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Optimization of Fly Ash and Silica Fume as Materials Substitution for Cement with Ternary Sistem C-A-S

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Abstract: This paper had successfully evaluated the optimization of fly ash and silica fume which have a potential to be substituted partially with cement based on C-A-S ternary system. The application of C-A-S Ternary System optimization concept is based on two basic philosophies of how to formulate the equation of balance and substance fusion between the cement and fly ash or between the cement and silica fume. The formulation of those equations is being constructed in the form of C-A-S Ternary triangle ($\text{CaO}-\text{AL}_2\text{O}_3-\text{SiO}_2$). The optimization value is procured from the transformation of chemical parameter to mathematics trigonometry form. Results indicated that by using the diagram phase of C-A-S ternary System, the optimum percentage of fly ash is 18.84% whereas silica fume is 11.78%.

Key words: Fly Ash • Silica Fume • Optimization Ternary System • Substitution

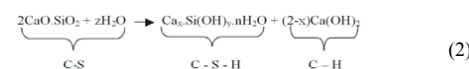
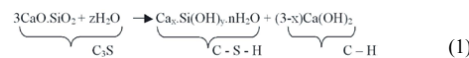
INTRODUCTION

This research is probing the aspect of microstructure substance in terms of material chemistry. The basic philosophy applied in this paper is the balance principle of the product of hydration reaction between major chemical compounds of cement tricalcium silica (C_3S) and dicalcium silica (C_2S). Cement that is mixed with water (H_2O) will form Hydrate Calcium Silica (CSH) and Calcium hydroxide (CH). Based from such philosophy, the usage of fly ash and silica fume is adopting the pozzolanic reaction concept which harness the potential of secondary reaction between the cement hydration products and the water like Calcium Hydroxide (CH). The main chemical compounds of fly ash and silica fume like silica fume (silicate oxide) will in turn produces secondary hydrate calcium silica (secondary CSH) [1]. Further analysis for applied philosophy and the concept of pozzolanic reaction has been processed by using the triangle system of calcium oxide (CaO) and silica oxide

(AL_2O_3) and silica oxide (SiO_2), know as C-A-S Ternary System [2]. The objective of this paper is therefore to optimize the fly ash and silica fume as materials substitution for cement using ternary system C-A-S.

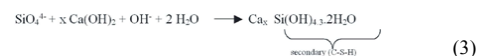
MATERIALS AND METHODS

Hydration Reaction of Cement: In determining the combined balance line of mineral with no calcium hydroxide ($\text{CH} = 0$), Tricalcium silica (C_3S) and dicalcium silica (C_2S) cement will react with water (H_2O) and forms Calcium Silica hydrate (CHS) and calcium hydroxide (CH) [1, 3].

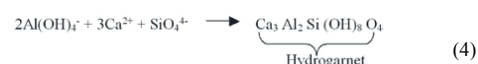


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The presence of pozzolanic materials will give an addition to ion silica which will react with calcium hydroxide (CH) to form secondary calcium silica hydrate (CSH) gel.



On the other hand, the aluminium (Al) that appears in pozzolanic material could possibly reacts with calcium hydroxide as well. This reaction will produce hydrogarnet phase (CASH).



The value of x that belongs to equations (1), (2) and (3) are the C/S ratio. *X-Ray* Diffraction (XRD) could be the alternative method to find the value of x. Components from the combined minerals are obtained as follows:

$$\left. \begin{aligned} S &= 1.S_{\text{CSH}} + 1.S_{\text{Hydrogarnet}} + 0.S_{\text{CH}} \\ C &= x.S_{\text{CSH}} + 3.S_{\text{Hydrogarnet}} + 0.S_{\text{CH}} \\ A &= 0.S_{\text{CSH}} + 1.S_{\text{Hydrogarnet}} + 0.S_{\text{CH}} \end{aligned} \right\} \quad (5)$$

In phase system ternary diagram, the balance of combined minerals is the summation of the components in equation (5)

$$C + S + A = 1 \quad (6)$$

Equation No. 5 indicates that if value of C/S = x = 1.65 at primary phase, C/S=3. A/S =1.0 at secondary phase, than the balance line's equation of combined minerals [2] should be:

$$A = \frac{(1 - 2.65S)}{2.35} \quad (7)$$

Formation Mixture Line of Cement with Fly Ash or Silica Fume: The result. obtained by using SEM (Scanning Electron Microscopy) equipped with ray EDAXnd device is an input to the formulation equation of cement material combining. Through the process of plotting the position into Ternary System triangle map with three parameters of main chemical compounds (SiO₂, Al₂O₃ and CaO). Other than that, those chemical compounds can be the input parameter in order to know the potential of pozzolanic characteristic [4-7].

