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Using the Application of the Taguchi Method to Investigate the Main Factors Affecting the Optimum Compressive Strength of Geopolymer Mortars Containing Calcined-Kaolin

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*Abduallah Muftah Menshaz — amenshaz81@gmail.com

Husen Abduallah Alhawat — haalhawat@elmergib.edu.ly

Civil Engineering, Department, Faculty of Engineering, Elmergib University, Libya

Abstract:

This study aimed to apply the Taguchi method to investigate the main factors affecting the compressive strength of the calcination kaolin-based alkali-activated mortars. The efficiency of the calcination temperatures of kaolin at 650 °C (Metakaolin, MK650) (as the source material) in producing alkali-activated mortar products was enhanced via four factors which were optimized using the Taguchi method L09. Four primary factors were examined following a statistical study of the initial silica modules (sodium silicate; Na_2SiO_3) (Ms) (designed as **A**); the NaOH concentration (moles) "**B**"; the Na_2SiO_3 to NaOH weight ratio "**C**"; and the alkaline-activator to solid material weight ratio "**D**". Each factor was examined at the three levels in order to obtain the optimum mixture. Nine mixtures were prepared per theL09 array proposed by the method. The performance of the specimens was evaluated by compressive strength (CS) tests. The results show that the optimum mixture consisted of A2B2C3D2 which achieved a CS of 62.86 MPa after 28 days of curing. The results show that the main binding phases consist of aluminosilicate type gel (N–A–S–H) and calcium silicate hydrate (C–S–H) gels, formed simultaneously, withinthe Metakaolin-based alkali-activated mortar.

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Keywords: Alkaline activated mortars, Calcination Kaolin, Compressive strength, Taguchi method.

دراسة العوامل الرئيسية المؤثرة على مقاومة الضغط لملاط الجيوبوليمر القائم على الكاولين المكلس باستخدام تطبيق طريقة تاغوشي

عبدالله مفتاح المنشاز * و حسين عبدالله الحوات

قسم الهندسة المدنية –كلية الهندسة – الخمس،جامعة المرقب– ليبيا

الملخص

هدفت هذه الدراسة إلى تطبيق طريقة تاغوشي لبحث العوامل الرئيسية التي تؤثر على مقاومة الضغط لملاط الجيوبوليمر القائم على الكاولين المكلس. تم تعزيز كفاءة درجات حرارة تكليس الكاولين عند 650 درجة مئوية (Metakaolin,MK650) (كمادة مصدر) في إنتاج منتجات الملاط المنشط القلوي من خلال أربعة عوامل باستخدام طريقة Taguchi L09.تم فحص أربعة عوامل أساسية بعد دراسة إحصائية لوحدات السيليكا الأولية(سيليكات الصوديوم) Na_2SiO_3يرمز له بالرمز "A"،تركيز NaOH المولات بالرمز "B" ، ونسبة الوزنNa_2SiO_3 إلى NaOH بالرمز "C"؛ والمنشط القلوي إلى نسبة وزن مادة المصدر بالرمز "D". تم فحص كل عامل على المستويات الثلاثة للحصول على الخليط الأمثل. تم تحضير تسعة مخاليط وفقاً لمجموعة L09 الذي اقترحته هذه الطريقة. تم تقييم أداء العينات عن طريق اختبارات قوة الضغط (CS). أظهرت النتائج أن الخليط الأمثل يتكون من A2B2C3D2 الذي حقق CS قدره 62.86 ميجا باسكال بعد 28 يومًا من المعالجة. أظهرت النتائج أن مراحل الارتباط الرئيسية تتكون من هلام من نوع ألومينوسيليكات (N–A–S–H) وهيدرات سيليكات الكالسيوم–C–S) (H، يتم تشكيلها في وقت واحد، داخل الملاط المنشط القلوي القائم على الميتاكاولين.

الكلمات المفتاحية: قذائف هاون قلوية منشطة، الكاولين المكلس، مقاومة الضغط، طريقة تاغوشي.

