Physiochemical characterisation of Malaysian white aggregate for use in sustainable concrete structure

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Abstract: The purpose of this study is to investigate the feasibility of using white aggregate as a construction raw material by determining its physiochemical properties, as well as its surface morphology. The results of a laboratory experimental programme aimed at establishing chemical and mineralogical characteristics of aggregate and its likely influence on concrete performance are reported in this paper. UV visible spectroscopy (UV-vis), X-ray fluorescence (ED-XRF) and X-ray diffraction (XRD) analyses were used for chemical and mineralogical characterisation, respectively. Surface morphology was determined by using scanning electron microscopy (SEM) instrument. It was observed that Malaysian white aggregate contains large amount of silt materials (about 9%). It also contains high amount of SiO_3 (62.5%) and K_2O (6.94%) compared with other natural aggregates. The surface morphology analysis clearly revealed the existence of palygorskite and smectite materials in the studied aggregate.

Keywords: stone aggregates; physiochemical properties; microstructure; wet chemistry.

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1 Introduction

Stone aggregates are fundamental elements of construction concrete, with estimated annual production of 10 billion cubic metres (Meyer, 2009). Since aggregates occupy between 70% and 80% of concrete volume, it is very important to know their physiochemical properties and their effect on concrete properties (De Brito and Saikia, (2013). Considering sustainable development, the understanding of construction material and its behaviour in asphalt and cement applications has always been a challenge. It is of utmost importance to assess and evaluate the properties at material level and as part of the mix for durability and monitoring point of view. The use of right quality and type aggregates is very important and needs to be well addressed for sustainable concrete structure (Braga et al., 2012; Smith et al., 2012; Pereira et al., 2012).

Last few decades, many researchers have clarified the influence of aggregate characteristics on the overall quality of concrete (Lo et al., 2008, 2007; Fragoulis et al., 2004). Aggregate characteristics have a major effect on fresh and hardened concrete behaviour. The main aggregates characteristics affecting the concrete properties are shape and texture; gradation; absorption; mineralogy; compressive strength and elasticity modulus; maximum size; specific gravity; sulphates attack resistance and hardness. Once the influence of each individual property is determined on concrete behaviour, it shall be possible to design more cost-effective mixes. Various studies have shown the effect of aggregates' physical properties on the strength and Young's modulus of concrete (Fragoulis et al., 2004; Topçu and Lu, 2010; Ruggero et al., 2010. However, the transition zone of expanded aggregates can be similar to the ordinary aggregates depending on the characteristics of the aggregates shell. To provide the information on surface morphology of the aggregates and concrete, scanning electron microscopy measurement is very common (Topçu and Lu, 2010; Ruggero et al., 2010). It is well known that aggregate texture, shape and gradation characteristics affect the strength, stiffness, retraction and permeability and durability of fresh concrete and they also affect the workability, finishing, bleeding and segregation of hardened concrete.

Owing to the sheer volume of construction activities in Bangladesh, the market of building materials including aggregates is stressed. The commercially available natural aggregates are either local black aggregate or imported aggregate. Recently huge amounts of white aggregates have been imported from Malaysia for construction purposes. Till now there no standard regulation or quality monitoring system for these aggregates which is mandatory for sustainable development of the country.

The study was undertaken to investigate the physiochemical and mineralogical composition of white aggregate recently imported from Malaysia, which included the assessment of their influence on concrete properties. It was also aimed to explain the surface morphology of the studied aggregate.

2 Materials and methods

2.1 Sample collection and processing

To carry out all experiments, four types of samples were prepared each time; which were coarse aggregate, fine aggregate, powdered sample and aqueous stock solution. As a requirement of ASTM standard procedure, coarse aggregate was collected in different size and shape, from different parts of raw material store place. Conventional method was applied to separate desired size of aggregate by sieve and used for physiochemical experiments. For elemental and morphological analyses, powdered sample was prepared by conventional grinding process. A hammer and 75 µm (sieve/200) were used to collect powder sample.

Figure 1 Image of Malaysia white aggregates (see online version for colours)

2.2 Stock solution preparation for wet chemistry tests

Aqueous stock solution was prepared ASTM standard method for wet chemical analysis. At first 50 gm of dry sample was weighted and transferred to the 500 ml volumetric flask. The volumetric flask filled up with distilled water. Kept the volumetric flask for 15 hrs on magnetic stirrer for digestion and temperature maintained up to 60°C. Sample transferred to centrifuge equipment after digestion, set the motion of centrifuge at 4000 rpm and continue this process for 10 min. After that clean solution was collected in a beaker to continue wet chemical analysis.

