

# Particle Packing Theory in Ultra-sustainable Concrete with High SCM Content

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## Abstract

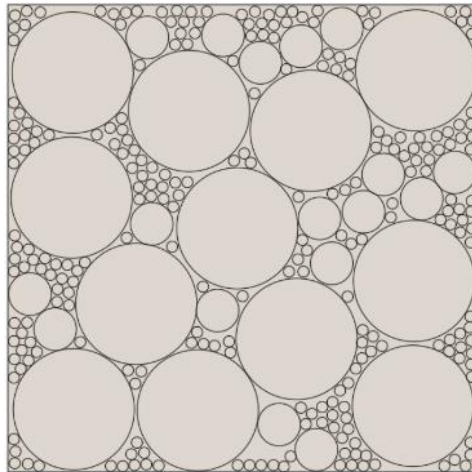
Concrete contains four main variables: aggregates, binder materials, water and chemical admixture, in which aggregates can account for up to 75% by mass of concrete. Based on a huge proportion of aggregates, it is worth a study with the aim of creating more sustainable concrete. Particle packing theory explains and predicts the arrangement of bulk materials and has been widely applied in related industries. In the paper, a number of established particle packing models are briefed, along with the experiments based on the application of particle packing theories. Concrete trials were designed to investigate the effect of total aggregate grading on the properties of fresh & hardened concrete. It found that concrete with very coarse grading compromises workability and enhances drying shrinkage. Additionally, an investigation on fine aggregate properties was carried out to evaluate the effect of grading. Manufactured sand and natural fine sand were blended at different portions to test flow time and voids content. Meanwhile, mortar mixes were further carried out, and results show that low flow time leads to better workability, and low voids content develops the relatively high compressive strength. The paper shows particle packing theory is a solid method to optimize the performance of concrete, and future studies are discussed.

**Keywords:** particle packing; aggregate grading; voids ratio; concrete mix.

## 1 Introduction

With the development of society, the construction industry is rapidly expanding. Cement, which is the most used construction material in the world, generates about 8 % of CO<sub>2</sub> emissions caused by human action annually [1]. To reduce CO<sub>2</sub> emissions, sustainable concrete has become a major focus of researchers. Sustainable concrete is defined to be environmentally friendly in different ways; reuse waste materials as part of components, or the production process could reduce the pollution of the environment, or the performance is outstanding and lifetime is long [2].

Particle packing theory explains and predicts the arrangement of bulk materials where fine particles could efficiently and effectively fill the voids among coarse particles, so that the whole combination becomes denser, shown in **Error! Reference source not found.** Because of this, it has been widely applied in related industries, such as ceramic, asphalt, powder technology and concrete [3]. In concrete production, particle packing theory could significantly optimize the mixture of concrete to improve the properties of concrete and create more sustainable concrete product.



*Figure 1: Particle packing with sphere particles*

Various particle packing models have been explored for nearly 100 years. However, the complexity of the arrangement of particles has led to a slow development of optimization on particle packing models. For instance, the shape of particles could vary from sphere to angular, the size could vary from atoms to rocks, the way of packing could either be random or within certain rules. Consequently, these different effects would form a nonlinear connection with particle packing models [3].

In concrete production, the theory is embodied in two aspects: the grading of aggregates and the particle-size distribution (PSD) of binder materials. The basic principle to apply the theory on aggregates is to reduce the voids among various aggregates to lower the binder materials demand; on binder materials is to reduce the voids to lower water demand[4].

### **1.1 Particle packing theory on binder materials**

As to binder materials, supplementary cementitious materials (SCM) have been used to partially replace cement in concrete and related products have been developed for a wide range of projects. Most of SCM are the by-products of industry, thus the application of it could create more sustainable concrete. Besides of this, SCM effectively affect the property of concrete from multiple aspects such as reaction rate, bearing effect on workability, fineness of materials on packing density[4]. Generally, mean particle size of SCM is smaller than cement particles, thus, the application of SCM would increase packing density of binder materials system which result in a denser structure with lower voids content and less water demand. Previous particle packing models have been applied based on the PSD of binder materials, however further research is needed to increase the practicality of the theory on such as the evaluation of effect of particle shape, electrostatic force among binder material particles, and the effect of early hydration reaction[4]. With these effects, compared with aggregates, the investigation of the theory on binder materials remains more difficulties and will be carried out based on experiments after study of the theory on aggregates in the future.

### **1.2 Particle packing theory on aggregates**

Aggregates can take up to 75% by mass of concrete, to evaluate aggregates packing in term of grading would significantly affect the quality of concrete. Particle packing theory has been applied as a solid approach to optimize aggregates grading. However, the application of the theory is very limited due to the large number of different aggregates and their properties such as aggregate shape, texture, flakiness and elongation[4]. Therefore, this paper is focusing on the investigation of the effect of aggregates grading and some indirect quantification of the effect of aggregates properties on the property of concrete.

Generally, based on workability required, researchers need to design concrete mix with the selected type of aggregate to meet the requirement of concrete property. Road Note No.4 mix design method[5], in Figure 2, and Ready-Mix curve, in Figure 3 **Error! Reference source not found.**, encourages researchers to select aggregate grading based on binder material content. Road Note No.4 mix design method was carried out by laboratory and field experiments in UK, which is also called the grading curve method. Based on predicted compressive strength, cement content and W/C ratio, aggregate grading could be determined from the curve. Overall, a lean mix, which means a low cement content, requires fine aggregate grading. By contrast, a rich mix requires coarse aggregate grading. Meanwhile, Ready-Mix curve was used in Ready-Mix Australia, which remains the same trend for the preferred aggregate grading curve depending on the cement content[6].