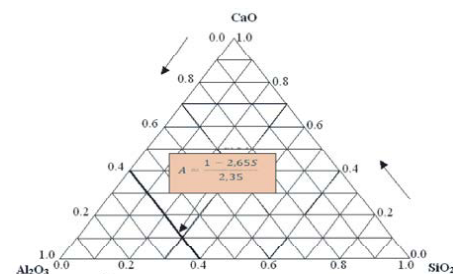


Fig. 1: Balance line's position of minerals CaO-Al₂O₃-SiO₂.

Table 1: The percentage of chemical compound C-A-S for cement + fly ash

C-A-S Composition (%) Cement + Fly ash			
Code	CaO	AlO ₃	SiO ₂
A1 (100-0)	72.19	4.73	14.16
A2 (90-10)	66.97	7.27	18.74
A3 (80-20)	62.87	7.77	24.36
A4 (70-30)	40.85	12.20	28.13
A5 (60-40)	37.85	12.17	29.44
A6 (50-50)	34.81	13.43	36.19
A7 (40-60)	25.52	17.95	41.14
A8 (30-70)	20.81	21.04	41.86
A9 (20-80)	20.41	21.74	42.59
A10 (10-90)	7.89	27.02	45.05
A11 (0-100)	4.20	33.48	52.83

Table 2: Percentage of chemical compounds C-A-S for cement + silica fume

C-A-S Composition (%) Cement + silica fume			
Code	CaO	AlO ₃	SiO ₂
B1 (100-0)	72.19	4.73	14.16
B2 (90-10)	66.32	2.08	23.43
B3 (80-20)	45.32	3.01	40.45
B4 (70-30)	38.85	3.03	46.08
B5 (60-40)	36.88	3.58	50.55
B6 (50-50)	31.54	4.69	59.08
B7 (40-60)	30.07	3.01	53.54
B8 (30-70)	11.32	2.51	69.72
B9 (20-80)	8.72	2.32	70.08
B10 (10-90)	2.77	3.61	79.40
B11 (0-100)	0.75	1.22	84.70

RESULTS AND DISCUSSION

Hidration Reaction of Cement: Equation No. 7 could be described in the phase system ternary diagram as a combined balance line to minerals with no content of calcium hydroxide (CH = 0) like what is shown in Figure 1 below:

Formation Mixture Line of Cement with Fly Ash or Silica Fume: The results of SEM/EDAX ray were shown in Tables 1 and 2 below:

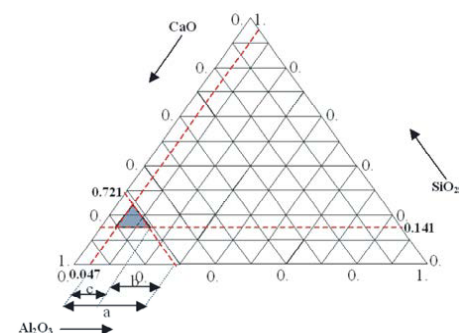


Fig. 2: The position between (0-100) on system ternary phase's diagram

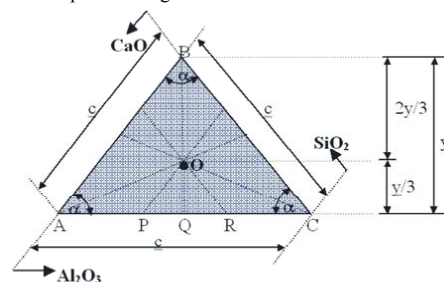


Fig. 3: The position of C100-0 at diagram of ternary's system phase (C-A-S)

Table 3: The position of material area of cement + fly ash

C-A-S Composition (%)			
Cement + Fly ash			
Code	a	b	c
A1 (100-0)	23.08	14.16	8.92
A2 (90-10)	25.76	18.74	7.02
A3 (80-20)	29.36	24.36	5.00
A4 (70-30)	46.95	28.13	18.82
A5 (60-40)	49.98	29.44	20.54
A6 (50-50)	51.76	36.19	15.57
A7 (40-60)	56.53	41.14	15.39
A8 (30-70)	58.15	41.86	16.29
A9 (20-80)	57.85	42.59	15.26
A10 (10-90)	65.09	45.05	20.04
A11 (0-100)	62.32	52.83	9.49

The result of SEM/EDAX ray based on Tables 1 and 2 were plotted into Ternary Triangle as shown in Figures 2 and 3 below:

Figure 2 above shows the material mixing position is forming an area. Those lines are required to be modified by determining the central area points in order to keep the position stated in Ternary system C-A-S coordinate.