2.3 Characterisation of sample

For characterisation of sample various instruments were used. Scanning electron microscope (SEM) was used for morphology analysis. The thermo-scientific ARL QUANT'X energy dispersive X-ray fluorescence (EDXRF) spectrometer analyser was used for determining m/m% for wide band metals. X-Ray detraction (XRD) used for identified the crystal structure, metallic component. We have used HACH (DR 2700) portable spectrophotometer for measuring the concentration of ion present in sample solution.

3 Results and discussions

3.1 Physiochemical analysis

Acid and crystallinity test

Acid test is performed to check the insoluble matter content and insoluble residues in aggregate. Acid test also gives the weather stability assumption. A good aggregate maintains its sharp edges and keeps its surface free from powder at the end of acid test. If the edges are broken and powder is formed on the surface, it indicates such an aggregate will have poor weathering quality. In our acid test experiment, we observed unbroken edges of the studied aggregate that produced little amount of powder and changed its colour. It is well known that the degree of crystallinity of silica aggregate is one of the major factors influencing alkali reactivity. Poorly-crystallised polymorphs of silica have higher Gibbs free energies and are thus thermodynamically less stable than well-crystallised species, which means that reactivity decreases over the range from amorphous to well-crystallised species of silica. This has been confirmed by our experiment that the sample shows good crystalline stability.

Silt and clay test

This test is generally conducted in order to determine the volumetric percentage of silt in fine aggregate. In this test, we have used mixed aggregate choose from different part of aggregates stock. To classify fine aggregate, we have used 4.75 mm sieve and followed BS 882 method for this measurement. Based on the analysis, the maximum silt content was observed about 9.2%. BS 882 and ASTM C40 recommends that no more than a maximum of 6% silt content for fines aggregates be used in concrete structure (Cho, 2013). Previous studies indicate that silt fine content has an effect on concrete's durability, especially when silt fine content in concrete is more than 5% (Cho, 2013). It was also reported that concrete exhibits higher chloride ion penetrability when silt fine content is more than 5%.

It was observed that the excess amount of silt materials may present as loose dust and may form a coating on the aggregate particles. Even thin coatings of silt or clay on aggregate particles can be harmful because they may weaken the bond between the cement paste and aggregate. If certain types of silt or clay are present in excessive amounts, water requirement may increase significantly. Excessive silt in aggregate may delay setting and hardening of concrete, may reduce strength gain, and in unusual cases may cause concrete deterioration.

Density, moisture and porosity test

The moisture content of an aggregate is an important factor when developing the proper water/cementitious material ratio. All aggregates contain some moisture based on the porosity of the particles and the moisture condition of the storage area. The density of the aggregates is required in mixture proportioning to establish weight-volume relationships. Specific gravity is easily calculated by determining the densities by the displacement of water. Table 1 represents the physical properties of white aggregate.

Table 1 Physical properties of coarse aggregate

Test type	*Measured value*	*Permissible/standard limit*	*Testing method*
Moisture content (%)	0.112	1–2%	ASTM C70
Water absorption (%)	0.21	Max 2%	ASTM C127
Density (kg/m^3)	2650	2,400	ASTM C29
Specific gravity	2.65	2.6–2.9	ASTM C127
Porosity (%)	12.6	-	ASTM C29

Comparing with standard value, our studied aggregate shows low moisture content as well as water adsorption rate. On the other hand, it shows 12.6% porosity. It is obvious that a porous aggregate produce concrete of unit weight, as that of light weight aggregates. Bond between aggregates and surrounding hydrates paste of cement strongly depends on porosity.

3.2 Wet chemical analysis

Water-extractable chloride, when present in sufficient amount, has a potential to initiate or accelerate corrosion of metals, such as steel, embedded in or contacting a cementitious system, such as mortar, grout, or concrete. This test method is applicable when aggregates contain a high background of naturally occurring chloride. Test method ASTM C1218/C1218M determines water-soluble chloride. Excessive amounts of mobile Sulphate, derived from aggregates or other constituents in concrete, can cause disruption due to expansion.

By ultraviolet visible spectrophotometer (UV-vis) we have measured the exact concentration of different ion present in aggregates solution and the obtained results are shown in Table 2. It was observed that the amount of dissolve ion presented in white aggregates are within standard limit.