1. Introduction

Aluminosilicate materials are generally mined from the earth such as for producing calcined kaolin (metakaolin, MK) [1, 2]. MK is an inorganic material; it is similar to organic materials because it reacts with solid polymers to form a strong alumina-silicate network by polycondensation. The usage of MK in geopolymer is good due to the high amount of amorphous phase and small particles [3, 4]. The formation of a geopolymer binder resulting from the reaction between the precursor materials and the alkali-activating solution involves a series of chemical reactions such as dissolution, transportation, and polymerization or polycondensation which release heat during the reaction. All these chemical reactions ultimately give rise to a solidified material with considerably high compressive strength (CS), equivalent to or higher than that of Portland cement-based materials [5]. MK-based geopolymers which are synthesized from high Si/Al ratios will remain stable even with prolonged aging [6]. Most natural precursors consist of materials such as MK, an anhydrous aluminosilicate obtained from the calcination of kaolin clay. Because of its disordered structure, it possesses enormous potential with regard to reactivity as a precursor material when it is mixed with an alkaline activating solution [7].

Many previous research have revolved around calcination conditions [8-12] such as the effects of calcination temperature which relates to alterations in the MK's chemical composition. Researchers have studied the influence of impurities in kaolin on the properties of alkaline-activated mortars (AAMs). Exposure temperature for a certain period may also affect the final properties of MK-based AAMs, which are related to chemical reactivity. The highest mechanical strength of MK-based AAMs, developed in these research works, was between 33-55 MPa.

The alkaline activator ratio ($Na_2SiO_3/NaOH$) is one of the factors that control the mechanical and chemical properties of kaolin-based geopolymer and alkaline activated binder (AAB). Furthermore, it has a direct relationship with silica modulus (Ms= SiO_2/Na_2O). [13] reported the impact of silica modulus (Ms= 1.6–2.4) on the

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polymerization of the MK-slag-based AAB, whereas [14] reported that the optimum (Na_2SiO_3/NaOH) in a lignite bottom ash based geopolymer was found within the range of 0.67 to 1.5. [15] have proven experimentally that the CS as well as the apparent density and the content of the amorphous phase of MK-based geopolymers, increase with an increase of NaOH concentration (within the range 4–12 mole/L). The CS increased with an increase in NaOH concentration mainly through the leaching of silica and alumina with high concentrations of NaOH [16]. The strength development in the alkaline activated binder was reported to also depend on the percentage Na_2O (Na_2O%), which had a direct relation to NaOH concentration (NC).

This study is an investigation into the characterization of geopolymers produced using calcined kaolin as source material. Four design factors were examined at three levels by the Taguchi method to obtain the optimal combination. The four factors are Na2SiO3 wt.%, NaOH concentration (mole), Na2SiO3/NaOH (weight ratio), and activated alkali/solids (weight ratio). A total of nine experiments were performed according to the L09 matrix proposed by Taguchi's method.

2. Materials and methods

2.1Materials and mixtures preparation

In the current studycalcination kaolin (MK) used in this work was produced in the laboratory, utilizing kaolin obtained from Associated Kaolin Industries, Malaysia, it conformed to the specification of BS 3892: part 1:1993. The MK was obtained by calcination of kaolin at temperatures of 650°C for 1 hour (10°C/min) to produce MK650.Then, characterization of the physical and chemical properties of the MK was undertaken.The chemical and physical properties of the MK in this study are provided in Table 1 and Table 2. A laser diffraction particle size analyzer (Mastersizer/E) was used to determine the particle size and specific surface area of the calcined kaolin or Metakaolin. The collected silica sand was sieved passed through a 1.18 mm and was retained on a 150 µm sieve as described in ASTM C778 [17]. River sand was estimated to have a fineness modulus of 2.8 and a specific gravity of 2.65. The alkaline activator (AA) used was a combination of silica modulus (Na_2SiO_3) (Ms= SiO_2/Na_2O ratio of 2, 2.2, and 3.3 respectively, and sodium hydroxide (NaOH) in a pellet form with 98% purity. NaOH and Na_2SiO_3 were purchased from Qrec (Asia) Sdn Bhd and Centre West Chemicals Sdn Bhd, respectively.

