

## Variation of Packing Densities and Liquid Film Thicknesses in Alkali Solution and Water in Natural Pozzolan Calcium Hydroxide Pastes and Mortars

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### ABSTRACT

The characteristic properties of concrete depend on the quality of mortar. In order to improve the properties of hardened concrete, the mortar needs to have good performance. The property of mortar is affected by wet packing density, liquid film thickness, type and quality of binder, gradation of fine aggregates, and binder-to-fine-aggregate ratio. The commonly used alkali solutions to produce polymer mortar and concrete are cursed by carbon dioxide emissions. This study used alkali solution and water to investigate the variation of packing densities and film thicknesses on paste and mortar mixes. of natural pozzolan and calcium hydroxide at a ratio of 75P25CH and standard sand. The standard proctor compaction tools and procedures were adopted to attain the maximum values of packing density of 0.937 for mortar mixed with alkali solution, 0.919 for mortar mixed with water, 0.906 for paste mixed with alkali solution, and 0.829 for paste mixed with water. For the case of film thicknesses, the optimum values obtained were 0.548  $\mu\text{m}$  for mortar mixed with alkali solution, 0.248  $\mu\text{m}$  for mortar mixed with water, 0.398  $\mu\text{m}$  for paste mixed with alkali solution, and 0.09  $\mu\text{m}$  mixed with water. The results indicate that alkali solution produced higher packing density and liquid film thickness than water for both paste and mortar, and the packing density and liquid film thickness increase with the increase of the number of compacting blows with 9 blows to 36 blows. The study recommends the establishment of standard compaction effort and procedure.

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## 1.0 Introduction

In the construction industry, studies on the quality and innovation of new building materials are progressively increasing. In the past few decades, researchers have focused on finding new, eco-friendly, and long-lasting binders like geopolymers to replace traditional cement. These binders improve the performance of mortar and concrete mixtures while saving money and protecting the environment (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Huseien et al., 2018; Nishiyama & Yokoyama, 2013). Several studies have indicated promising results of using pozzolanic material to produce polymer concrete and mortars for construction projects (Satya Pavan, 2018). It has been observed that polymer mortar and concrete have good mechanical strength in the hardened state, good durability in service, and a reduced environmental footprint (Malisa & Chengula, 2022). The disadvantage of polymer is that it requires the use of industrially produced chemicals, such as sodium and potassium hydroxide, which may be difficult to obtain and sometimes expensive.

Previous studies show that the performance of polymer concrete depends on the quality of the mortar produced (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Provis, 2018; Singh et al., 2015). Hence, the production of high performance mortar should be the first step in the mix design of polymer concrete (Huseien et al., 2018; Raj et al., 2014). To produce high-quality mortar, some factors need to be considered (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Kotak & Thaker, 2016; Kwan et al., 2012). The most important factors that contribute to the good performance of mortar are the rheology and packing of the particles, as well as the shape and size of the solid materials in fresh mortar (Alonso et al., 2017; Han et al., 2019). There are things about the materials that affect these factors, such as their liquid content (LC), packing density (PD), specific surface area (SSA) of solids, and liquid film thickness (LFT) (Huseien et al., 2018; Kwan & Fung, 2009; Li & Kwan, 2014).

Packing density is the number of solid particles arranged and packed together in a certain volume. Liquid film thickness is the amount of liquid that surrounds these particles while they are being mixed (Kwan & Fung, 2009; Li & Kwan, 2014; Zhao et al., 2022). The relationship between PD and LFT in paste and mortar involves the arrangement of particles in the mixture and how that arrangement influences the amount of liquid needed to coat the particles (Kwan & Fung, 2009; Kwan & Li, 2012; Zhao et al., 2022).

The concept of packing density and liquid film thickness is that there exists a certain volume of voids between the particles in the mixes (Kwan et al., 2012; Zhao et al., 2022). The amount of liquid added must be at least sufficient to fill up the voids. The extra solution needed to fill up the voids is called "excess liquid" (Kwan & Li, 2012). The excess liquid exists in the form of liquid films coating the surfaces of the particles in the mixes. The larger the volume of excess liquid, the thicker the liquid films will be, and the particles will be further apart, resulting in reduced friction resistance and interlocking between them. This ultimately leads to increased workability and negatively impacts the strength and durability of hardened mortar and concrete (Robayo-Salazar et al., 2018; Zhao et al., 2022).

