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Statistical Modelling of Ultrasonic Pulse Velocity of Fly Ash Based Geopolymer Mortar using Response Surface Methodology

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ABSTRACT

The fly ash based geopolymer has emerged as a capable and sustainable binder material in construction industry. Ultrasonic pulse velocity (UPV) method is a non-destructive technique for investigating the mechanical performance of concrete. Experimental investigation was performed for studying the effect of NaOH Molarity, Na₂SiO₃/NaOH and curing temperature on the ultrasonic pulse velocity of geopolymer mortar. Experiments were designed based on central composite design (CCD) technique of response surface methodology (RSM). Statistical model was developed and statistically validated and found significant as the difference between adjustable R-squared and predicted R-squared less than 0.2. Finally, the optimized mix proportion was assessed for maximized value of UPV. Experimental validation on the optimized mix reveals the close agreement between experimental and predicted values of UPV with significance level of more than 95%. The proposed technique improves the yield, the reliability of the product and the processes.

1. Introduction

World population is increasing continuously. In 2017, the it was 7.6 billion, however this figure will reach up-to 9.8 billion in 2050^[1]. The rapid increase in population will lead toward urbanization results in the growth of construction industry. Owing to the better mechanical properties and easy handling, concrete is mostly utilized building material in construction industry. Cement, sand and water are the main ingredients of concrete. However, the production of cement contributes in polluting the atmosphere. During the produce of one ton of cement, approximately 0.99 tons of carbon di-

oxide contribute into atmospheric gasses^[2]. Yearly, about 2.9 billion tons of cement is being consumed in construction industry. Hence cement industry alone is responsible for 10% of total greenhouse gas emissions^[3]. Furthermore, production of cement is also an energy extensive procedure. Approximately 3.1 to 5 GJ of energy is needed for the production of 1 ton of cement from the raw material^[2]. Cement industry utilized 12% of total energy used.

Geopolymer is an eco-friendly, sustainable and alternative binder material to OPC^[4]. The term geopolymer was firstly introduced by^[5]. Alkaline solution and base material are the main ingredients of geopolymer. Base material

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could be of geological origin or industrial waste material rich in alumina and silica. Polymerization takes place when aluminium and silicate elements present in base material, reacts with alkaline medium^[6]. Approximately 60% less energy is required to manufacture fly ash based geopolymer when compared to OPC industry^[7]. Furthermore reduction in atmospheric carbon dioxide emission of fly ash based geopolymer was 80% to the OPC^[8].

Non-destructive testing techniques (NDT) are getting popularity among the researchers for the measurement of mechanical and physical properties of concrete. These techniques include ultrasonic wave reflection^[9], Infrared thermography^[10], resonant frequency measurements^[11], Ultrasonic pulse velocity^[12] and spectral analysis of surface waves^[13]. However, ultrasonic pulse velocity test is considered as most reliable, quick and safe among other NDT^[14]. Magnitude of UPV can be used to accurately predict compressive strength, poisson's ratio and elastic modulus of concrete^[10]. Therefore, UPV is more compact and easy method to assess the quality of concrete.

Ultrasonic pulse velocity (UPV) is influenced by the verity of parameters especially those responsible for physical and mechanical properties of concrete. In their study Tarek Uddin investigate the variation of UPV of concrete using different types of aggregates and varying sand to aggregate ratio^[15], they concluded that aggregate type has significant influence on the UPV value and develop correlation of UPV with compressive strength and elastic modulus^[15]. Another study deals with the UPV of cementitious mortar having variable water to cement ratio (w/c). They concluded that by increasing w/c, the UPV decrease. Further they developed regression models to predict porosity and permeability of concrete with the help of UPV^[16]. In their study Tirupan Mandal et al. investigated the effectiveness of UPV value for the prediction of strength and modulus of cementitious stabilized materials CSMs. They proposed UPV as convenient and reliable method to assess the strength of CSMs^[17]. The effect of moisture content on UPV is studied by the Uldis Lencis et al. Higher UPV was observed for the samples with higher moisture content^[18]. Furthermore, concrete moisture content and UPV was correlated for the better understanding. Another study focuses on the effect of moisture content and heat on the UPV. They concluded that the increment in moisture content as well as temperature caused in higher UPV^[19].

There is enough literature available for Ultrasonic pulse velocity of cementitious concrete, however, research on the influence of different variables on the UPV of geopolymer is fledgling. In their study, Weibo ren investigated the effect of elevated temperature on UPV of geopolymer.