Table 1. Chemical compositions of the metakaolin using XRF analysis (% by weight)

Materials	Oxides									
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	TiO_2	K_2O	P_2O_5	Na_2O	LOI
MK	55.985	37.44	1.430	0.02	0.40	1.26	2.93	0.087	-	1.390

Table 2. Physical properties of constituent materials

Properties	Materials
	MK
Specific gravity	2.6
Median particle size d_{50} (μm)	3.95
Specific surface area (m^2/g)	1.008

2.2 Exposing Samples to High Temperature

In this study, the Taguchi method was used to design the mixtures and to obtain an optimum mix design by looking at the influence of different parameters on their mechanical properties. Four primary factors were examined following a statistical study of the initial silica modules (Na_2SiO_3) (Ms) (designed as **A**); the NaOH concentration (moles) “**B”**; the Na_2SiO_3 to NaOH weight ratio “**C”**; and the alkaline-activator to solid material weight ratio “**D”**. Each factor was examined at the three levels as depicted in Table 3. The level of each considered factor and values of the tested factors were chosen based on previous research [18-21].The amount of water added to each mixture was calculated based on a percentage of

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geopolymer paste at 5% by weight ratio [22]. The added water was prepared as 8 M, 10 M, and 12 M of NaOH concentration. NaOH is a basic ingredient for the geopolymerization process and the solution contains Na ion and water molecules [14]. All of the AAMs were made with sand-to-solid material weight ratios of 1.5 which are used in previous research [23]. The design suggested by the Taguchi method for four factors and three levels is the L09 array as shown in Table 4, while Table 5 shows the values and trial mixture proportions used in the series of nine mixtures. All cubic specimens with the dimension of 50 x 50 x 50 mm were cast with a prepared mix in double layers wherein every layer was shacked in a vibration table for 15 seconds. Then, all moulds were kept at room temperature (25± 2°C/70 ± 5% relative humidity) for 12 hours.

Following that, the samples were de-moulded and wrapped with heat-resistant vinyl bags to prevent losing moisture, and immediately placed in an oven at 75 °C for 24 hours [24]. Finally, the samples were left in ambient conditions (25± 2°C and 70±5% RH) till the day of testing (i.e., 3, 7, 14 and 28 days).

Table 3. Introduced levels for each factor in Taguchi experimental design of MK

Factor	Unit	Level 1	Level 2	Level 3
A: $Na_2Si\,O_3$	Ms	2	2.2	3.3
B: NaOH concentration	Mole	8	10	12
C: Na_2SiO_3-to-NaOH	wt. ratio	1.5	2	2.5
D: Alkaline-Activator-to- Solid Material	wt. ratio	0.78	0.80	0.82

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TABLE 4. Taguchi method of orthogonal arrays [L09 (3×4)] of the experimental design

Trial Mix	Factor A	Factor B	Factor C	Factor D
TMM1	3	2	1	3
TMM2	2	3	1	2
TMM3	2	1	2	3
TMM4	3	1	3	2
TMM5	3	3	2	1
TMM6	1	3	3	3
TMM7	1	2	2	2
TMM8	2	2	3	1
TMM9	1	1	1	1

TABLE 5. Mix proportions of alkali-activated mortar used for Taguchi optimization (kg/m^3)

Trial Mix	MK	Sand	Na_2SiO_3	NaOH		Added water
				pellet	water	
TMM1	712	1070	350	70	160	60
TMM2	715	1070	347	80	150	60
TMM3	712	1070	389	50	140	60
TMM4	715	1070	414	40	120	60
TMM5	718	1080	383	70	120	60
TMM6	712	1070	417	60	110	60
TMM7	718	1080	383	60	130	60
TMM8	718	1080	411	50	110	60
TMM9	718	1080	345	60	170	60

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3. Results and discussions

The CS test was performed on the 9 trial mixtures which were prepared by the Taguchi method. The estimation of the compressive strength (response index) for each factor was calculated by averaging the CS at 3, 7, 14 and 28 days for each trial mixture containing a particular factor as shown in Fig. 1. For example, the response index for factor A1 which was tested on trial mixtures marked with factor A1, as shown in Fig. 1 (i.e. TMM6, TMM7, and TMM9). Thus, the response index for factor A1 at 3 days after curing was the average of the response index for trial mixtures of TMM6, TMM7, and TMM9. Similarly, the calculations were done for factors A1, A2, and A3 at 3, 7, 14 and 28 days, respectively. Results have been plotted in Fig. 2a. The plot indicates that factors A2 gave the highest response index; therefore, this is the optimum value for factor A. Optimum values for factors B (Fig. 2b), C (Fig. 2c) and D (Fig. 2d) were calculated in the same way.