Table 4: The position of material area of cement + silica fume mix

C-A-S Composition (%)			
Cement + Silica fume			
Code	a	b	c
B1 (100-0)	23.08	14.16	8.92
B2 (90-10)	31.60	23.43	8.17
B3 (80-20)	51.79	40.45	11.34
B4 (70-30)	58.12	46.08	12.04
B5 (60-40)	59.54	50.55	8.99
B6 (50-50)	63.77	59.08	4.69
B7 (40-60)	66.92	53.54	13.38
B8 (30-70)	86.17	69.72	16.45
B9 (20-80)	88.96	70.08	18.88
B10 (10-90)	93.62	79.40	14.22
B11 (0-100)	98.03	84.70	13.33

The following example is given in order to explain the modification method above using Al (100-0). Figure 2 illustrates the position of material area on Table 1 with chemical compounds 72.19% CaO - 4.73% Al₂O₃ - and 14.16% SiO₂ have been obtained:

$$a = (100 - \text{Al}_2\text{O}_3) - [100 - (100 - \text{CaO})]$$

$$= (100 - \text{Al}_2\text{O}_3) - \text{CaO}$$

$$= 100 - (\text{CaO} + \text{Al}_2\text{O}_3) \quad (8)$$

$$b = \text{SiO}_2 \quad (9)$$

$$= 100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2) \quad (10)$$

The percentage value of a, b and c can be obtained by inputting the value of CaO, Al₂O₃ and SiO₂

$$a = (100 - 4.73) - [100 - (100 - 72.19)]$$

$$= (100 - 4.73) - 72.19$$

$$= 23.08 \%$$

$$b = 14.16 \%$$

$$c = a - b$$

$$= 100 - (72.19 + 4.73 + 14.16)$$

$$= 8.92 \%$$

Plotting value (a), (b) and (c) resulted in Tables 3 and 4 below.

Since the sides of Ternary triangle are congruent the length of side (c) of C-1000 materials triangle are equal to α ($\alpha=60^\circ$) [8-9]. The details on the materials coordinate positions are shown in Figure 3.

Based on Figure 3, it is obtained

$$y = c \sin \alpha \quad (8)$$

Substituting the Equation(3) to the equation (4):

$$y = [100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)] \sin \alpha \quad (9)$$

Focus on the OPQ triangle and OQR. triangle:

$$\begin{aligned} \text{PQ} &= \frac{y/3}{\tan \alpha} \\ &= \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)] \cos \alpha}{3} \end{aligned} \quad (10)$$

$$\begin{aligned} \text{QR} &= \text{PQ} \\ \text{OP} = \text{OR} &= \frac{y/3}{\sin \alpha} \end{aligned} \quad (11)$$

$$= \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \quad (12)$$

Then,

$$\begin{aligned} \text{AP} &= \frac{c}{2} - \text{PQ} \\ &= \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{2} - \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)] \cos \alpha}{3} \end{aligned}$$

By $\alpha=60^\circ$:

$$\begin{aligned} \text{AP} &= \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \\ \text{CR} &= \frac{c}{2} - \text{QR} = \text{AP} \end{aligned} \quad (13)$$

$$\frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \quad (14)$$

Thus, the coordinates of C100-0 material in ternary system (C-A-S) are as follows:

$$\begin{aligned} \text{C} &= \text{CaO}_{\text{material}} + \text{CR} \\ &= \text{CaO}_{\text{material}} + \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \\ &= 72,19 + \frac{[100 - (72,19 + 4,73 + 14,16)]}{3} \\ &= 75.16\% \end{aligned} \quad (15)$$

$$\begin{aligned} \text{A} &= \text{Al}_2\text{O}_{3\text{material}} + \text{AP} \\ &= \text{Al}_2\text{O}_{3\text{material}} + \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \\ &= 4,73 + \frac{[100 - (72,19 + 4,73 + 14,16)]}{3} \\ &= 7.70\% \end{aligned} \quad (16)$$

Table 5: Point's coordinate of mixed material

Code	C-A-S composition(%) Cemen + Fly ash		
	CaO	AlO ₃	SiO ₂
A1 (100-0)	75.16	7.70	17.13
A2 (90-10)	69.31	9.61	21.08
A3 (80-20)	64.54	9.44	26.03
A4 (70-30)	47.12	18.47	34.40
A5 (60-40)	44.70	19.02	36.29
A6 (50-50)	40.00	18.62	41.38
A7 (40-60)	30.65	23.08	46.27
A8 (30-70)	26.24	26.47	47.29
A9 (20-80)	25.50	26.83	47.68
A10 (10-90)	14.57	33.70	51.73
A11 (0-100)	7.36	36.64	55.99

