameter of geopolymer sample (mm),

H = height of mold (mm),

h = height of geopolymer sample (mm),

q_u = compressive strength (N/mm^2),

q_{umax} = maximum compressive strength (N/mm^2),

r = correlation coefficient,

V = volume of mold $\pi D^2H/4$ (mm^3),

V' = volume of geopolymer sample $\pi d^2h/4$ (mm^3),

w = weight ratio of solution of sodium hydroxide and sodium silicate to binder,

w_{opt} = optimum value of w at yielding q_{umax},

ΔV = amount of volume shrinkage of geopolymer sample V-V' (mm^3),

$\Delta V/V$ = volume shrinkage ratio of geopolymer sample (%),

α = power,

β = power,

γ = power (=1),

ρ_s = density of particle (g/cm^3),

η = weight ratio of sodium hydroxide to sodium silicate,

η_{opt} = optimum value of η at yielding q_{umax}

ρ_t = density of geopolymer sample at q_{umax} (kg/m^3)

φ = diameter of plate of loading

$\chi = SiO_2{}^{\alpha} \times Al_2O_3{}^{\beta} \times CaO^{1.0}$

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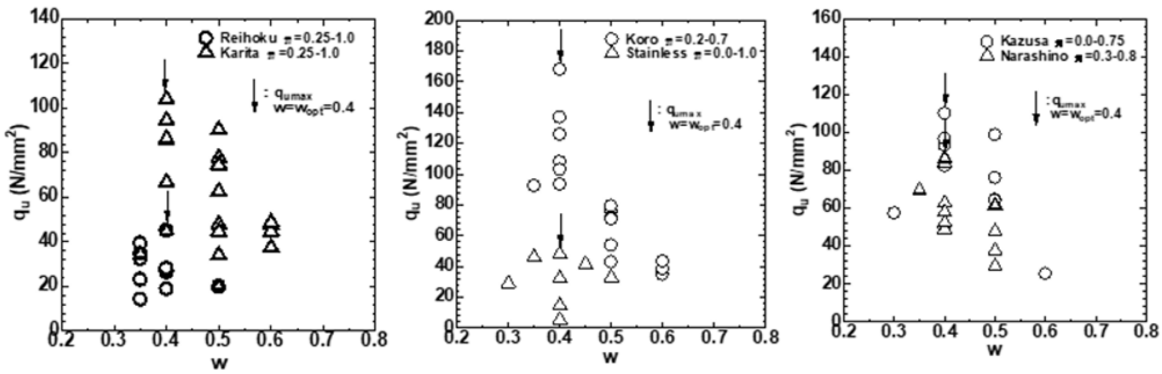
Fig. 11 Relationship between q_{umax} and $\chi\ (= SiO_2 \times Al_2O_3 \times CaO)$

Fig. 12 Relationship between q_{umax} and $\chi\ (= {SiO_2}^{0.59} \times {Al_2O_3}^{0.69} \times CaO^{1.0})$



(a) Fly ash **(b) Slag 1** **(c) Slag 2**

Fig. 1 Relationship between q_u and w for various value of η in each starting geopolymer materials: (a) Fly ash, (b) Slag 1 and (c) Slag 2

