

Using Definitive Screening Design to Assess Factor Significance on the Compressive Strength and Volumetric Weight of a Ternary Blend Geopolymer

Martin Ernesto Kalaw^{1,*}, Ana Karmela Sumabat², Hoc Thang Nguyen³, Jonathan Dungca⁴, Florinda Bacani², Alvin Culaba¹, Susan Gallardo² and Michael Angelo Promentilla²

¹Mechanical Engineering Department, De La Salle University, Philippines

²Chemical Engineering Department, De La Salle University, Philippines

³ Faculty of Materials Engineering, Ho Chi Minh City University of Technology, Vietnam

⁴ Civil Engineering Department, De La Salle University, Philippines

*Corresponding Author: martin.kalaw@dlsu.edu.ph

Abstract: In the building sector which has been Portland cement (OPC)-based for the past century, geopolymers have emerged to have the potential to become the new norm. Its technical properties have been shown to be comparable if not better and its production results in as much as 80% reduction in CO₂ emissions compared to OPC. Moreover, sustainability is accessible since geopolymers, synthesized via alkali activation of amorphous alumino-silicate materials, can be formed from alumina- and silica- rich industrial and agro-industrial wastes such as coal ash and rice hull ash.

Synthesis of geopolymers are determined by three sets of factors: raw materials used (type, mineral composition, mix ratio, particle size, etc.), alkali activator (type, mix proportion, pH, liquid-to-solid ratio, etc.), and synthesis and curing conditions (curing time, curing temperature, applied pressure, etc.). These set of factors have to be identified for optimum properties of the geopolymers formed. As raw materials compositions are inherently variable, the effect of each factor cannot be taken absolutely.

In this study, the raw material mix ratio is set as 1:1:1 mass ratio of coal fly ash (CFA), coal bottom ash (CBA), and rice hull ash (RHA) and the curing time at 28 days. The order of significance of the effects of (1) NaOH/water glass ratio as alkali activator, (2) liquid-to-solid ratio, (3) curing temperature, and (4) particle size of bottom ash on the compressive strength and volumetric weight of the geopolymers formed will be determined. The Definitive Screening Design (DSD), a new, robust screening design that allows for three level tests at only 2m+1 runs (i.e. m = 4, 9 runs), will be used as the design of experiments.

Keywords: Green building materials, insulation, geopolymers, building simulation, carbon emissions

1. INTRODUCTION

1.1 Overview

The production and use of geopolymers as an alternative binder to ordinary Portland cement (OPC) consume much less energy and generates as much as 80% less greenhouse gas emissions (Davidovits, 1991; Barbosa et al., 2000). And its development from industrial waste materials such as blast furnace slag and coal ashes increases its potential for sustainability.

Optimized geopolymer production using different precursor materials have been able to produce alternative binders that are of comparable strength if not better, lighter, and with better fire and high temperature performance than OPC (Davidovits, 1994).

These present the prospect of geopolymer technology in replacing OPC as the binder of choice in the construction industry.

In this study, this prospect is considered by investigating the potential of using coal ash and

rice hull ash mixtures as raw materials for geopolymer production. Coal ashes (fly ash and bottom ash) and rice hulls are among the top agro-industrial by-products that are generally disposed of in landfills. The fly ash and bottom ash, mostly from coal fired power plants, have high alumina and silica content (Li and Xu, 2009). And the rice hull ash, from local biomass-fired power plants, is a rich source of amorphous silica (Siddique and Iqbal Khan, 2011).

1.2 Geopolymer Formation

Geopolymers are inorganic polymers formed from the alkaline activation of amorphous aluminosilicate materials resulting in a three-dimensional polymeric network. The products formed are the synthetic equivalent of natural rocks thus the term “geopolymer” (geo – meaning earth). As rock-like materials, they also possess properties similar to rocks such as hardness, chemical stability and longevity (Davidovits, 1994; Kumar et al., 2007).

The quality of the geopolymer formed depends on three sets of factors: the precursor materials used (type, mineral composition, mix ratio, particle size, etc.), the alkali activator used (type, mix proportion, pH, liquid-to-solid ratio, etc.), and synthesis and curing conditions (curing time, curing temperature, applied pressure, etc.). These set of factors have to be identified for optimum properties of the geopolymers formed. However, as the precursor materials compositions are inherently variable, the effect of each factor cannot be taken absolutely.