The response index for Factor A is shown in figure 2a. The results revealed that the strength of response index increased with the increase of Factor A1 up to A2 and reduce to Factor A3. For all mixtures the strength increased with the curing time. The 28 days' strength in A1 was found to be 62.60 MPa, which was increased by 5.53% in A2 and decreased by 15.80% in A3, when Na_2SiO_3 (Ms). Therewith, when NaOH concentration, the results revealed that the response index increased with the increase of Factor B1 to B2 by 4.53%. Then, the response decreased with the increase of Factor B2 to B3 by 2.42% which clarify in figure 2b. While that, sodium-based aluminosilicate is formed, attracting OH– to its structure, lowering the total amount of hydroxide groups and allowing the formation of N–A–S–H gel as a secondary reaction product [25, 26].

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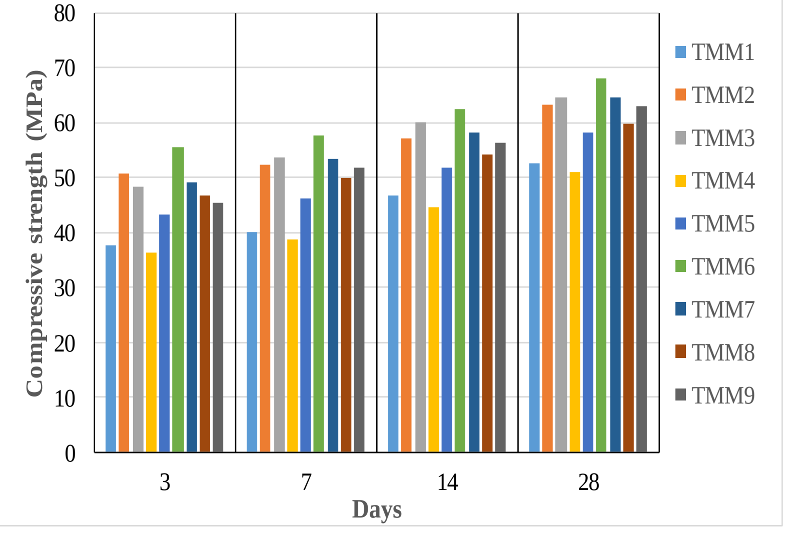

Figure 1. Changes of compressive strength of the trial mixes.

Nonetheless, when study effect of Na_2SiO_3 to NaOH weight ratio, the response index for Factor C is shown in figure 2c. The response index slightly increased with the increase in Factor C up to C3, where the amount of aluminosilicate gel form is correlated with the compressive strength of geopolymer, in which a higher formation of gel will lead to higher geopolymer strength [20]. The 28 days' strength in C1 was found to be 59.61 MPa, which was increased by 0.10% and 4.99% in C2 and C3, respectively. However, alkaline activator to solid weight ratio, the response index for Factor D is shown in figure 2d. The response index increased with the increase in Factor D1 up to D2 and reduce to Factor D3. It was evident that the highest CS was noted at 28 days for D2 followed by D1 and D3, accordingly. The 28-day strength in D1 was found to be 60.35 MPa, which was increased by 2.47% and decreased by 1.09% in D2 and D3 respectively.

Finally, the statistical analysis of the Taguchi method can determine the ideal combination while simultaneously meeting the highest response index of metakaolin-based AAMs mixtures. The CS obtained from the various trial mixtures (Figure 1) ranged from 36.34 to 68.06 MPa at 3, 7, 14, and 28 days. The optimum levels achieved for the effective performance of each response are shown in figure 3. However, the results can be verified by fabricating a new mixture and producing specimens according to the optimum levels of all factors and testing at 3, 7, 14, and 28 days. The range of the CS obtained for all mixtures was from 48.46 to 62.86 MPa. Consequently, it can be concluded that the Taguchi method can be effectively applied to optimize the CS of the AAMs.