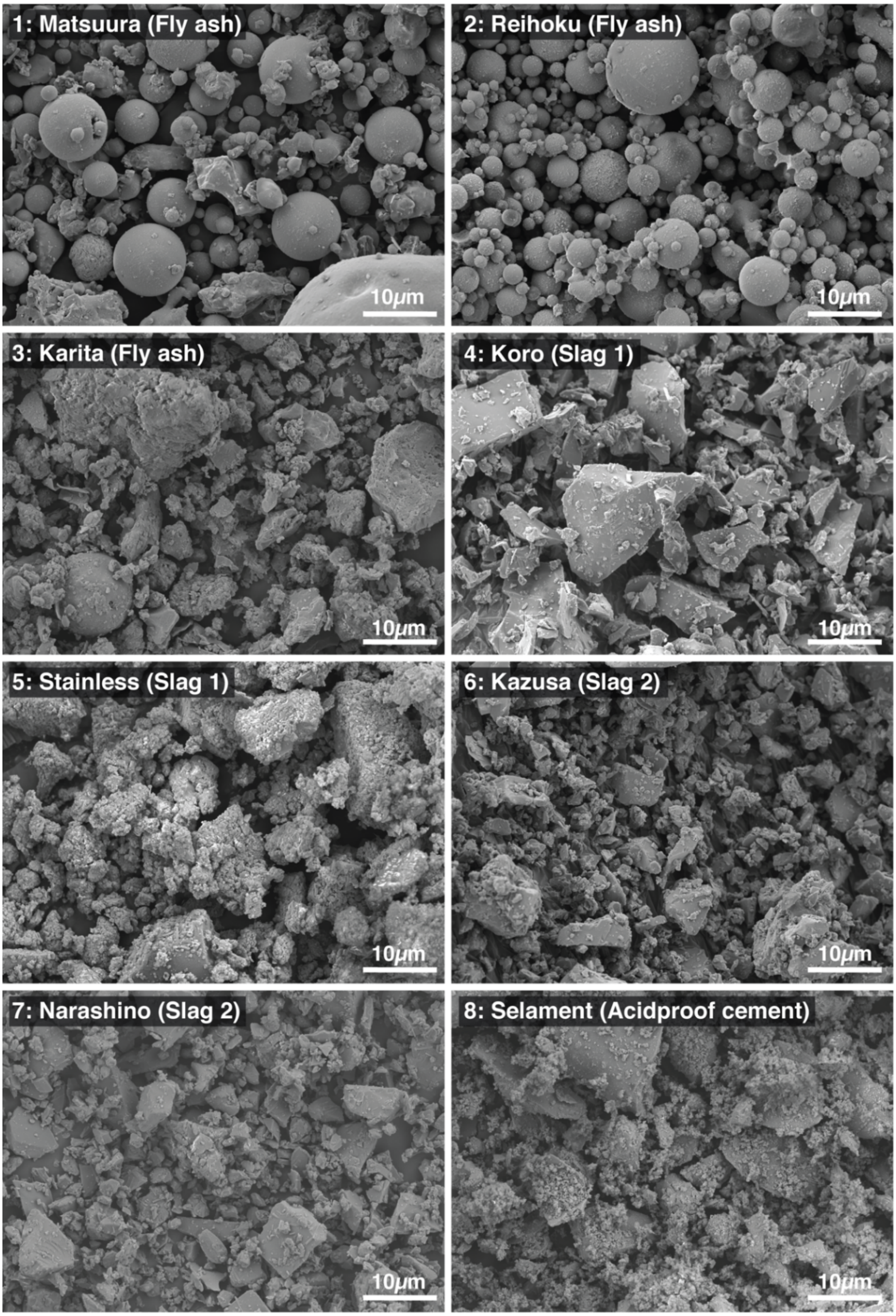


Fig. 2 SEM observation of 8 starting geopolymer materials (Mag x2000)