The previous studies have revealed that the combined effects of liquid content (LC), packing density (PD), and specific surface area (SSA) may be evaluated in terms of liquid film thickness (LFT) of the solid-liquid mixture (Alonso et al., 2017; Chengula & Middendorf, Bernhard, Msambichaka, 2018; Kotak & Thaker, 2016). Liquid film thickness has a significant impact on the flowability and workability of the mortar (Han et al., 2017, 2019; Kotak & Thaker, 2016; Kwan & Fung, 2009). A thicker liquid film tends to increase the flowability and workability of mortar and concrete. This is because liquid film provides lubrication between particles, allowing relative movement as the internal friction is reduced (Kotak & Thaker, 2016; Li & Kwan, 2011; Zhang et al., 2019). However, optimum liquid film is required to produce good mortar and concrete performance. Generally, proper packing density with optimal liquid film thickness can

contribute to the improved strength and durability of the mortar and concrete (Kwan & Fung, 2009; Zhao et al., 2022).

The characteristics of fine aggregates, such as shape, size, strength, texture, and amount of solid materials, have effects on the rheology and packing of the particles in mortar and concrete mixes (DENER, 2022; Pereira et al., 2015; Wazien et al., 2016). Other researchers have found that mortar flows better when the average size of the particles in the mixture gets smaller (Alonso et al., 2017; Chengula & Middendorf, Bernhard, Msambichaka, 2018; Hamisi Chengula, 2022; Han et al., 2017; Li & Kwan, 2014). However, when the mean size of the fine aggregates gets smaller, up to 0.3 mm, their solution absorption is critical to the flowability behaviour (Chengula & Middendorf, Bernhard, Msambichaka, 2018; DENER, 2022; Han et al., 2017). Moreover, researchers found that the liquid demand of a polymer mortar is closely related to the packing density of the fine aggregate (Gong et al., 2014; Kashani & Ngo, 2018; Sanjeev Mukherjee, Vardhman Jain, Akshay Gupta, 2020). Mortar made of finer aggregate particles would need a higher liquid content for a given workability, and this is because of increased specific surface areas of the solids (Chen et al., 2021; Chengula & Middendorf, Bernhard, Msambichaka, 2018; Kotak & Thaker, 2016; Wu et al., 2022).

On another hand, the chemical composition of precursor materials has a direct relation to the properties of produced mortar. The type and amount of chemical composition affect the hydration mechanisms (Yanguatin et al., 2017). Therefore, a sufficient quantity of chemical oxides is required for the performance of polymer mortar and concrete.

This study examined the variations in packing densities and liquid film thicknesses in mixtures of pozzolan-calcium hydroxide paste and mortar mixes, taking into account the impact of compaction efforts and the types of liquids, specifically alkali solutions and water.

## **2.0 Materials and Methods**

### *2.1 Materials*

The materials used in this study are fine aggregates, sodium hydroxide pellets, sodium silicate solution, natural pozzolan, calcium hydroxide, and tap water. The fine aggregates were obtained from Mbarali, natural pozzolan from the Songwe area in the Mbeya Region. Calcium hydroxide, sodium hydroxide pellets, and sodium silicate solutions were obtained from authorised local chemical suppliers in Mbeya and Dar es Salaam, Tanzania.

The natural pozzolan-calcium hydroxide binders used for this study were mixed at a proportion of 75% natural pozzolan and 25% calcium hydroxide (75P25CH). Several studies have shown that pastes and mortars made from natural pozzolan-calcium hydroxide binders have good compressive strengths at this level of pozzolan-calcium hydroxide (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Malisa & Chengula, 2022). The sodium hydroxide pellets were diluted with distilled water to make a solution of 8 moles. It was 2.5 times as much sodium silicate ( $\text{Na}_2\text{SiO}_3$ ) as sodium hydroxide (NaOH) that was mixed together to make polymer paste and mortars. Other studies have indicated that 8 moles of sodium hydroxide solution and a ratio of 2.5:1 of  $\text{Na}_2\text{SiO}_3$  and NaOH are used to make alkali solutions for polymer pastes, mortars, and concrete that show good mechanical properties (Malisa & Chengula, 2022).