Table 6: Coordinate points of mixed material

Code	C-A-S Composition (%) Cement + Silica fume		
	CaO	AlO ₃	SiO ₂
B1 (100-0)	75.16	7.70	17.13
B2 (90-10)	69.04	4.80	26.15
B3 (80-20)	48.98	6.79	44.23
B4 (70-30)	42.86	7.04	50.09
B5 (60-40)	39.88	6.58	53.55
B6 (50-50)	33.10	6.25	60.64
B7 (40-60)	34.53	7.47	58.00
B8 (30-70)	16.80	7.99	75.20
B9 (20-80)	15.01	8.61	76.37
B10 (10-90)	7.51	8.35	84.14
B11 (0-100)	5.19	5.66	89.14

$$\begin{aligned} \text{S} &= \text{SiO}_{2\text{material}} + \text{OR} \\ &= \text{SiO}_{2\text{material}} + \frac{[100 - (\text{CaO} + \text{Al}_2\text{O}_3 + \text{SiO}_2)]}{3} \\ &= 14,16 + \frac{[100 - (72,19 + 4,73 + 14,16)]}{3} \\ &= 17.13\% \end{aligned} \quad (17)$$

The coordinates of every mixed material can be determined by inputting the data in Table 5 into Equations Nos. 15, 16, and 17.

Similarly, Table 6 could be calculated as well below:

Those coordinate points above are illustrated in ternary system diagram phase (C-A-S) forming a straight line as mixed material line with starting point A1.100-0 (100% cement) and point A11.0-100 (100% pozzolanic material) as illustrated in Figure 4.

Using a linear regression, a mixture can be determined as follow:

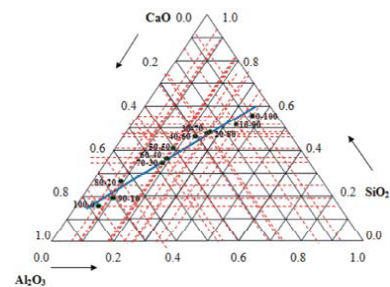


Fig. 4: Lines coordinate of cement + fly ash mix

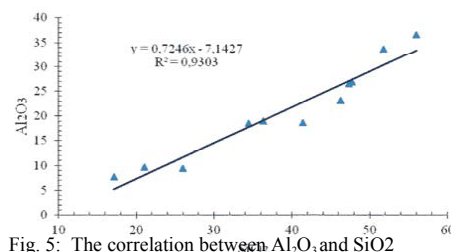


Fig. 5: The correlation between Al_2O_3 and SiO_2

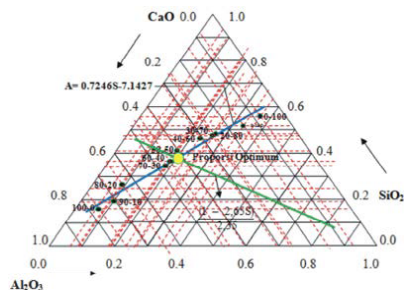


Fig. 6: Intersection point between mixing line with balance line

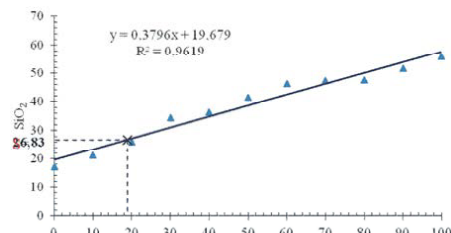


Fig. 7: The relationship between SiO_2 with increasing percentage (C + FA)

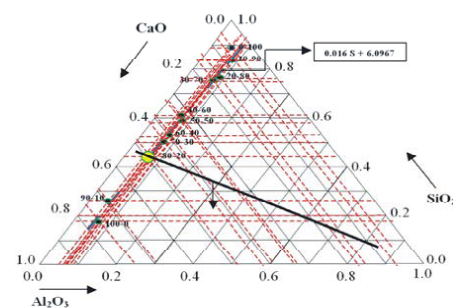


Fig. 8: Intersection between materials mixing line of cement + silica fume with minerals balance line

Based on Figure 5, an equation can be easily obtained as below:

$$A = 0.7246 S - 7.1427 \quad (18)$$

where A is Al_2O_3 chemical compound and S is SiO_2 chemical compound.

The Optimum Proportion of Cement + Fly Ash Mixture:
The optimum proportions are determined from an intersection point between mineral balance lines (Equation 7) with mixing lines by inserting Equation No. 18 as shown in Figure 6 [10-12].