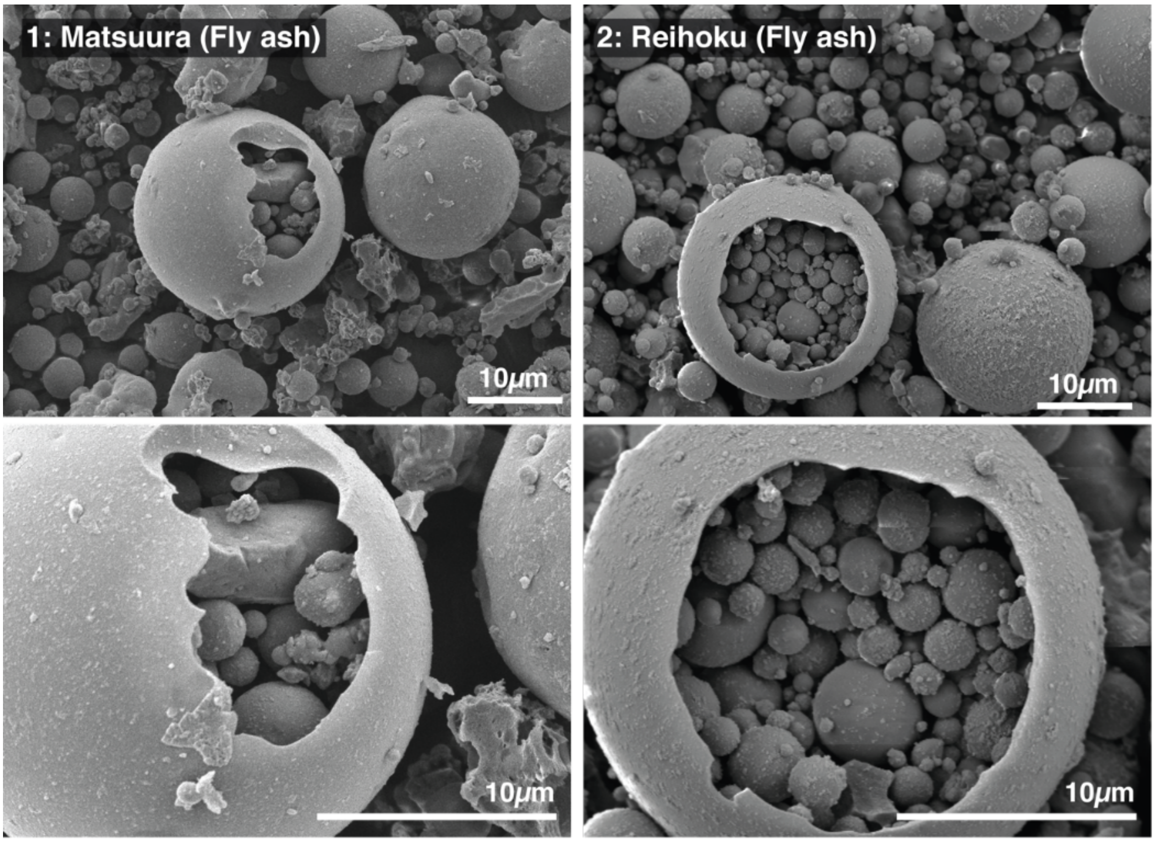
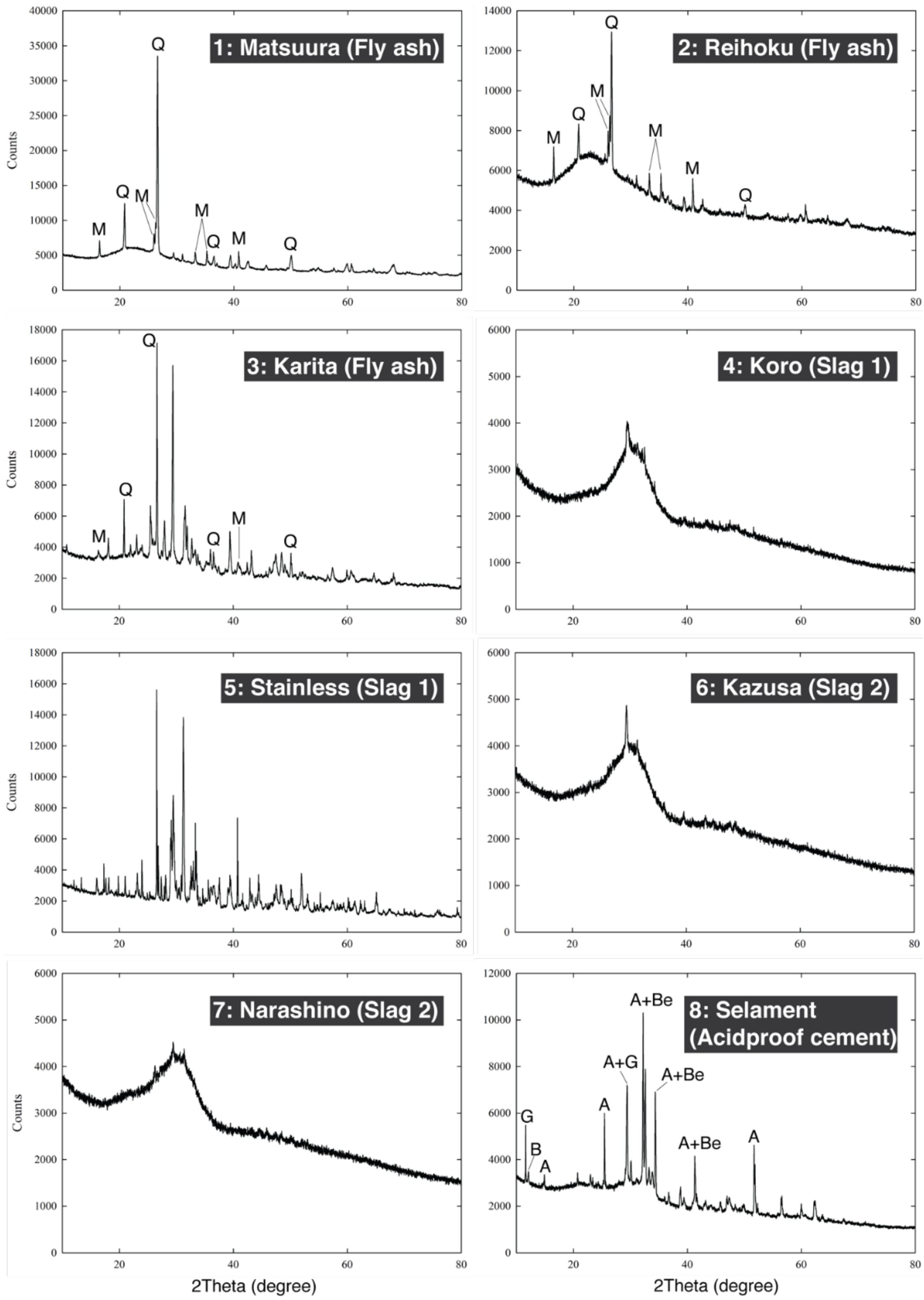