### *2.2 Methods*

The methods used in this study involve the preparation of standard sand by sieving and determinations of physical properties, which include specific surface areas, specific densities, and water absorption/adsorption for binder and sand. The ingredients were mixed together in order to produce pastes and mortar by using an electric pan mortar mixer. Using the standard Proctor compaction test as described by MoW (2000), fresh paste and mortar with different liquid-to-solid ratios were pressed together. The packing density and water film thickness for the pastes and mortars with different liquid-to-solid ratios and pressing efforts were then found.

### 2.2.1 Mixing and Compaction Procedures

The paste mixes' binder was made up of powdered natural pozzolan and calcium hydroxide mixtures that were 75% pozzolan and 25% calcium hydroxide (75P25CH). Pozzolanic binder (75P25CH) and standard sand were used to produce mortar mixes at a proportion of 1:3 as described in British standards, EN 196-1 (EN 196-1, 2005). The mixtures were then mixed with either alkaline solution or water at varying liquid-to-solid ratios. The alkali solution and water were used as two different liquids in this study because they are commonly used liquids for paste, mortar, and concrete mixes (Alonso et al., 2017; Gong et al., 2014; Kwan & Li, 2012).

Pastes and mortars were mixed by using a pan-type portable mixer (ELE International 240 V, 3 amps, 50 Hz, 1 phase). and compacted by using the standard Proctor compaction test, the procedure stipulated by MoW (2000) is shown in Figure 1.

Figure 1  
 Photos A Pan-Type Mortar Mixer for Mixing Paste and Mortar, B Proctor Tools Used for Compaction of Materials



### 2.2.2 Evaluation of Packing Density and Liquid Film Thickness

The mass of fresh paste and mortar in a known volume of mold with varying liquid-to-solid ratios was measured using an electric balance. The liquid solids ratios were varied, and for each ratio, the bulk densities and packing densities were calculated.

The maximum packing density was obtained at the minimum void content in a mold, resulting in improved strength and durability of hardened mortar and concrete specimens (Zhao et al., 2022). Liquid film thickness (LFT) is the average thickness of the liquid film wrapping the solid particles. The value of LFT is defined as the ratio of

excess liquid to the solid surface area. Normally, LFT is related to packing density (PD), liquid consumption, and total surface area of the solid particles (Wu et al., 2022). Equations 1, 2, and 3 are used to determine bulk density, packing density, and liquid film thickness of fresh mixtures (Kotak & Thaker, 2016; Raj et al., 2014).

$$Pb = \frac{M_m}{V_c} \quad (1)$$

$$PD = \frac{\rho_b}{\sum R_i \rho_i} \quad (2)$$

$$LFT = \frac{v_l \sum M_i}{\rho_l \sum R_i SSA_i} \quad (3)$$

Where:  $M_m$  - Mass of wet mixtures,  $V_c$  - Volume of container,  $PD$  - Packing density of mortar;  $\rho_b$  - Bulk density of mortar (gm/cm<sup>3</sup>);  $R_i$  - Fractions of ingredient materials of mortar;  $\rho_i$  - Densities of ingredient materials of mortar (gm/cm<sup>3</sup>) LFT - Liquid film thickness (cm);  $SSA_i$  - specific surface areas of solids (cm<sup>2</sup>/gm);  $M_i$  - Masses of solids;  $\rho_l$  - Density of liquid (gm/cm<sup>3</sup>);  $v_l$  - excess liquid ratio,  $R_i$  - Fractions of ingredient materials of mortar.

Table 1 represent masses of fresh mortar and paste used to compute packing densities and liquid film thickness.