Temperature was varied from 200 °C to 800 °C with an increment of 200 °C. Morphological destruction in geopolymer with the increment in temperature results in decreased value of UPV^[20]. In another study, the addition of more alkaline solution contributes in strength development of geopolymer with higher UPV^[21]. Curing days and NaOH concentration has shown considerable influence on UPV of geopolymer concrete. Increasing magnitude of UPV was observed with the increase in curing days further NaOH concentration has shown variable influence on UPV^[12].

The main objective of this article is to investigate the influence of geopolymer primary variables (NaOH molarity, Na₂SiO₃/NaOH, curing temperature) on ultrasonic pulse velocity (UPV) using response surface methodology (RSM). ANOVA model was also developed for the prediction of UPV. Geopolymer mix was also optimized for maximum UPV. Experimental validation was performed on the Optimized mixture.

2. Experimental Program

2.1 Material

High calcium fly ash (HCFA) of Malaysian origin was utilized as a base material for the synthesis of geopolymer. Chemical characteristics of HFA are given in table 1. It is well noted that CaO was more than 10% for HCFA. Further, the summation of SiO₂, Al₂O₃ and Fe₂O₃ was less than 70%, Hence the fly ash confirms the basic requirement of high calcium fly ash as per ASTM 618-10.

Table 1. Chemical characteristics of fly ash

| Element | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | CaO | MgO | K ₂ O | SO ₃ | TiO ₂ | P ₂ O ₅ | LOI |
|------------|------------------|--------------------------------|--------------------------------|-----|------|------------------|-----------------|------------------|-------------------------------|-----|
| Percentage | 35.5 | 12.73 | 23.6 | 19 | 2.27 | 2.1 | 1.5 | 1.46 | 1.2 | 0.6 |

Note: LOI: Loss on ignition

As shown in Figure 1, FESEM image of HCFA reveals the spherical shape of fly ash particles.

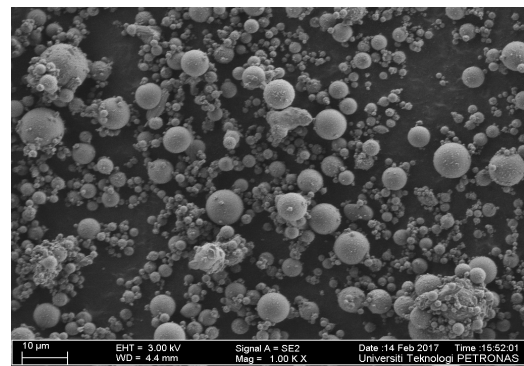


Figure 1. FESEM image of HCFA

Micro silica sand with maximum particle size of 710um was used as filler in geopolymer.

Mineralogy of HCFA as well as micro silica sand was observed by X-ray diffraction (XRD), shown in Figure 2.

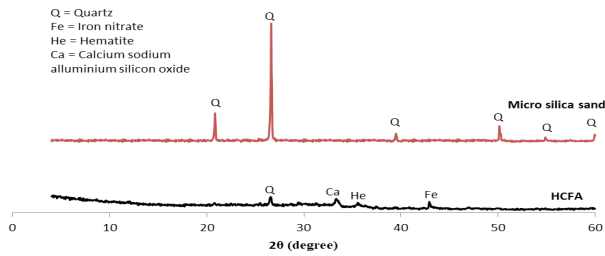


Figure 2. X-ray diffraction (XRD) of HCFA and micro silica sand

Sodium hydroxide of 99% purity was provided by R&M chemicals. Sodium silicate was supplied by Sino chemicals Malaysia. Chemical composition of Na₂SiO₃ are given in Table 2.

Table 2. Chemical composition of Na₂SiO₃

| Molecule | Percentage |
|-------------------|------------|
| Na ₂ O | 14.7 |
| SiO ₂ | 29.75 |
| H ₂ O | 55.52 |

2.2 Methodology

Influence of NaOH molarity, Na₂SiO₃/NaOH and curing temperature on the ultrasonic pulse velocity was investigated by 20mix designs proposed by central composite design (CCD) of response surface methodology (RSM). Commercially available software (design Expert®) was used to statistically design and analyse the experiments. For each mix design a group of three cylinders was casted and cured for 28 days.

2.2.1 Mixing Proportions and Specimen Preparation

A set of three geopolymer cylinders having length of 200 mm and diameter of 100 mm were casted for each mix design as shown in table 3.