Fig. 3 SEM observation of Matsuura & Reihoku (Top: x2000 & Bottom: x5000)



(M=Mullite ($Al_6O_{13}Si_2$); Q=Quartz (SiO_2); G=Gypsum ($CaSO_4 \cdot 2H_2O$); B=Brownmillerite ($Ca_2(Al,Fe)_2O_5$); A=Alite ($3CaO \cdot SiO_2$); Be=Belite ($2CaO \cdot SiO_2$))

Fig. 4 XRD observation of 8 starting geopolymer materials SEM results

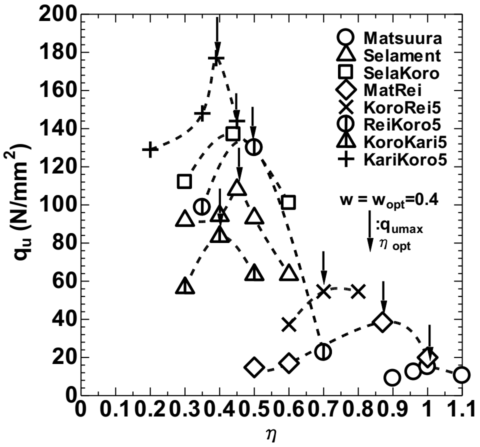


Fig. 6A Compression test results for geopolymer samples (Fly ash, Acidproof cement, Fly ash+Fly and Fly ash+Slag1)