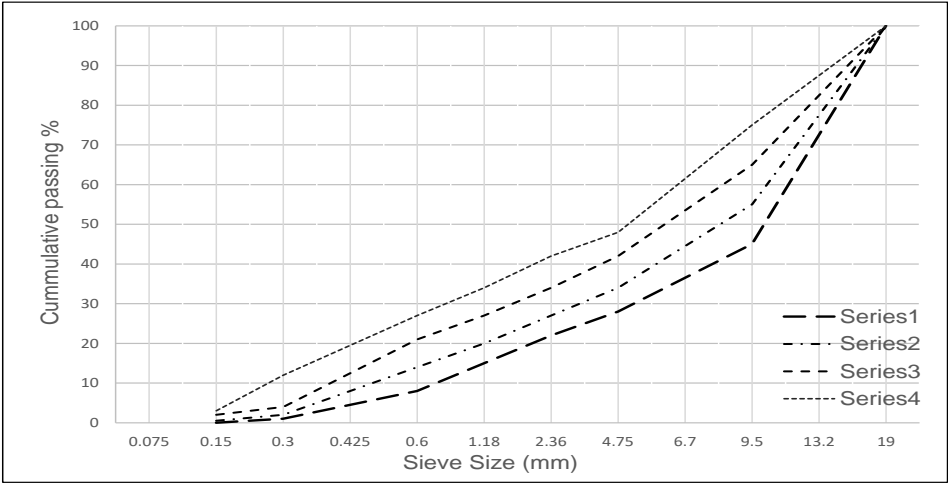


Figure 2: Aggregate grading chart of Road No. 4 mix design method

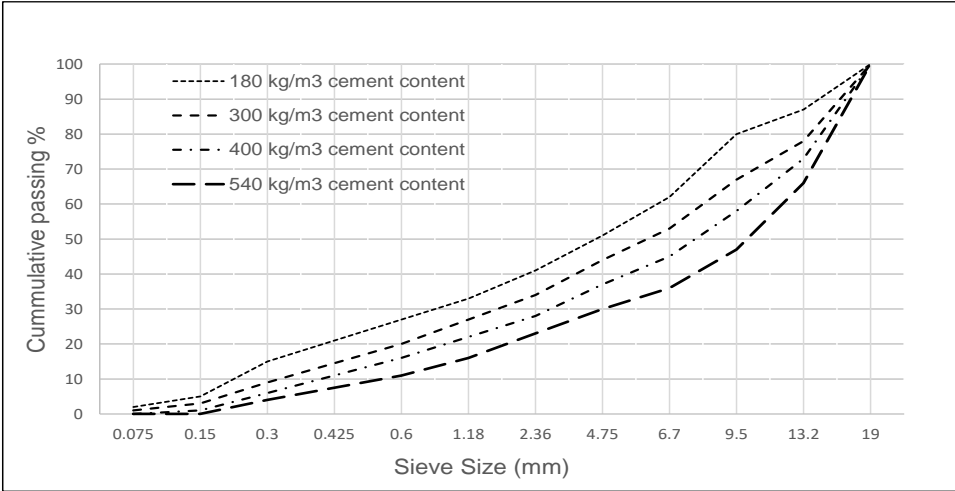


Figure 3: Aggregate grading charts of Ready-Mix Curve

Overall, these methods report grading curves are based on cement content and aggregate properties including aggregate shape, texture, flakiness and elongation. In practice, free water is related to workability, whereas W/C ratio is a nonnegotiable element to the target compressive. Thus, it could be predicted that fresh and hardened concrete properties will be affected by the chosen aggregate grading curve.

Particle packing curves and mathematic packing models are the most common approaches to optimize the grading curves. Fuller curve[7] is the most popular and used size distribution curve. The curve is designed to lead the mixture of particles to maximum packing density, which is the ratio of solid particles volume to total volume, to achieve the lowest voids volume. It is based on the equation shown in Equation 1 and plotted as red lines in Figure 4, for the application of aggregate grading.

$$P(d) = 100 \times \left( \frac{d}{d_{\max}} \right)^n \quad (1)$$

$P(d)$ : cumulative volume of particles passes the sieve with size  $d$ ;  $d$ : size of particles to determine;  $d_{\max}$ : maximum diameter of the coarse particles;  $n$ : distribution index of the particles.