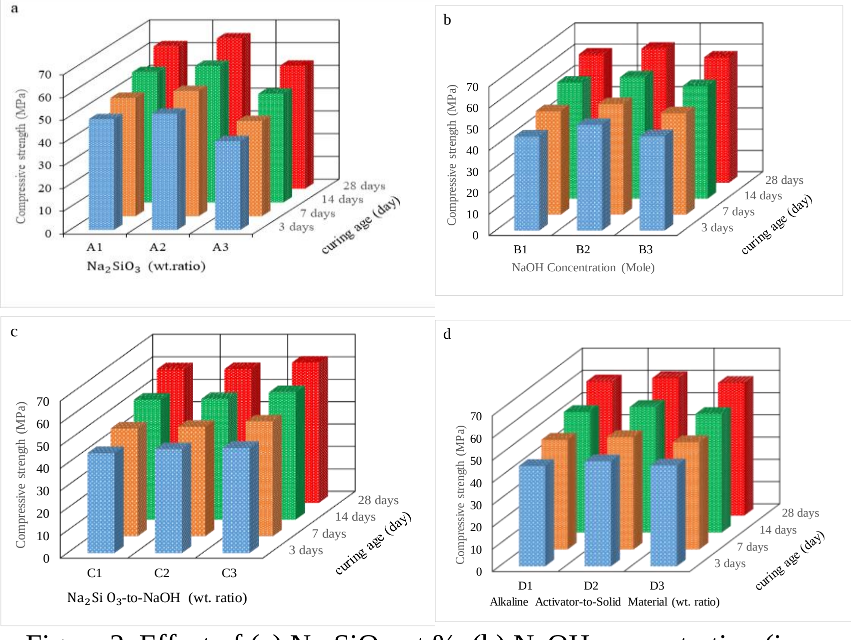

Figure 2. Effect of (a) Na_2SiO_3 wt.%, (b) NaOH concentration (in terms of molar), (c) Na_2SiO_3-to-NaOH weight ratio, (d) alkaline activator-to-solid material weight ratio on each response of compressive strength at different curing ages.

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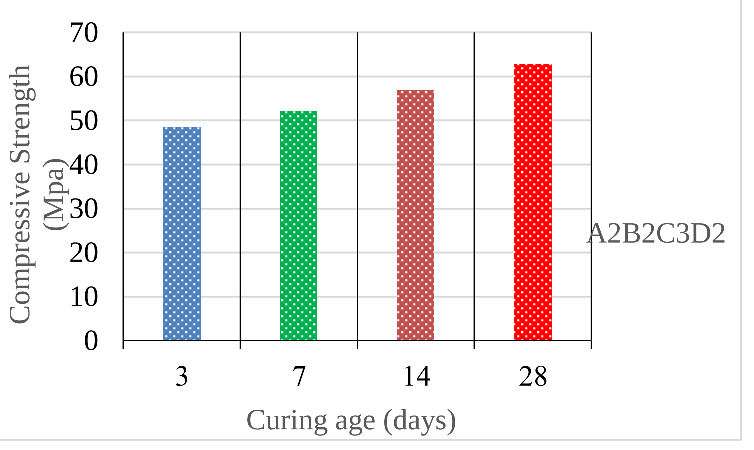

FIGURE 3. Optimization of the factors combination of MK based AAMs at different curing ages

4. Conclusions

This research used the application of the Taguchi method to investigate the main factors affecting the optimum compressive strength of geopolymer mortars containing calcined kaolin. TheTaguchi method L09 indicated that the optimum of metakaolin can be achieved with an alkaline activator (2.2 of silica modules (Na_2SiO_3), 10 M of NaOH, Na_2SiO_3 to NaOH = 2.5, and alkaline-activator to solid material = 0.52). The optimum of compressive strength the metakaolin-based alkali-activated mortars is to synthesize the optimum geopolymer mortar which produces 62.86 MPa after 28 days of curing. The high compressive strength is due to the formation of gel binders (C–S–H and N–A–S–H) in the geopolymer.

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