Table 3. Experimental mix design

| Run | Factor 1 | Factor 2 | Factor 3 | Alkaline solution | Extra water | W/GP solids ratio | Fly ash | Sand |
|-----|--------------------|--------------------------|--|-------------------|-------------|-------------------|---------|------|
| | NaOH molarity M | Curing temperature °C | Na ₂ SiO ₃ /NaOH | | | | | |
| 1 | 10 | 105 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 2 | 10 | 67.5 | 0.48 | 0.38 | 0.039 | 0.23 | 1 | 0.3 |
| 3 | 20 | 67.5 | 1.75 | 0.38 | 0.096 | 0.23 | 1 | 0.3 |

| | | | | | | | | |
|----|----|------|------|------|-------|------|---|-----|
| 4 | 4 | 90 | 2.5 | 0.38 | 0.035 | 0.23 | 1 | 0.3 |
| 5 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 6 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 7 | 4 | 45 | 1 | 0.38 | 0.007 | 0.23 | 1 | 0.3 |
| 8 | 16 | 45 | 1 | 0.38 | 0.081 | 0.23 | 1 | 0.3 |
| 9 | 16 | 45 | 2.5 | 0.38 | 0.077 | 0.23 | 1 | 0.3 |
| 10 | 4 | 45 | 2.5 | 0.38 | 0.035 | 0.23 | 1 | 0.3 |
| 11 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 12 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 13 | 4 | 90 | 1 | 0.38 | 0.007 | 0.23 | 1 | 0.3 |
| 14 | 10 | 30 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 15 | 0 | 67.5 | 1.75 | 0.38 | 0 | 0.23 | 1 | 0.3 |
| 16 | 16 | 90 | 1 | 0.38 | 0.081 | 0.23 | 1 | 0.3 |
| 17 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |
| 18 | 10 | 67.5 | 3 | 0.38 | 0.060 | 0.23 | 1 | 0.3 |
| 19 | 16 | 90 | 2.5 | 0.38 | 0.077 | 0.23 | 1 | 0.3 |
| 20 | 10 | 67.5 | 1.75 | 0.38 | 0.054 | 0.23 | 1 | 0.3 |

Note: alkaline solution, Extra water and sand are the mass ratio of fly ash.

NaOH molarity, curing temperature and Na₂SiO₃/NaOH were determined by RSM. Fly ash, sand, W/GP solids and alkaline solution were selected from available literature^[22]

2.2.2 Ultrasonic Pulse Velocity Equipment

The equipment used for measurement of UPV consisted of electrical pulse generator and electro-acoustical transducer as the main components. Ultrasonic pulse was transmitted from the concrete by a transducer. The pulse was received by a transducer placed on the opposite end. Measurement was taken in the form of pulse travelling time between two transducers. Pulse travelling time was converted into pulse velocity by using equation 1.

$$UPV = L/T \tag{1}$$

Where

V= Pulse velocity (m/s)

L=Distance between centre of transducer faces (m)

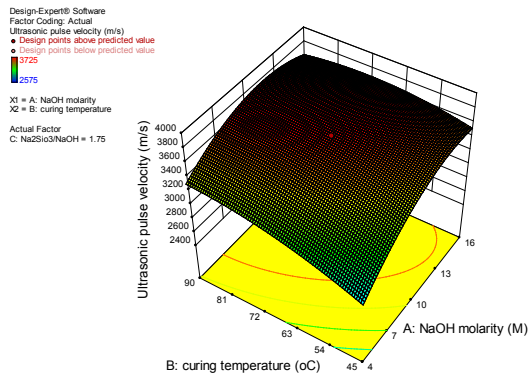
T= Transit time (s)

The measuring of pulse velocity was conducted with direct transmission method on 100 mm x 200 mm geopolymer cylinders. Standard testing procedure adopted as per recommendations of ASTM C 597-02^[23].

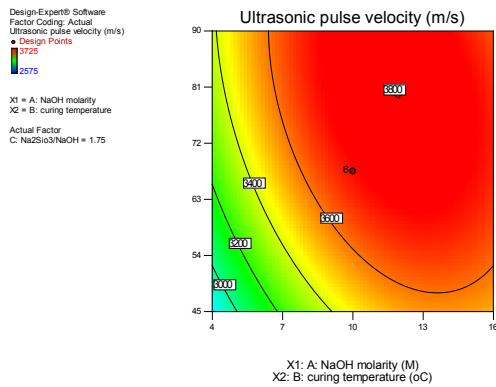
2.3 Results and Discussions

Effect of NaOH molarity, Na₂SiO₃/NaOH and Curing temperature on the ultrasonic pulse velocity is shown in Figure 3 and Figure 4 as response surface diagrams. And intervals of UPV are illustrated with the help of contour diagrams. As shown in Figure 3, influence of curing tem-