The precursor aluminosilicate materials can be divided into two main groups: (1) calcined materials, such as fly ash, metakaolinite, slag, construction residues, pozzolanic wastes, etc., and (2) non-calcined materials, for instance, kaolinite, feldspars, rock-type aluminosilicate minerals, mine tailings, etc. (Xu & van Deventer, 2003).

Of the common alkali activators, such as NaOH, Na₂SO₄, waterglass, Na₂CO₃, K₂CO₃, KOH, and K₂SO₄, the most utilized for geopolymer synthesis are a mixture of sodium or potassium hydroxides (NaOH, KOH) and sodium waterglass (nSiO₂-Na₂O) or potassium waterglass (nSiO₂-K₂O) (Pacheco-Torgal et al., 2007).

1.3 Factor Screening

Because of the many factors needed to be considered, testing a new formulation (precursor material mix) may involve a significant number of experimental/test runs. Thus it is imperative that the number of factors considered be as small as possible.

In this study, a ternary mix of coal fly ash (CFA), coal bottom ash (CBA), and rice hull ash

(RHA) is considered. Samples of these materials are shown in Figure 1. The proportion of the 3 materials, using a mixture design model, are three factors already. A fourth factor considered in the model is curing time.



Fig. 1. Precursor materials used in the ternary mix geopolymer.

For a manageable experimental design, other factors are set on a fixed value but these values need also be identified.

In this study, four additional factors are considered for significance:

- X1 = NaOH-water glass ratio (alkali activator)
- X2 = liquid-solid ratio
- X3 = curing temperature
- X4 = particle size of CBA

The evaluation of the order significance of these four factors on the geopolymer formed is evaluated using the Definitive Screening Design (DSD) run on the JMP software.

DSD is a small, robust screening design that allows for three level tests with only $2m + 1$ runs (m = number of factors). Thus for $m = 4$ factors, at 3 levels each, only nine runs are needed. In this screening design, the main effects are not biased by any second-order effect and all quadratic effects can be quantified. Figure 2 shows the color map of the factor correlations for a 4-factor DSD.

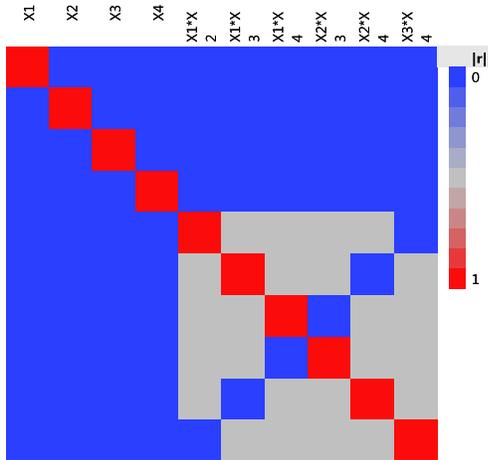


Fig. 2. Color Map on Correlations for 4 factor DSD (From JMP software)

For four factors, Table 1 shows the 9 runs of the Definitive Screening Design with three levels -1, 0, and +1 for each factor.

Table 1. Definitive screening design for 4 factors

RUN	X1	X2	X3	X4
1	0	1	1	1
2	0	-1	-1	-1
3	1	0	1	-1
4	-1	0	-1	1
5	1	-1	0	1
6	-1	1	0	-1
7	1	1	-1	0
8	-1	-1	1	0
9	0	0	0	0

2. METHODOLOGY

The definitive screening design (DSD) is used to determine the significance of four input factors:

- X1 NaOH/water glass ratio
- X2 liquid-to-solid ratio
- X3 curing temperature
- X4 particle size (of coal bottom ash)

on two response variables:

- S_c compressive strength and
- γ volumetric weight

of the geopolymers formed from a 1:1:1 mass ratio of coal fly ash (CFA), coal bottom ash (CBA) and rice hull ash (RHA).

Table 2 shows the 3 levels used for each factor in the DSD.

Table 2. Levels used for each factor in DSD

Factor	-1	0	+1
X1	Pure WG	50-50	75-25
X2	50%	60%	70%
X3	40°C	60°C	80°C
X4	Very fine	Fine	Coarse

In the formation of the geopolymer specimens, all coal ash used come from the same source and batch and the same is true for the rice hull ash. Cubical specimens (50mm x 50mm x 50mm) are formed using the factor levels shown in Table 2. The specimens remain in the molds for 24 hours, then de-molded and placed in an oven for another 24 hours, and then allowed to cure at ambient temperature for a total of 28 days. The process of specimen formation is shown in Figure 3.

3. RESULTS AND DISCUSSION

Table 3 shows the results of the material testing on the geopolymer specimens.