Table 1  
 Calculated Masses for Material Mixes

Mixes	Solution	Alkali Solution to Solids ratios	0.15	0.175	0.2	0.25	0.3
Mortar	Alkali	Mass of cylinder + wet mortar (g)	5758.9	5840.2	5821.4	5761.3	5705.1
		Mass of cylinder (g)	3586.9	3586.9	3586.9	3586.9	3586.9
		Mass of Alkali Mortar (g)	2172	2253.3	2234.5	2174.4	2118.2
Mortar	Water	Water to Solids ratios	0.1	0.12	0.14	0.16	0.18
		Mass of cylinder + wet mortar (g)	5749.8	5786.2	5756.1	5731.8	5702.1
		Mass of cylinder (g)	3586.9	3586.9	3586.9	3586.9	3586.9
		Mass of Water Mortar (g)	2162.9	2199.3	2169.2	2144.9	2115.2
Paste	Alkali	Alkali Solution to Solids ratios	0.3	0.35	0.4	0.45	0.5
		Mass of cylinder + wet mortar (g)	5246.9	5417.4	5560.3	5535.5	5488.1
		Mass of cylinder (g)	3586.9	3586.9	3586.9	3586.9	3586.9
		Mass of Alkali Paste (g)	1660	1830.5	1973.4	1948.6	1901.2
Paste	Water	Water to Solids ratios	0.2	0.25	0.3	0.35	0.4
		Mass of cylinder + wet mortar (g)	5190.2	5371.3	5331.7	5276.6	5208.1
		Mass of cylinder (g)	3586.9	3586.9	3586.9	3586.9	3586.9
		Mass of Water Paste (g)	1603.3	1784.4	1744.8	1689.7	1621.2

### 3.0 Results and Discussions

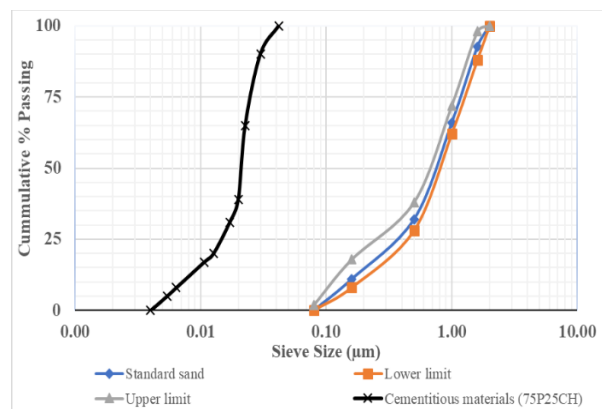
#### 3.1 Physical and Mechanical Properties of the Used Materials

In this study, sand from Mbarali was graded and blended in order to meet the gradation standard given in EN 196-1 (EN 196-1, 2005). Four different sizes of sand were categorised into four groups: group 1 is from 2.0 mm to 1.18 mm, group 2 is from 1.18 mm to 0.6 mm, group 3 is from 0.6 mm to 0.15 mm, and group 4 is from 0.15 mm to 0.075 mm and blended at proportions of 39%, 23%, 30%, and 8%, respectively, to obtain sand gradation that lay within the proposed envelopes (Fig. 2). Specific density and water absorption of sand were determined following procedures stipulated by MoW (2000). The specific surface area of sand was determined by using an empirical formula (equation 4) that uses the particle size distribution curve of the materials (Chengula & Middendorf, Bernhard, Msambichaka, 2018). Specific density and specific surface area of binder (75P25CH) were determined by using the Enslin-Neff traditional method used in geotechnical engineering. Specific density and water absorption of aggregates in mortar and concrete can influence the compressive strength and durability of the hardened mortar and concrete (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Han et al., 2019). Aggregates with higher specific gravity are generally denser and therefore contribute to higher

compressive strength when well-graded (Wu et al., 2022). However, higher water absorption results in increased porosity, which eventually reduces the compressive strength and durability of hardened mortar and concrete.

The specific surface area (SSA) of materials significantly affects the fresh and hardened properties of mortar and concrete. Previous research has indicated that SSA of materials affects flow properties, packing density, and compressive strength of hardened mortar and concrete (Chengula & Middendorf, Bernhard, Msambichaka, 2018; Wu et al., 2022). Figure 2 shows particle size distribution curves of sand and binder (75P25CH) materials used for paste and mortar mixes for this study.