perature on UPV was altered with the NaOH molarity. At low molar NaOH, the effect of curing temperature was more prominent and increase in curing temperature caused the improving UPV value. However curing temperature shows relatively less influence on the geopolymer synthesis with higher molar NaOH solution. For 16M NaOH solution the increase in UPV was seen with the initial increment in curing temperature however further increase in curing temperature shows negative effect on UPV. This could have happened due to weak product formation. The same phenomenon was also reported by J.G.S. van Jaarsveld [24].



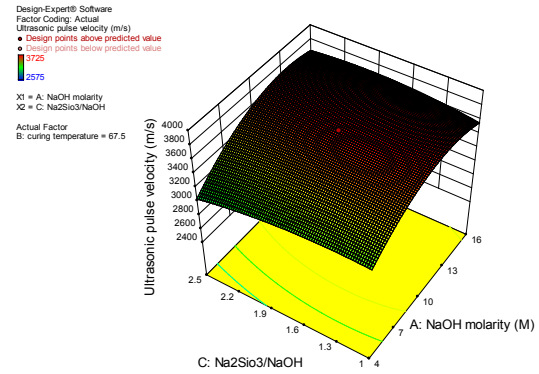
(a) 3D response surface



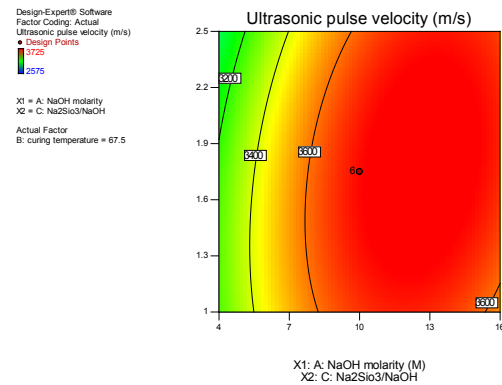
(b) contour diagrams

Figure 3. Ultrasonic pulse velocity versus Curing temperature and NaOH molarity

Influence of NaOH molarity on the ultrasonic pulse velocity is shown in Figure 4. Geopolymer synthesis with low concentration of NaOH produces weaker product and leaves unreacted fly ash particle, furthermore, concentrated NaOH produced higher geopolymeric reaction [25]. Improvement in UPV was observed with the increase in NaOH molarity from 4M to 10M. However further increase in NaOH concentration does not contribute in UPV. this could be happened due to fully reacted fly ash particles at 10M.



(a) 3D response surface



(b) contour diagrams

Figure 4. Ultrasonic pulse velocity versus NaOH molarity and Na₂SiO₃/NaOH

Nasir et al, utilized response surface methodology (RSM) for the development of ANOVA models for Nano silica modified concrete [26]. However, ANOVA equation 2 is given for the prediction of Ultrasonic pulse velocity (UPV) of geopolymer.

$$\text{Ultrasonic pulse velocity} = +705.65796 + 211.97143 * x_1 + 41.19801 * x_2 + 192.98417 * x_3 - 0.78889 * x_1 * x_2 + 15.33333 * x_1 * x_3 + 1.31852 * x_2 * x_3 - 7.37575 * x_1^2 - 0.21232 * x_2^2 - 134.20792 * x_3^2 \quad (2)$$

Where x_1 , x_2 , and x_3 represents NaOH molarity, curing temperature and Na₂SiO₃/NaOH respectively

2.4 ANOVA Model validation

The developed ANOVA model was statistically validated for the reliable usage to predict UPV value. ANOVA results for the model are given in the table 4, and the proposed model was significant.

As enlisted in table 5, Predicted R² is in good agreement with Adjusted R² and the difference between them was less than 0.2. Additionally, the value of adequate precision was more than four. Hence, the model can be used to predict UPV value [27].