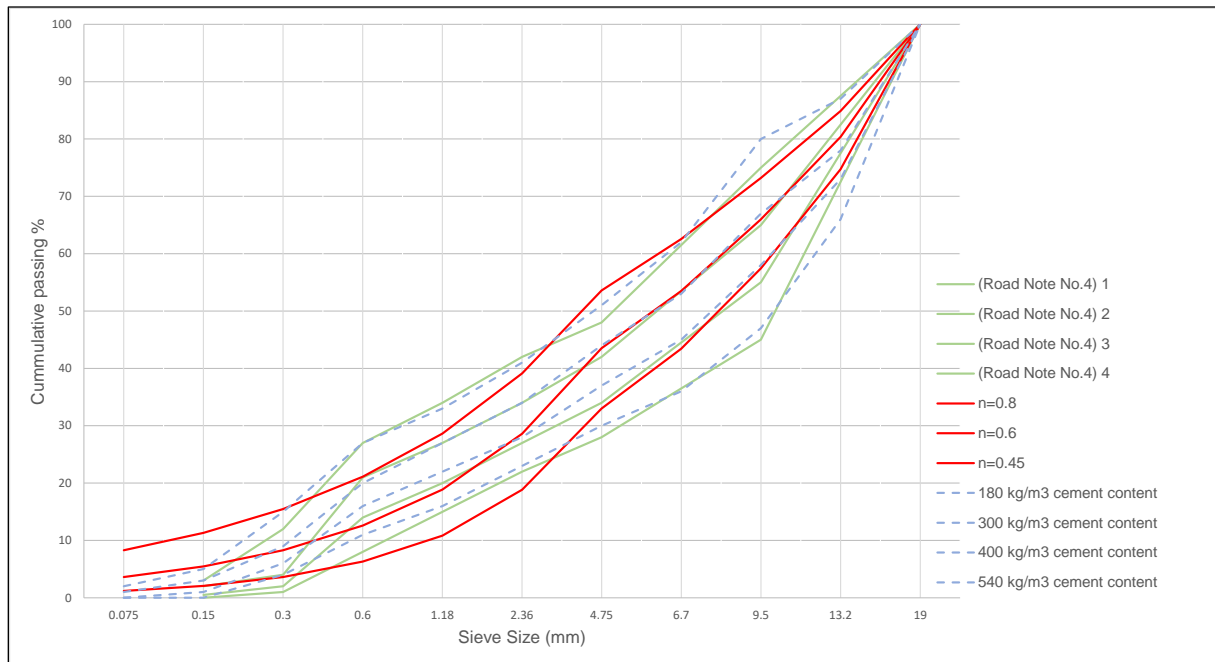


Figure 4: Aggregate Grading of Fuller Curve  $n=0.8$ ,  $n=0.6$  &  $n=0.45$

Considering different properties of aggregates such as aggregate shape, texture, flakiness and elongation, the distribution index  $n$  needs to be adjusted [8]. In Figure 4,  $n=0.45$  defines the curve for coarse grading,  $n=0.6$  defines the curve for medium grading and  $n=0.8$  defines the curve for fine grading. By comparison with Road Note No.4 curve (green curves) and Ready-Mix curve (blue curves), it is found the Fuller curves are very similar to them with sieve size above 4.75mm which is the grading range of coarse aggregate. When sieve size is lower than 4.75mm, Fuller curves require higher content of aggregates between 1.18mm to 2.36mm and a decent amount of dust under 0.075mm. This is due to dust could significantly fill up the very fine voids and achieve the maximized packing density. However, the increasing amount of these particles would significantly lower the workability of concrete [9], which is the lack of the theory. In another word, with the benefit of the densest structure, theoretic models may have defects on other concrete properties.

Besides the grading curves, mathematic packing models were studied since the 1930s. Furnas model is the first particle packing theory with the application of a mathematics model to describe the packing structure of a binary mix [10, 11]. Gradually, with the attraction to perfect packing models, researchers have created a number of different theories, such as Aim and Goff model [12], Linear Packing Density Model [13] and compressible packing model (CPM) [11]. Compared with previous models, CPM considers the interaction between particles and the effect of different packing forms to determine packing density. However, there is still a lack of consideration of particle properties, such as particle shape and reaction between binder particles. Therefore, the application is still limited in predicting realistic situations because of the complexity of concrete components.

## 2 Materials and Methods

### 2.1 Raw material

To investigate the effect of aggregate grading on concrete properties, general purpose Portland cement is the only binder material involved to simplify the experiments. Four different aggregates were prepared in the experiments including manufactured granodiorite aggregates: coarse aggregate 1, coarse aggregate 2 & coarse sand; and natural dune sand: fine sand. The grading chart of these four aggregates was tested and listed in Table 1.

Table 1: Grading of coarse aggregate 1, coarse aggregate 2, coarse sand and fine sand.

Sieve size		19.0mm	13.2mm	9.5mm	6.7mm	4.75mm	2.36mm	1.18mm	600µm	425µm	300µm	150µm	75µm
Cumulative Pass %	Coarse aggregate 1	98	52	11	1	1	1	1	0	0	0	0	0
	Coarse aggregate 2	100	100	89	49	18	4	3	0	0	0	0	1
	Coarse sand	100	100	100	100	99	83	58	39	31	25	14	9
	Fine sand	100	100	100	100	100	100	100	100	96	71	25	1

### 2.2 Testing Method

All the experiments were carried out in a laboratory, as per AS 1012, NZS3111:19-1986 and BS EN 12350-5 standards to retain the consistent procedures and minimum manual handling errors. Road Note No.4 mix curve and Ready-Mix Curve have proven practicality; however, they were mostly widely used for natural aggregates. With the decline in the availability of natural resources, manufactured materials have become prominent replacements for concrete production. Compared with natural aggregates, manufactured aggregates are more angular and flakier. Thus, experiments with manufactured aggregates were carried out to determine the practicality.

From **Error! Reference source not found.**, the similarity of Road Note No.4 mix curve and Ready-Mix Curve is clearly observed. The major difference is Road Note No.4 mix curve has a lack of information of aggregate size under 0.15mm. To investigate the effect of aggregate grading, concrete mix design is based on Ready-Mix Curve to evaluate concrete properties, including workability, compressive strength and drying shrinkage.