Figure 2  
 Graph for Particle Size Distribution for Sand and Cementitious Materials (75P25CH)



S

$$SA = \frac{1}{\rho_s} \left[ 10 \left[ 4.550 \frac{(A^{0.0085} + B^{0.065})}{(A^{0.085} B^{0.065})} + 4.265 \frac{(2.75^{0.0035} C + 2.75^{0.00045} D)}{(2.75^{0.0035} C + 0.00045 D)} \right] \right]^{0.25} \quad (4)$$

$$A = S_{25}; B = \frac{1}{3} [S_{25} + 2S_{50}]; C = \frac{1}{5} [2S_{50} + 3S_{75}]; D = \frac{1}{7} [3S_{75} + 4S_{99.9}]$$

Si – Sieve size at %ge ‘i’ cumulative passing (µm);  $\rho_s$  – Specific density of materials (g/cm<sup>3</sup>) and SSA – Specific surface area (cm<sup>2</sup>/g).

The Enslin-Neff analysis measures the water uptake capacity under free swelling conditions until the material reached its maximum absorption. The water absorption was calculated by using equation 5 (Fehervari et al., 2021).

$$w_a = \frac{w_{max}}{m_{dry}} \times 100 \quad (5)$$

$W_a$  - is water absorption,  $W_{max}$  - is mass of water at maximum absorption,  $M_{dry}$  - is dry sample of powder materials

The results for physical properties of materials are presented in Table 2. The specific densities for binder (75P25CH) and sand materials were found to be 2.44 and 2.60, respectively. The values are related to studies carried out by Partschefeld et al. (2020) and Yanguatin et al. (2017). Water absorption for sand was found to be 0.53%. According to ASTM C 127-15, the water absorption requirement for standard sand shall not exceed 1% by mass; therefore, the sand meets the standard requirement in terms of absorption (ASTM, 2015). Water adsorption for binder materials (75P25CH) was determined to be 61%. It is important to determine the water adsorption properties of binders for construction, as it can affect the properties of fresh mortar and concrete. The combined density of sodium silicate and sodium hydroxide solution was determined to be 1.52 g/cm<sup>3</sup>.

Table 2

*Physical and Mechanical Properties of Materials*

Properties	Specific densities (g/cm <sup>3</sup> )	Specific surface areas (cm <sup>2</sup> /gm)	Water Absorption/ adsorption (%)
Alkali Solution	1.52	N/A	N/A
75P25CH binder	2.44	4905	61
Sand	2.60	88.3	0.53

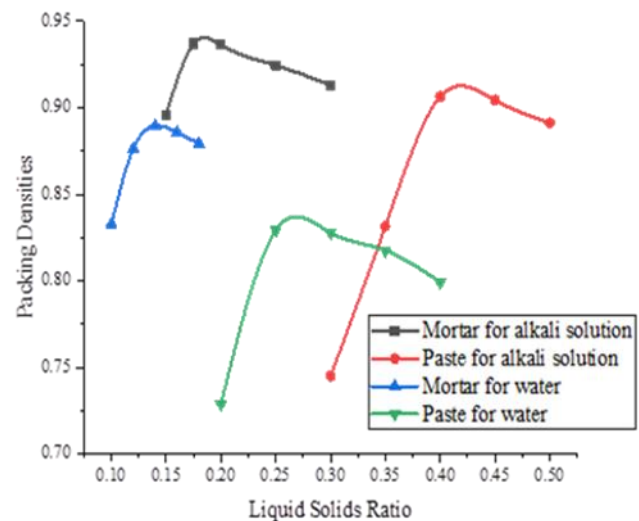
*3.2 Packing Density and Voids Ratios*

The graphs of packing density against liquid to solid ratios for mortar and paste were plotted. The optimum liquid is considered as the liquid-solid ratio needed to achieve the maximum packing density of

the mixture. The maximum packing densities were found to be 0.937 and 0.906 for mortar and paste mixed with alkali solution, respectively. The packing density for mortar and paste mixed with water was determined to be 0.919 and 0.829, respectively. Figure 3 indicates the relationship between the packing density and liquid-to-solid ratios for both paste and mortar. The maximum packing densities were found to be 0.937 and 0.906 for mortar and paste mixed with alkali solution, respectively. The packing density for mortar and paste mixed with water was determined to be 0.919 and 0.829, respectively.