Table 4. ANOVA results for full regression model

| | |
|------------------|---------------------------------|
| Response | Ultrasonic pulse velocity (m/s) |
| Sum of squares | 2.325 x 106 |
| Mean square | 2.583 x 105 |
| F-value | 1397.02 |
| p-value prob > F | < 0.0001 |
| Remarks | significant |

Table 5. Model Validation results

| | |
|--------------------|---------------------------------|
| Response | Ultrasonic pulse velocity (m/s) |
| Standard deviation | 13.60 |
| Mean | 3413.30 |
| R2 | 0.9992 |
| Predicted R2 | 0.9942 |
| Adjusted R2 | 0.9985 |
| Adequate precision | 118.024 |

Figure 5 and Figure 6 represents Perturbation curves and Normal plot of residuals for ultrasonic pulse velocity. Despite the higher sensitivity of NaOH molarity (A) and curing temperature (B), the developed model was less sensitive for Na₂SiO₃/NaOH (C). Furthermore, the spread of Normal plot of residuals was approximately follow the 45-degree straight line. Hence model can be used for any predicted value.

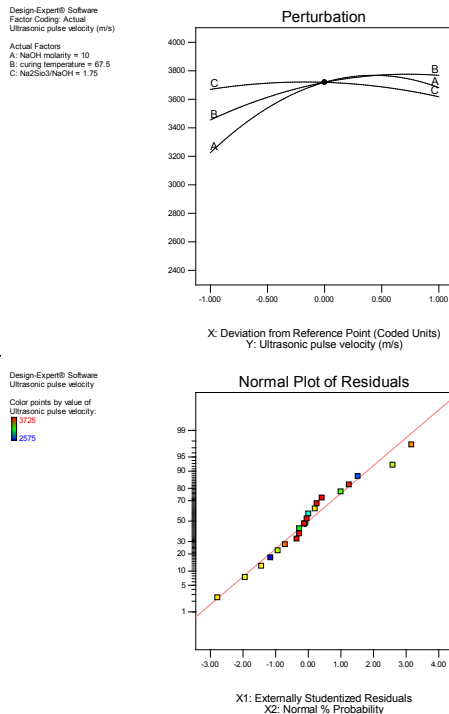


Figure 5. Perturbation curves for UPV Figure 6 Normal plot of residuals for UPV

2.4 Optimizations

In this study Multi-objective optimization approach was utilized to identify the ideal value of independent variable [28]. The main objective was to get maximize UPV for all factors simultaneously. The criteria for the prediction of the optimal solution through RSM is given in table 6. The graphical representation of the optimized solution in figure 6 shows the design variables of NaOH molarity of 9.20, Na₂SiO₃/NaOH of 1.456 and Curing temperature of 79.76 °C are able to achieve maximize value of UPV as 3743.12 m/s with desirability equals to one. A set of experiments was performed to validate the predicted optimized value of UPV. Test results in table 7 are found in great concurrence with the predicted outcomes with error less than 5%, therefore the developed model for UPV prediction can be used with more than 95% confidence level.

Table 6. Optimisation benchmark

| Factors and Response | Goals | Lower limit | Upper limit |
|--|----------|-------------|-------------|
| NaOH | In-range | 4 | 16 |
| Na ₂ SiO ₃ /NaOH | | 1 | 2.5 |
| Curing Temperate | | 45 | 90 |
| UPV | Maximize | 2575 | 3725 |

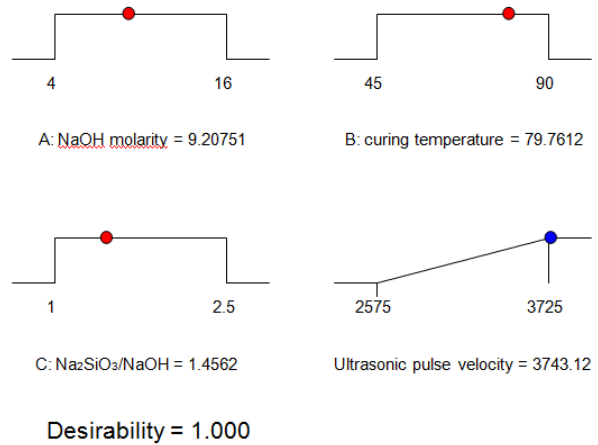


Figure 6. Optimisation ramps

Table 7. Model verification

| Response | Predicted | Experimental | Error (%) |
|----------|-----------|--------------|-----------|
| UPV | 3743.12 | 3920 | 4.72 |

3. Conclusion

In this investigation, RSM method was utilized to set up the optimal proportions of geopolymer paste for achieving maximize UPV. Conclusions drawn from this examination

study are given beneath.

(1) Statistical model for the prediction of UPV of geopolymer binder have been developed and validated.

(2) The developed statistical models can be used to design the experiment for any target value of UPV.

(3) The RSM optimization technique reduces the design time and improves the performance of the existing process and product, improves reliability and achieves robustness of the product and process.

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