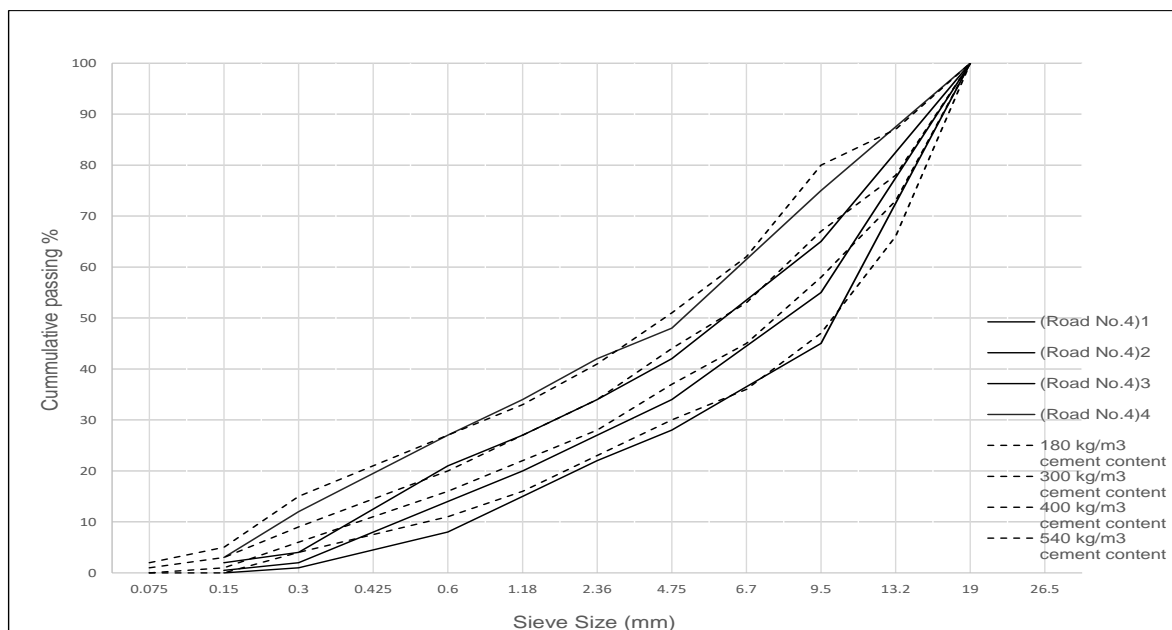


Figure 5: Aggregate grading charts (Road Note No.4 Mix Curve & Ready-Mix Curve)

### 3 Experiment Results

#### 3.1 Effect of total aggregate grading on the properties of fresh and hardened concrete

Two sets of concrete mixes were designed to evaluate the fresh and hardened concrete properties based on different preferred grading curves with certain cement content listed in

Table 2. The first set of mixes are from Mix 1 to Mix 3, containing 395 kg/m<sup>3</sup> cement, with a W/C ratio of 0.56. Aggregate grading was modified to fit three curves: Mix 1 fits 400 kg/m<sup>3</sup> cement preferred grading curve as the coarse mix, Mix 2 fits 300 kg/m<sup>3</sup> cement preferred grading curve as the medium mix and Mix 3 fits 180 kg/m<sup>3</sup> cement preferred grading curve as the fine mix, shown in **Error! Reference source not found.**

Similarly, the second set of mixes is from Mix 4 to Mix 6, containing 300 kg/m<sup>3</sup> cement, with a W/C of 0.73. Aggregate grading was modified to fit three curves: Mix 4 fits 400 kg/m<sup>3</sup> cement preferred grading curve as the coarse mix, Mix 5 fits 300 kg/m<sup>3</sup> cement preferred grading curve as the medium mix and Mix 6 fits 180 kg/m<sup>3</sup> cement preferred grading curve as the fine mix, shown in **Error! Reference source not found.**

Table 2: Six concrete mix designs

Concrete Trials No.	Mx 1	Mix 2	Mix 3	Mix 4	Mx 5	Mix 6
Mix Description	395 kg/m <sup>3</sup> cement			300 kg/m <sup>3</sup> cement		
	Coarse	Medium	Fine	Coarse	Medium	Fine
Cement (kg/m <sup>3</sup> )	395	395	395	300	300	300
Coarse aggregate 1 (kg/m <sup>3</sup> )	870	680	460	870	680	460
Coarse aggregate 2 (kg/m <sup>3</sup> )	400	450	580	400	450	580
Coarse sand (kg/m <sup>3</sup> )	550	590	500	570	620	540
Fine sand (kg/m <sup>3</sup> )	65	155	330	125	210	370
Water Reducer (ml/100kg)	350	350	350	350	350	350
Retarder (ml/100kg)	180	180	180	180	180	180
W/C ratio	0.56	0.56	0.56	0.73	0.73	0.73

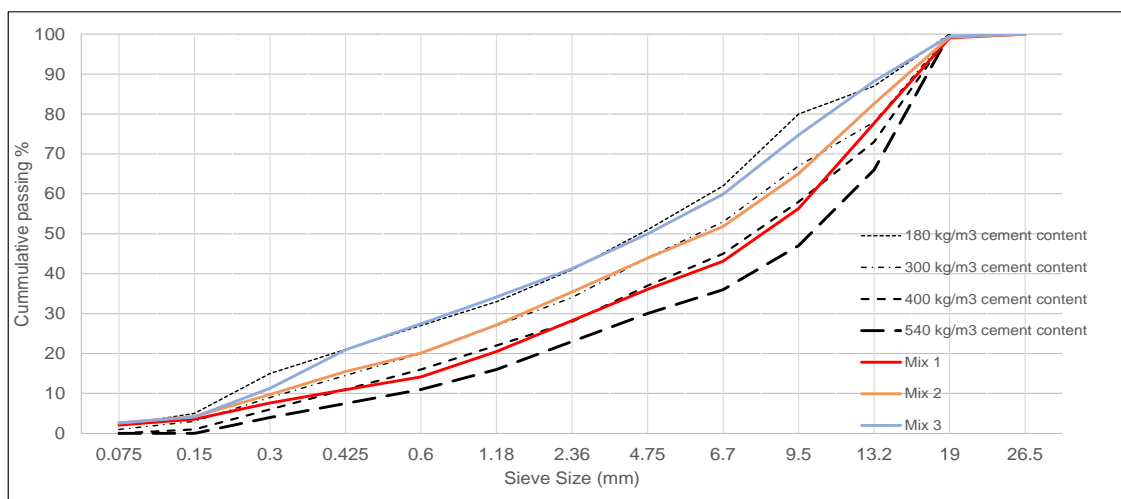


Figure 6 Aggregate grading charts of 395 kg/m<sup>3</sup> cement content concrete mixes