Figure 3

*The Graph of Packing Density for Fresh Paste and Mortar Mixes*

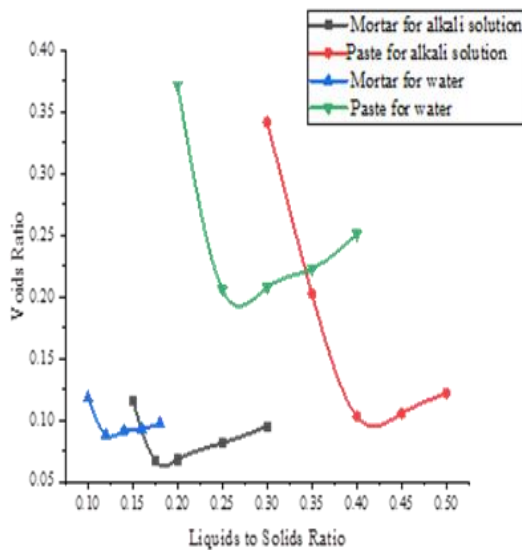


The alkali solution to solid ratios at maximum packing densities are 0.175 and 0.4 for mortar and paste respectively. The water-to-solid ratios at maximum packing densities are 0.12 and 0.25 for mortar and paste, respectively. The results indicate that more alkali solution is required to attain the maximum packing density for mortar and paste. This is attributed to the high viscosity of alkali solution compared to water. The optimum packing densities based on alkali solutions for mortar and paste are higher than those for water. These results stem from the higher density of alkali solution compared to water.

The graphs show that when liquid is added, the voids ratio decreases to a minimum value before beginning to increase for paste and mortar mixes. At the minimum void ratio, the liquid solids ratio represents the amount of liquid required to achieve the maximum packing density. Figure 4 shows the relationship between void ratios under different liquid-solid ratios.

Figure 4

Graphs of Voids Ratios Versus Liquid to Solid Ratios



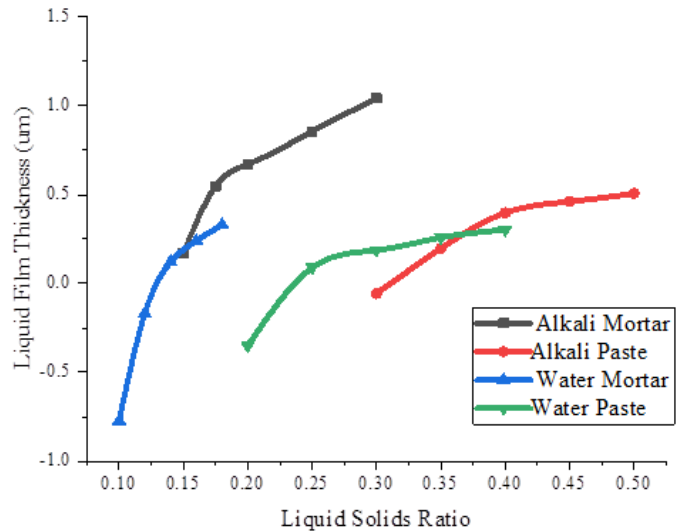
### 3.3 Liquid Film Thickness

Liquid film thickness of fresh paste and mortar mixed with alkali solution and water were determined. The optimum liquid film thickness is observed at the turning point of the graphs. The optimum alkali solution film thickness for mortar and paste were determined to be 0.548  $\mu\text{m}$  and 0.398  $\mu\text{m}$  respectively at maximum packing density. On the other hand, water film thickness for mortar and paste were 0.248  $\mu\text{m}$  and 0.090  $\mu\text{m}$  respectively. The results reveal that liquid film thickness for mortar is higher than that of paste. This trend complies with the results from other previous researchers when cement was used as binder (Li & Kwan, 2011). However, the values are greater than that obtained using Portland cement as binder. This likely attributed to the different

properties of pozzolanic binder materials and Portland cement particularly liquid adsorption properties and initial setting time. Figure 5 shows the relationship between liquid film thickness and liquid solids ratios.