Table 2 Water soluble ion content amount present in aggregate

Ion name	*Measured value (mg ion/kg sample)*	*Standard limit*	*Method*
Nitrate (NO_3^-)	1.5	Max. 3%	ASTM C1580
Chlorine (Cl^-)	0.33	Max. 0.1 % by mass	ASTM C1218
Phosphate (PO_4^{2-})	0.21	No data	No data
Sulphate (SO_3^{2-})	27.48	15-55	ASTM C1580

3.3 Chemical composition analysis

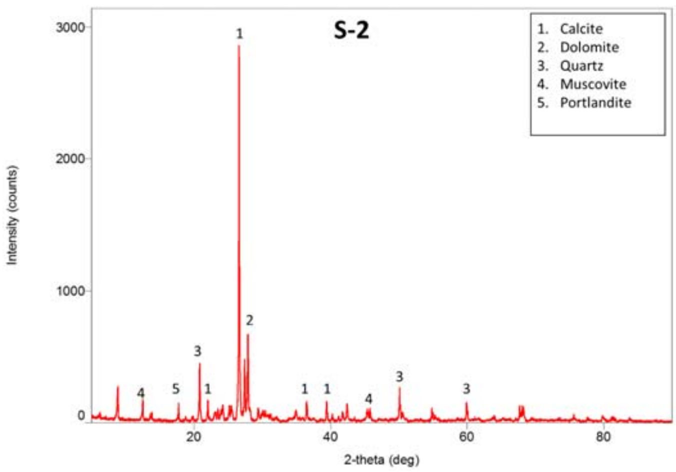
We have done EDXRF measurement for elemental analysis applications to check the presence of major and minor element in percentage. The details are presented in Table 3.

Table 3 Percentage of elements present in white aggregates

	Na_2O (wt.%)	MgO (wt.%)	Al_2O_3 (wt.%)	SiO_2 (wt.%)	CaO (wt.%)	K_2O (wt.%)	Fe_2O_3 (wt.%)
Value	2.03	0.83	16.80	62.50	3.89	6.94	5.02

The obtained EDXRF result resembling that white aggregate is enriching is SiO_2 and K_2O. The natural aggregates usually contain 30–50% silica (Lerch, 1955). High amount of silica content in aggregate may lead alkali-silica reaction in concrete mixture. The alkalis in the cement may react with certain silica in the aggregate and form gel around the aggregates. This gel was reported to cause deterioration of the concrete (Lerch, 1955; Neville, 1973). It was reported in many literatures that the most effective method to remove excess fine particles (somewhere it was noted as dust/silica) that can contribute to geotextile clogging is the agitation and pressure washing of aggregates before using in concrete structure.

Figure 2 XRD spectra of white aggregate (see online version for colours)

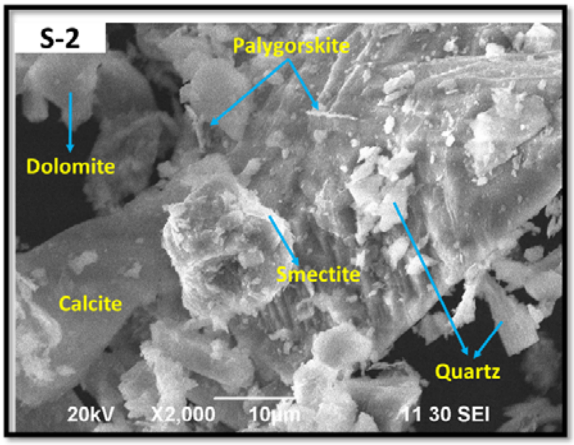


X-ray diffraction was done to identify the crystalline phases and the corresponding intensities of various compounds of the aggregate. The existence of calcite and dolomite in aggregate can be clearly observed for white aggregates. However, the intensity and peak region for quartz shows higher due to the existence of more silica in white aggregate confirmed by EDXRF experiment. Moreover, clear existence of muscovite and portlandite is observed. The muscovite peak comes from the hydrated phyllosilicate mineral of aluminium and potassium. Quartz and feldspar are more likely to have a solid origin. However higher amount of quartz usually observed for recycled concrete aggregates (Lerch, 1955).

3.4 Surface morphology study

SEM experiment was done to check the microcrystalline structure of the aggregates. Figure 3 shows the SEM image of white aggregate. From the image, presence of calcite, dolomite and some quartz is identified. On the other hand, the development of palygorskite $(Mg,Al)2Si_4O_{10}(OH){\bullet}4(H_2O)$ fibres between calcite crystals near smectite flakes was observed. Elongate palygorskite fibres developed as cement between calcite crystals, and fibre and fan-shaped bundles at the edge of smectite are clearly identified for white aggregate.

Figure 3 SEM image of white aggregate (see online version for colours)



4 Conclusions

White aggregate studied in this work contain large amount of silt materials and the experimental value is about 9%, whereas, the permissible world standard is maximum 4-6%. So, it is a big concern and there is a need for treatment before use in concrete structure. From EDXRF, it is clear that white aggregate contains high amount of silica and potassium. High amount of potassium can make aggregate more reactive. Moreover, the alkali-silica reactivity of aggregate (due to high silica content) is a critical parameter with regard to the concrete durability, and which affects its structural weakening and shortens its useful life. This finding is in good agreement with the XRD results. From

SEM, it was observed that palygorskite and smectite materials clearly exist in studied aggregate. An extensive study is suggested to correlate these findings with concrete structure.

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