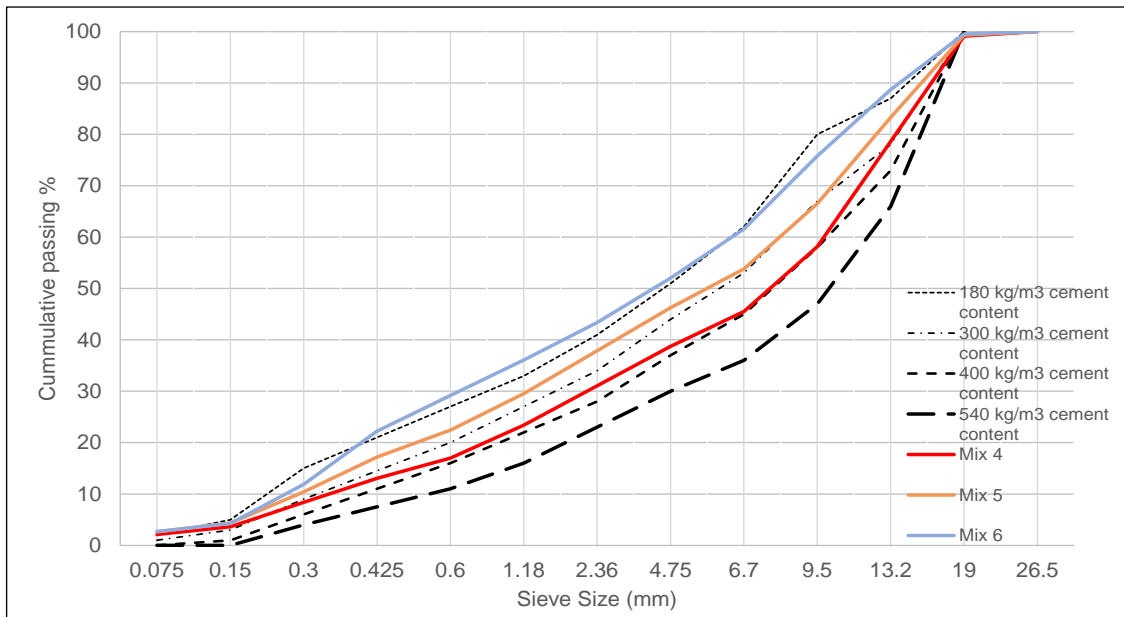


Figure 7 Aggregate grading charts of 300kg/m<sup>3</sup> cement content concrete mixes

### 3.2 Fresh concrete property

Slump (AS1012.3.1) and flow table test (BS EN 12350-5) was applied to investigate the fresh property of concrete. With a fixed W/C ratio of 0.56, from **Error! Reference source not found.**, it can be observed Mix 1 has a cylindrical shape of slump, however, the measurement of slump reached 110mm. Meanwhile, Mix 2 and Mix 3 obtained 115mm and 125mm slump but with better slump shape. In this case, slump results are not able to act as an indicator of workability. By applying vibration, flow table test presents the workability practicality. It is observed the flow of Mix 1 is 380mm, however, segregation appeared. Meanwhile, Mix 2 and Mix 3 both have a workable flow of 405mm and 410mm.

Mix 1, which was designed with the Ready-Mix aggregate grading of 400 kg/m<sup>3</sup> cement content, achieved the lowest workability. Thus, traditional grading curves require some adjustment when using manufactured aggregates. The main reason for the difference is believed to be from the angular shape of manufactured aggregates compared with the rounded shape of natural aggregates. Meanwhile, the production process generates more crushed surface areas with a higher roughness and more flaky pieces, which would increase the friction of aggregates in concrete and reduce the workability of concrete. Mix 2 gave the most cohesive concrete with no segregation of aggregates in the flow test. This mix used medium aggregate grading recommended by Ready-Mix for concrete with binder content of 300 kg/m<sup>3</sup>.

To evaluate the effect on workability from aggregate properties such as shape, surface texture and flakiness, more concrete mixes will be carried out with natural aggregates. Three concrete mixes will be designed based on the same theory with same grading and W/C ratio will be adjust with the target slump of 120±10mm.