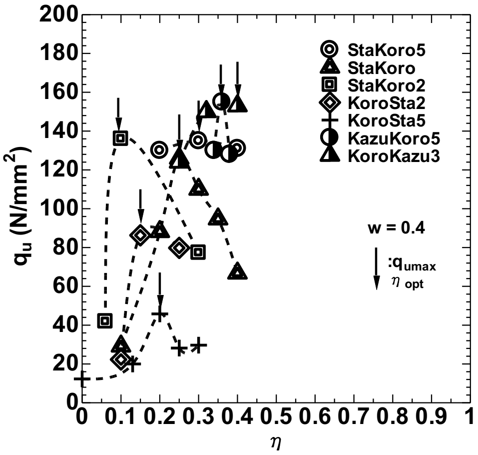


Fig. 6B Compression test results for mixed binder geopolymer samples (Fly ash+Slag1, Slag1+Slag1 and Slag1+Slag2)

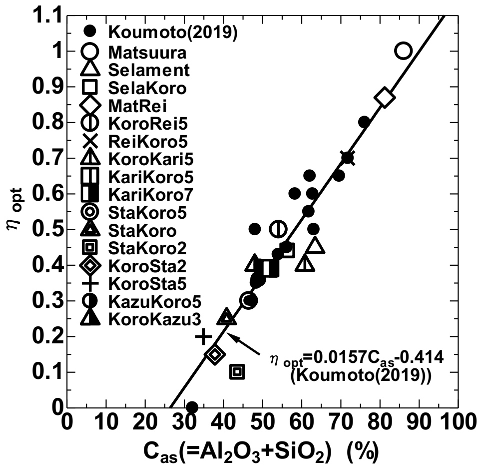
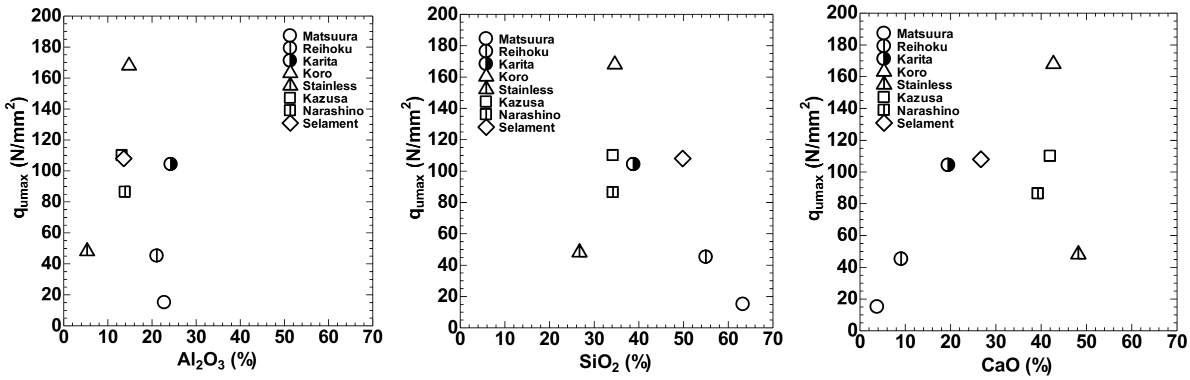


Fig. 7 Correlation between η_{opt} and C_{as}



(a) SiO_2 **(b) Al_2O_3** **(c) CaO**

Fig. 8 Relationship between q_{umax} and each chemical composition: (a) SiO_2, (b) Al_2O_3 and (c) CaO for 8 starting geopolymer materials