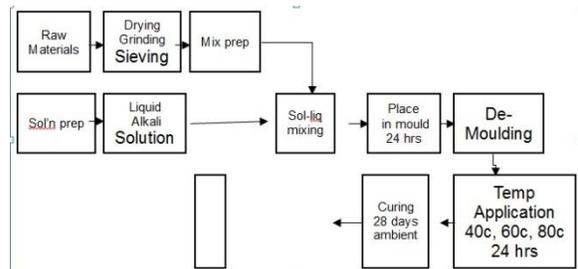


Fig. 3. Flow diagram of geopolymer specimen production

Table 3. Measured responses for each test run

RUN	X1	X2	X3	X4	S_c , MPa	γ , kg/m ³
1	0	+1	+1	-1	2.027	1130
2	0	-1	-1	+1	4.712	1365
3	+1	0	-1	-1	3.203	1226
4	-1	0	+1	+1	2.973	1274
5	+1	+1	0	+1	2.531	1237
6	-1	-1	0	-1	Br	1263
7	+1	-1	+1	0	4.718	1361
8	-1	+1	-1	0	1.260	1085
9	0	0	0	0	2.698	1221

Note: Br - broken specimen

Using the data in Table 3, Figure 4 shows the results of the screening for compressive strength. In this figure, it can be seen that the order of significance of the factors contributing to compressive strength is: liquid-solid ratio, NaOH-waterglass ratio, CBA particle size, curing

temperature with curing temperature having the least effect.

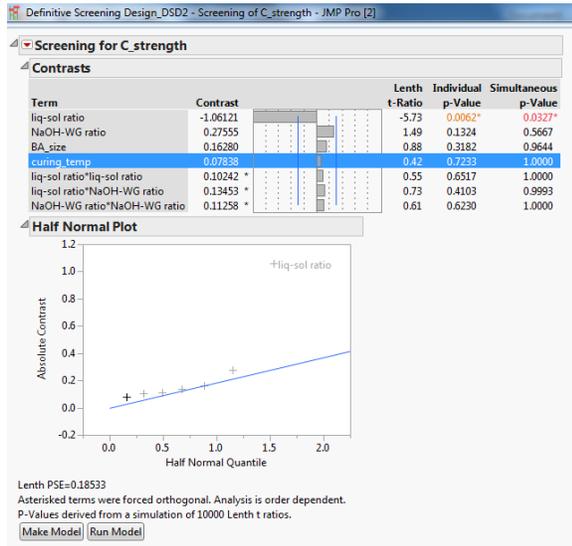


Fig. 4. Results of screening for compressive strength.

Using generalized regression model for compressive strength, the parameter estimates are shown in Figure 5.

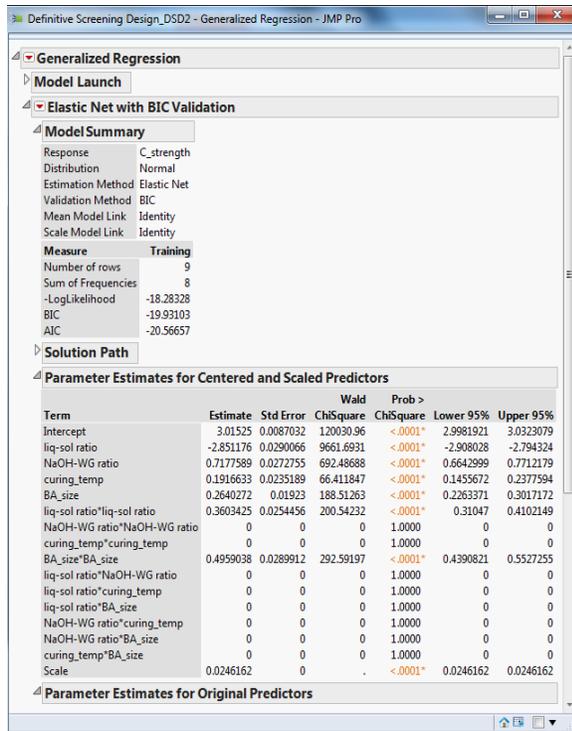


Fig. 5. Parameter estimates for compressive strength using generalized regression