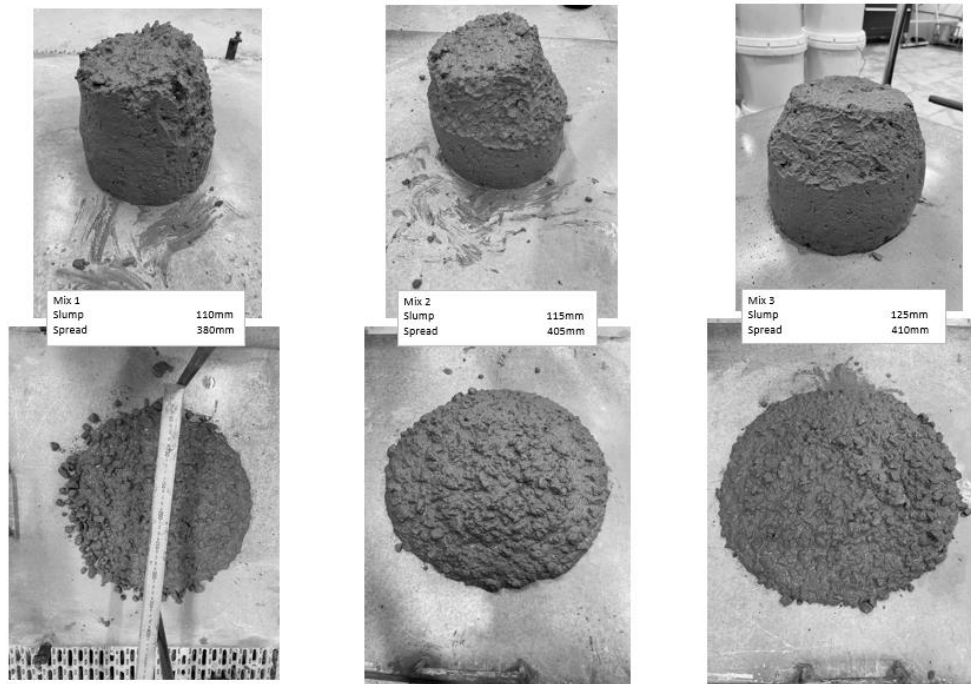


Figure 8 Workability of 395kg/m<sup>3</sup> cement mixes with w/c ratio of 0.56

Similar results were found on Mix 4, Mix 5 and Mix 6, which gave a slump of 110mm, 115mm and 125mm respectively. At the same time, with the spread of Mix 4 reaching 390mm, aggregates were exposed on top of concrete, which presents poor workability. Meanwhile, Mix 5 and Mix 6 remained a good spread shape and reached 415mm and 425mm, shown in **Error! Reference source not found.** Therefore, with a higher amount of coarse aggregates content and better packing structure, Mix 5 would be the preferred mix design.

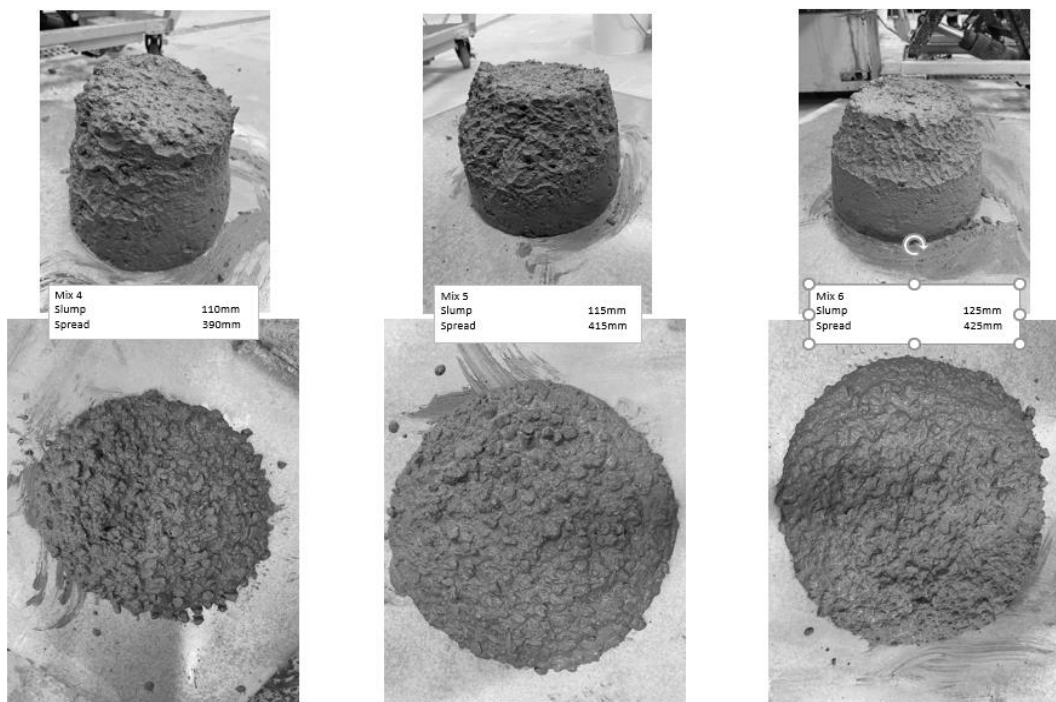


Figure 9 Workability of 300kg/m<sup>3</sup> cement mixes with w/c ratio of 0.73

### 3.3 Hardened concrete property

With the same W/C ratio, both sets of concrete mixes developed nearly equivalent compressive strength within their groups as shown in Table 3 (AS1012.9). In the first set of three mixes, Mix No. 3 obtained



the highest compressive strength, which is roughly 2 MPa higher than Mix No.1 at 28 days. However, in the second set, Mix No.4 achieved higher compressive strength than another two mixes. At 3 days, it led by nearly 3MPa compared with Mix No.6, and the difference reduced to less than 1 MPa at 28 days.