Figure 5

The Relationship between Liquid Film Thickness and Liquid Solids Ratio for Paste and Mortar

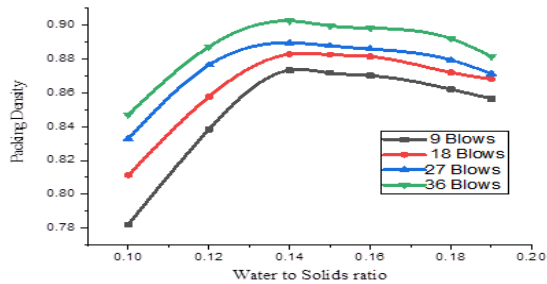


### 3.4 Packing Density and Water Film Thickness for Mortar at Varying Compaction Efforts

The packing densities and water film thickness for mortar were determined at varying compaction efforts for 9 blows, 18 blows, 27 blows, and 36 blows. The graphs indicate that the packing density increases with increasing compaction efforts at a given water-solids ratio. This is contributed to the reduction of voids with the increase of compaction efforts. Therefore, there is a need to introduce standard compaction efforts and procedures to be used in the determination of packing density and liquid film thickness for mortar and concrete mixes. Moreover, the shape, size, and strength of solids affect the pore system in the mixes, which then affects packing densities and water film thicknesses of fresh mixtures. Figure 6 shows the packing densities at varying compaction efforts.

Figure 6

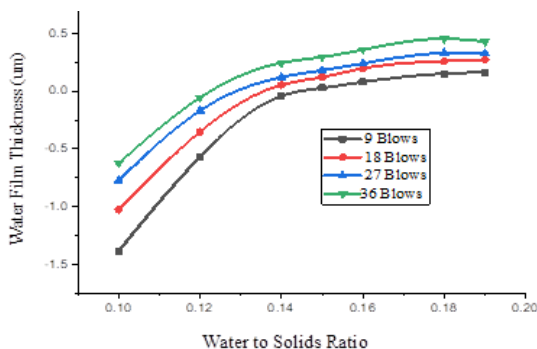
Graph of Packing Densities versus Water Solid Ratios for Various Compaction Efforts



The thickness of the water film increases uniformly as the compaction effort increases. The graphs for the water film thickness against the water solids ratio were plotted to indicate the relationship between water film thickness and compaction effort. The obtained water film thicknesses were 0.041  $\mu\text{m}$ , 0.055  $\mu\text{m}$ , 0.122  $\mu\text{m}$ , and 0.246  $\mu\text{m}$  for 9 blows, 18 blows, 27 blows, and 36 blows, respectively. The reason behind this trend is likely attributed to the reduction of voids between solid particles when compaction efforts increase, which in turn increases the volume of excess water in the mixes to wet the solid particles at the same water-to-solid ratio. The optimum water film thickness is observed at turning points of the graph. Figure 7 illustrates how the water film thickness changes with varying compaction efforts and water solids ratios.

Figure 7

Water Film Thickness for Fresh Mortar at Varying Compacting Efforts



#### 4.0 Conclusion and Recommendation

A design of good-performance mortar and concrete can be done by using packing density and water film thickness techniques. The lower liquid-to-solids ratio contributes to a good performance of mortar and concrete. The packing density and liquid film thickness for paste and mortar were mostly influenced by compacting effort and properties of the materials used in the mix. The packing densities obtained in this study at the compaction effort of 27 blows were 0.937 for mortar mixed with alkali solution, 0.919 for mortar mixed with water, 0.906 for paste mixed with alkali solution, and 0.829 for paste mixed with water. Liquid film thickness obtained was 0.548  $\mu\text{m}$  for mortar mixed with alkali solution, 0.248  $\mu\text{m}$  for mortar mixed with water, 0.398  $\mu\text{m}$  for paste mixed with alkali solution, and 0.090  $\mu\text{m}$  for paste mixed with water. The packing density and liquid film thickness for mortar and paste mixed with alkali solution were greater than that mixed with water. This is due to the different material properties present in the mixes. It has been revealed that packing densities and liquid film thickness increase with increasing compaction effort at a given liquid-to-solids ratio.

It is recommended that standard compaction efforts and procedures for determining the packing density and water film thickness of fresh paste, mortar, and concrete are necessary. This is due to the variation in packing density and liquid film density at different compaction efforts.

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## 7.0 Conflicts of Interest

The authors declare that they have no potential conflicts of interest with regard to the authorship and/or publication of this research work.

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