Thus the compressive strength regression model is:

$$S_c = 3.01525 + 0.7177589X_1 - 2.851176X_2 + 0.1916633X_3 + 0.2640272X_4 + 0.3603425X_2^2 + 0.4959038X_4^2$$

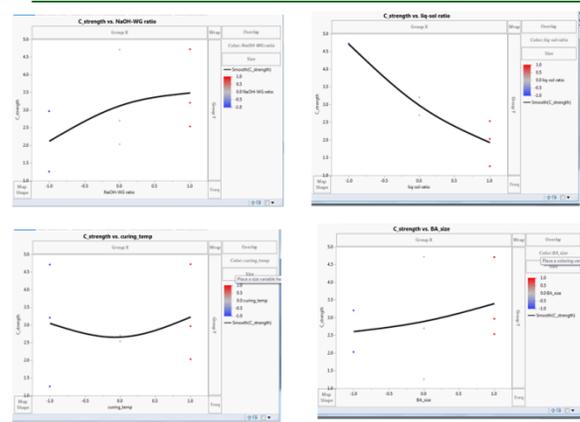


Fig. 5. Individual correlation of factors on the compressive strength (clockwise from top left: vs NaOH-WG, liquid-solid, BA size, curing temperature)

Comparing with the individual correlation of each factor, as seen from Figure 5, the compressive strength regression model captures the non-linear effects of X2 (liquid-solid ratio) and X4 (CBA particle size). Figure 5 also shows that except for X2 (liquid-solid ratio), increase in the other factor levels also increases the response (compressive strength).

Figure 6 shows the results of the screening for volumetric weight. In this figure, the order of significance of the factors contributing to volumetric weight is: liquid-solid ratio, CBA particle size, NaOH-waterglass ratio, curing temperature with curing temperature also having the least effect.

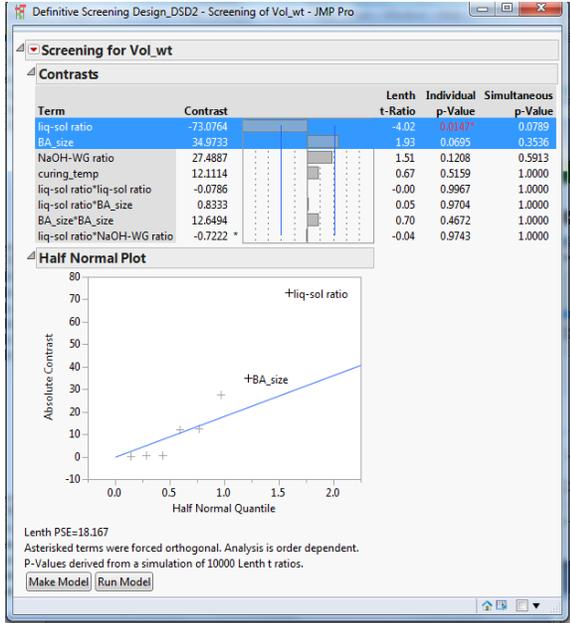


Fig. 6. Results of screening for volumetric weight

Using generalized regression model for volumetric weight, the parameter estimates are shown in Figure 7.

Thus, the volumetric weight regression model is:

$$\gamma = 1240.2222 + 80.273861X_1 - 217.037X_2 + 34.141805X_3 + 102.72752X_4 + 1.1075383X_1^2 + 35.755771X_4^2$$

Comparing with the individual correlation of each factor, as seen from Figure 8, the volumetric weight regression model captures the non-linear effects of X1 (NaOH-waterglass ratio) and X4 (CBA particle size). Figure 8 also shows that except for X2 (liquid-solid ratio), increase in the other factor levels also increases the response (compressive strength).