Table 3 Compressive Strength Results

	3 days Compressive Strength (MPa)	7 days Compressive Strength (MPa)	28 days Compressive Strength (MPa)
Mix 1	32.0	38.5	45.0
Mix 2	31.5	39.0	46.0
Mix 3	33.0	41.0	46.5
Mix 4	22.0	27.5	32.5
Mix 5	20.0	26.5	31.5
Mix 6	19.0	26.0	32.0

Drying shrinkage results show the values as predicted. Coarse aggregates grading developed the lowest drying shrinkage in both sets of trials, shown in Figure 10 & Figure 11. Mix 1 and Mix 4 developed about 25% and 8% reduction drying shrinkage value at 28 days compared with Mix 3 and Mix 6. The results show the coarse aggregates grading has a higher effect on drying shrinkage property with a low W/C ratio.

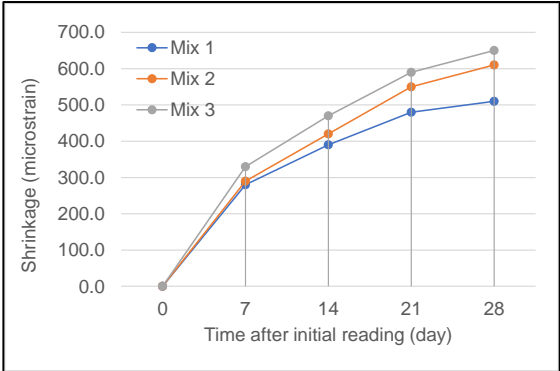


Figure 10 Drying shrinkage of 395kg/ m<sup>3</sup> cement content concrete

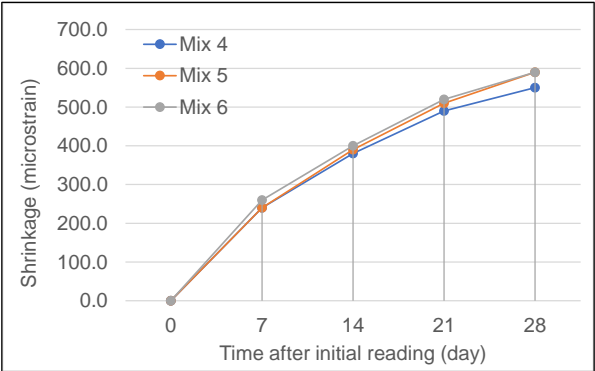


Figure 11 Drying shrinkage of 300kg/ m<sup>3</sup> cement content concrete

#### 4 Characteristics of aggregates

To investigate the effect of fine aggregate grading, 'New Zealand flow cone test' NZS 3111:19-1986 was applied to evaluate the flow time and voids content by combining different proportions of coarse sand and fine sand, shown in Table 4.

Table 4: Results of New Zealand Flow Cone Test

Sample No.	1	2	3	4	5	6	7	8	9	10	11
Sample Description	100% Coarse Sand	90% Coarse sand + 10% Fine sand	80% Coarse sand + 20% Fine sand	70% Coarse sand + 30% Fine sand	60% Coarse sand + 40% Fine sand	50% Coarse sand + 50% Fine sand	40% Coarse sand + 60% Fine sand	30% Coarse sand + 70% Fine sand	20% Coarse sand + 80% Fine sand	10% Coarse sand + 90% Fine sand	100% Fine sand
Flow Time (s)	23.90	23.70	22.55	21.95	21.30	20.80	20.35	20.10	19.95	19.90	19.75
Average Percentage of Voids (%)	38.90	38.71	38.37	38.37	39.26	39.54	39.76	41.08	41.74	42.40	42.97
Packing density (%)	61.10	61.29	61.63	61.63	60.74	60.46	60.24	58.92	58.26	57.60	57.03
Packing Density (t/m <sup>3</sup> )	2.63	2.63	2.62	2.62	2.61	2.61	2.61	2.60	2.60	2.59	2.59

With the increasing of the portion of fine sand, flow time reduced nearly 20% from 23.90s on Sample No.1 to 19.75s on Sample No.11. The reason is assumed, as natural sand, the particle shape of fine sand is relatively more rounded. Therefore, when increasing fine sand, a growing number of particles perform the bearing effect, reducing the friction among particles and improving the flowability of the sample.

Meanwhile, voids ratio dropped from 38.90% on Sample No.1 to 38.37% on sample 3 & 4, then significantly increased to nearly 43% on Sample No.11.

From Table 1, it is found fine sand holds a very narrow grading range and contains a large number of fine particles from 150 $\mu$ m to 450 $\mu$ m. Thus, proper addition of it would adjust the grading of Coarse sand and enhance the particle packing structure. However, with continually increasing fine sand, these fine particles started dominating, leading samples to an over fine total grading curve with a poor packing structure, shown in Figure 12. Fuller curves with size distribution index  $n=0.3$ ,  $n=0.4$  &  $n=0.5$  were plotted in **Error! Reference source not found.** as the reference curves for the comparison of grading curves. It is observed, Sample No.1 has the highest similar trend of Fuller curves while Sample No.11 has the lowest similarity. Thus, voids content of Sample No.1 to Sample No.4 is relatively lower than others.

Meanwhile, the grading curves of these samples are all located outside of the range of Fuller curves because of the low content of coarse (above 2.36mm) and fine (under 150 $\mu$ m) particles in the grading. In concrete production, fine particles under 75 $\mu$ m could significantly reduce the workability of concrete, therefore, maximum packing structure may not have the practicality.