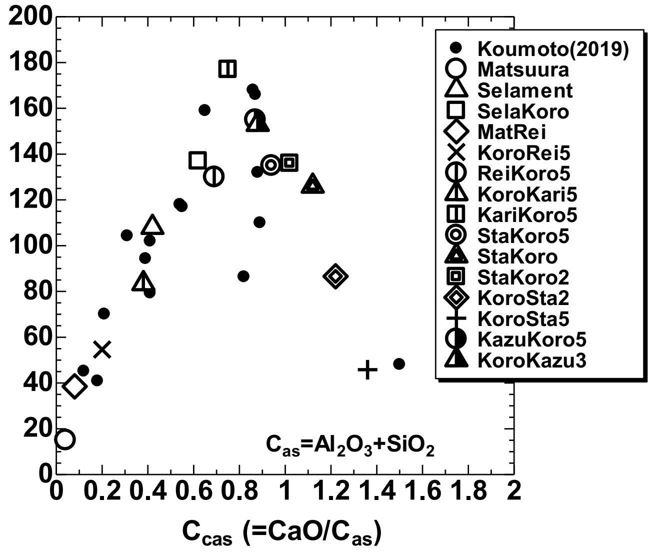


Fig. 9 Relationship between q_{umax} and C_{cas}

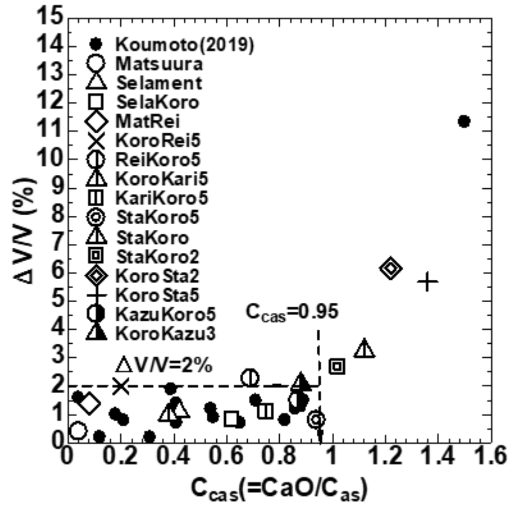


Fig. 10 Relationship between *ΔV/V* and C_{cas}

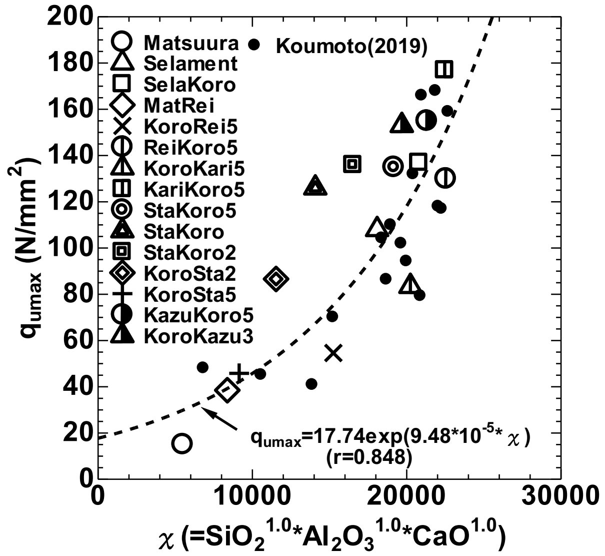


Fig. 11 Relationship between q_{umax} and χ (=$SiO_2 \times Al_2O_3 \times CaO$)

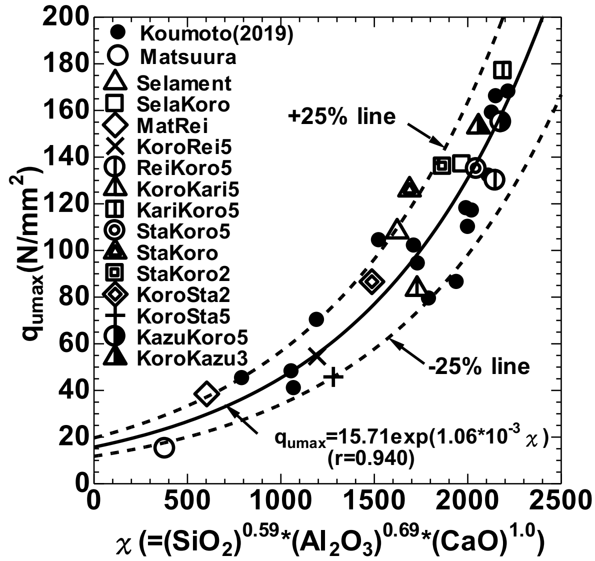


Fig. 12 Relationship between q_{umax} and χ (= $SiO_2^{0.59} \times Al_2O_3^{0.69} \times CaO^{1.0}$).