Calculating the percentage of SiO_2 by substituting equation (18) and equation (7), the following equations can be obtained:

$$\begin{aligned} 0.7246 S - 0.071427 &= 1 - 2.65 S / 2.35 \\ (0.7246 S - 0.071427) \times 2.35 &= 1 - 2.65 S \\ (1.70281 S - 0.167853) &= 1 - 2.65 S \\ 1.70281 S + 2.65 S &= 1 + 0.16785 \\ S &= 0.2683 \\ S &= 26.83 \% \end{aligned}$$

By plotting value S, the relationship between SiO_2 with increasing percentage can be obtained as illustrated in Figure 7 below:

$$S = 0.3796x + 19.679 \quad (19)$$

By substituting the value of S to the Equation No. 19:

$$\begin{aligned} S &= 0.3796x + 19.679 \\ 26.83 &= 0.3796x + 19.679 \\ - 0.3796 x &= 19.679 - 26.83 \\ - 0.3796 x &= - 7.151 \\ X &= 18.84 \% \end{aligned}$$

Table 7: Test's results of X - Ray diffraction

Specimen Code	(%) Composition	Water (gr)	CSH* (%)	CH** (%)	Detail Composition
(1)	(2)	(3)	(4)	(5)	(6)
C + FA (1A)	90 - 10	104	64.38	35.62	
(1B)	95 - 15	104	67.73	32.27	
(1)	81.16 - 18.84	105	74.48	25.52	Optimum
(1C)	80 - 20	106	72.70	27.30	
(1D)	75 - 25	106	65.30	34.70	
C + SF (2A)	95 - 5	151	56.39	43.61	
(2B)	90 - 10	151	56.98	43.02	
(2)	88.22 - 11.78	152	66.20	33.80	Optimum
(2C)	85 - 15	153	65.30	34.70	
(2D)	80 - 20	153	58.40	41.60	

Note: * CSH: Calcium silicate hydrate ** CH : Calcium hydroxide

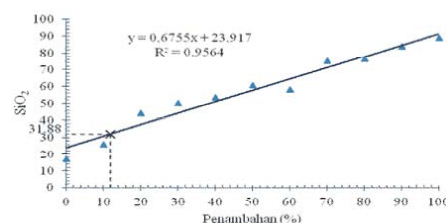


Fig. 9: Relationship between Silica Oxide (SiO₂) with the percentage increment (cement + silica fume)

Optimum Proportions of Cement + Silica Fume Mixture:

The optimum proportion is determined from the intersection's point between minerals balance line (Equation No. 7) with material mixing line as substitutions which was stated on Equation (0.016 S+6.0967) [13].

The equation to calculate the percentage of SiO₂ can be derived by substituting equation (0.016 S+6.0967) and Equation No. 7 as follow:

$$\begin{aligned}
 0.016 S + 0.060967 &= 1 - 2.65 S / 2.35 \\
 (0.016 S + 0.060967) 2.35 &= 1 - 2.65 S \\
 (0.0376 S + 0.143272) &= 1 - 2.65 S \\
 0.0376 S + 2.65 S &= 1 - 0.143272 \\
 S &= 31.88\%
 \end{aligned}$$

Plotting value S resulted in Figure 9 below:

An equation can be obtained from Figure 9 as:

$$S = 0.6755 X + 23.917 \quad (20)$$

By inserting the value of S to Equation (20), the value of "X" it can be obtained as follows:

$$\begin{aligned}
 S &= 0.6755x + 23.917 \\
 31.88 &= 0.6755x + 23.917
 \end{aligned}$$

$$\begin{aligned}
 - 0.6755x &= 23.917 - 31.88 \\
 - 0.6755x &= -7.960 \\
 X &= 11.78\%
 \end{aligned}$$

The Microstructures using Method of X – Ray

Diffraction: The optimum composition of mixing water at a normal consistency resulted in a hardened specimen. Such specimen was further tested by X – Ray diffraction to discover the maximum value of CSH and minimum CH (Table 7) [14-15].

CONCLUSION

Based on C-A-S Ternary's System, the optimum's percentage of fly ash for cement is 18.84 % while 11.78% for silica fume. Optimization using C-A-S Ternary System is one of the effective methods that could be used to find the mixtures optimum's percentage between cement and pozzolanic materials as well. The percentage value of optimum proportions obtained is a function of maximum quantity of calcium silicate hydrate (CSH) compounds and minimum quantity of calcium hydroxide (CH). Optimum mix proportion indicates the physical and mechanical behaviour of an optimal concrete. The concept used eliminates calcium hydroxide compound through pozzolanic reaction.

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