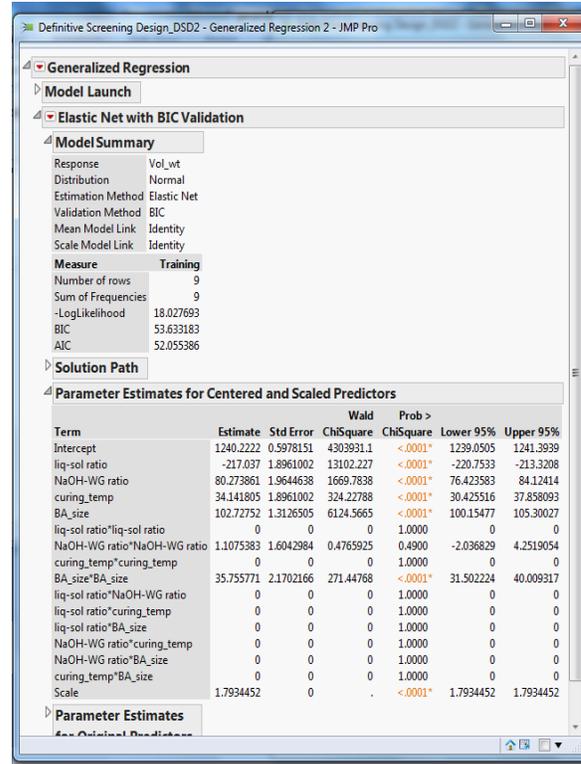


Figure 7. Parameter estimates for volumetric weight using generalized regression

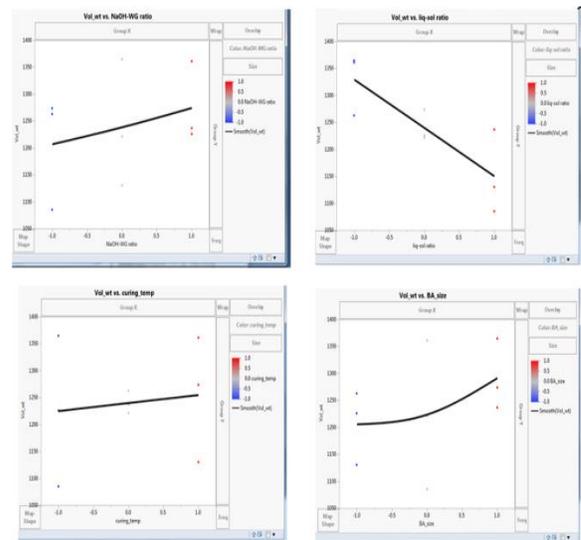


Fig. 8. Individual correlation of factors on the volumetric weight (clockwise from top left: vs NaOH-WG, liquid-solid, BA size, curing temperature)

4. CONCLUSIONS

The screening shows that curing temperature has the least effect on compressive strength and volumetric weight thus this factor may be excluded from the model.

Thus, the regression equations may be represented as:

$$S_c = 3.01525 + 0.7177589X_1 - 2.851176X_2 + 0.2640272X_4 + 0.3603425X_2^2 + 0.4959038X_4^2$$

and

$$\gamma = 1240.2222 + 80.273861X_1 - 217.037X_2 + 102.72752X_4 + 1.1075383X_1^2 + 35.755771X_4^2$$

The screening has also shown that for compressive strength, the liquid-solid ratio followed by the NaOH-waterglass ratio are the most significant factors while for volumetric weight, the liquid-solid ratio followed by the BA size are the most significant factors.

5. ACKNOWLEDGEMENTS

The authors express their appreciation to the ME, CHE and CIV Departments of De La Salle University for the use of their facilities and to JICA and AUN/SEEDNet for providing financial and technical support. Thanks are also extended to Mr. Khoi, Mr. Hien and the students and staff of the Ceramics Lab of the Ho Chi Minh City University of Technology for helping with the laboratory tests.

6. REFERENCES

- Barbosa, V., MacKenzie, K., & Thaumaturgo, C. (2000). Synthesis and characterisation of materials based on inorganic polymers of alumina and silica: sodium polysialate polymers. *Int. L. Inorg. Mater.*, 2, 309-17
- Davidovits, J. (1991). Geopolymers-inorganic polymeric new materials. *Journal of Thermal Analysis*, 37, 1633-1656, doi:10.1007/BF01912193.
- Davidovits, J. (1994). Properties of geopolymer cements, First International Conference on Alkaline Cements and Concretes, Scientific Research Institute on Binders and Materials, Kiev State Technical University, Kiev, Ukraine, 1994, pp. 131-149
- Kumar, S., Kumar, R., Bandopadhyay, A., & Mehrotra, S. (2007). Novel geopolymeric building materials through synergistic utilisation of industrial waste. *International Conference Alkali Activated Materials – Research, Production and Utilization*.

Li Hui and Xu Delong. (2009). The future resources for eco-building materials: II. Fly ash and coal waste, *Journal of Wuhan University of Technology-Mater. Sci. Ed.* Aug.2009

Pacheco-Torgal, F et al, “Alkali-Activated Binders: A Review”, *Constr Build Mater* (2007), doi: 10.1016/j.conbuildmat.2007.10.015.

Siddique, R., and Iqbal Khan, M. (2011). Supplementary cementing materials, *Engineering Materials*, DOI: 10.1007/978-3-642-17866-5_5, Springer-Verlag, Berlin

Xu, H. & van Deventer, J.S.F., (2003). Effect of Source Materials on Geopolymerization, *Ind. Eng. Chem. Res.* 42, 1698-1706