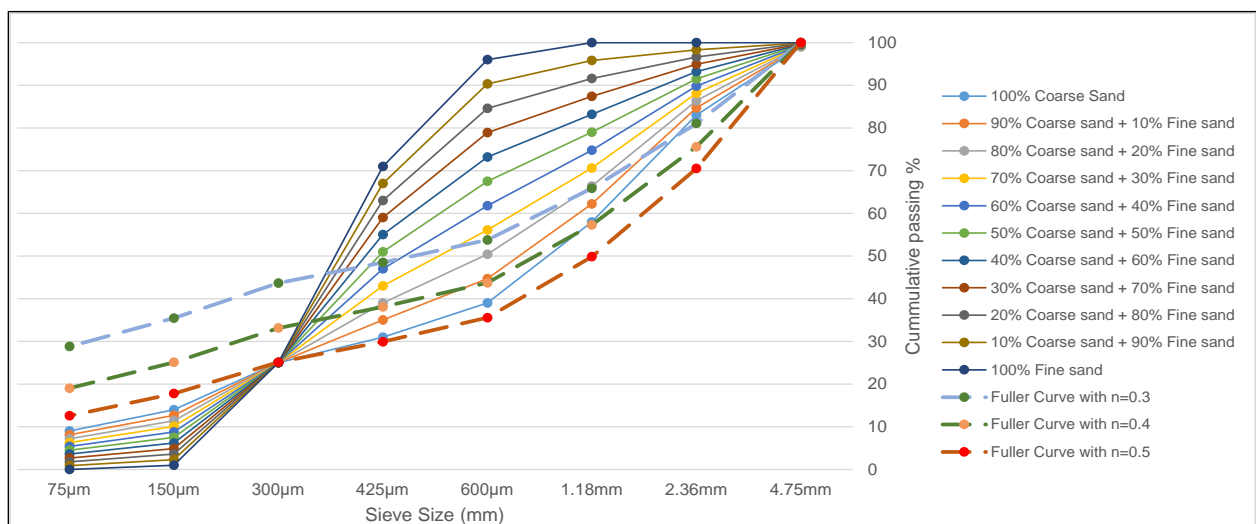


Figure 12 Total Grading of Sand combination and Fuller Curves with  $n=0.3$   $n=0.4$  &  $n=0.5$

To further investigate the effect of total grading of these samples, six mortar mixes were involved aiming at monitoring the compressive strength developing and the flow difference. Sample No.1, 3, 5, 7, 9 & 11 were brought into mortar mixes Mix M1, M2, M3, M4, M5 and M6, with a remaining 20% difference on the portion of sand, shown in Table 5. The W/C ratio of these mortar mixes is fixed at 0.49 to make the flow and compressive strength comparable. The results show an increasing trend when increasing fine sand use. By comparing with the flow time from 'New Zealand flow cone test', it is found mortar has

better workability by involving the sand with lower flow time. Therefore, the flow time could be applied to the prediction of the workability of mortar mixes. The reason is the same as flow time because fine sand is more rounded and shows the bearing effect that could reduce the friction among particles.

Mix No.	Mix M1	Mix M2	Mix M3	Mix M4	Mix M5	Mix M6
Portion of Coarse sand	100%	80%	60%	40%	20%	0%
W/C ratio	0.49	0.49	0.49	0.49	0.49	0.49
Flow	66%	66%	71%	72%	76%	89%
7 days Density (kg/m <sup>3</sup> )	2327	2307	2268	2185	2143	2130
28 days Density (kg/m <sup>3</sup> )	2331	2320	2294	2206	2153	2138
7 days Compressive Strength (MPa)	45.6	46.9	45.6	38.6	37.5	34.7
28 days Compressive Strength (MPa)	54.3	54.9	54.3	44.2	40.6	37.2
Packing density of sand (%)	61.10	61.63	60.74	60.24	58.26	57.03

*Table 5: Test results of Mortar mixes*

Compressive strength tests were carried out and listed in Table 5. Mix M1 which is a 100% coarse sand mix, achieved 45.6 MPa and increased slightly to 46.9 MPa from Mix M2 with 20% replacement with fine sand, then kept dropping to 34.7 MPa from Mix M6 which is a 100% fine sand. Similarly, Mix M2 achieved highest 28 days compressive strength 54.9 MPa and Mix M6 achieved lowest 28 days compressive strength 37.2 MPa. Late age compressive strength will be tested in the following days.

The trend of change on compressive strength fits the trend of the previous particle packing density results. Combination with 20% fine sand achieved the highest packing density and compressive strength, while 100% fine sand has the lowest packing density and compressive strength. It presents the benefit from the optimization of aggregate grading on compressive strength.

## 5 Conclusion

This study investigated the effect of aggregates grading and some indirect quantification of the effect of aggregates properties on the fresh & hardened concrete properties. Below summarizes the important findings from this study:

- Concrete mixes were designed with traditional preferred grading curves 'Ready-Mix curve'; however, concrete properties were not achieved as expected. Therefore, an update on these curves is required with the application of manufactured materials.
- On another hand, concrete trials show a significant improvement in the drying shrinkage property by increasing coarse aggregates. Meanwhile, concrete compromises the workability and results in low practicality. It is due to theoretical particle packing models having a huge lack of connection with the properties of materials, such as aggregate shape and texture. Thus, further study is urgently required to consider these effects to achieve better applicability.
- From the experiments on fine aggregates, results demonstrate the association between compressive strength and packing density, as well as flow of mortar and the flow time of fine aggregates, which proves that particle packing theories could be applied to optimize concrete properties and create more sustainable concrete product.
- From traditional experiments, results may not be accurate enough to distinguish the minor difference as desired. Optimization of experiment procedures is required to involve even less manual handling. Furthermore, some of the experiments can only be implemented as quality control methods, which requires further investigation to evaluate and quantify the effect on the concrete product. Therefore, an improvement on traditional test methods or new test methods may need to be developed.

## 6 Acknowledgement

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