Self-cleaning geopolymer concrete - A review

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Abstract. Concrete is the most widely used construction materials for building technology. However, cement production releases high amounts of carbon dioxide (CO₂) to the atmosphere that leads to increasing the global warming. Thus, an alternative, environmental friendly construction material such as geopolymer concrete has been developed. Geopolymer concrete applies greener alternative binder, which is an innovative construction material that replaces the Portland cement. This technology introduced nano-particles such as nanoclay into the cement paste in order to improve their mechanical properties. The concrete materials also have been developed to be functioned as self-cleaning construction materials. The self-cleaning properties of the concrete are induced by introducing the photocatalytic materials such as titania (TiO₂) and zinc oxide (ZnO). Self-cleaning concrete that contains those photocatalysts will be energized by ultraviolet (UV) radiation and accelerates the decomposition of organic particulates. Thus, the cleanliness of the building surfaces can be maintained and the air surrounding air pollution can be reduced. This paper briefly reviews about self-cleaning concrete.

1. Introduction

Concrete is the most widely used construction materials in the world. Concrete is the second most consumed substances on earth after water. Concrete is used everywhere such as office buildings, residential apartment, bungalow, any public places, etc [1]. Unfortunately, the main components contain in the cement contributed to the greenhouse gas emissions. Dust emissions during the cement manufacturing are one of the main issues that have been faced by the industries. A major source of air pollution is caused by concrete dust released by building demolitions and natural disasters [2]. Thus, the building surfaces will become dull and dirty. In addition, concrete also faces the problem of tending to become dirty when exposed to polluted area. These situations have pressured the industries to develop an innovative technology and find alternative materials that are environmentally friendly in order to support the sustainable development. Geopolymer concretes with self-cleaning behavior are green alternative to overcome this problem. This is because they offer tremendous environmental benefits such as decreasing the CO₂ emissions, economical, durable and environmentally friendly.
Geopolymer cement has their own unique properties which are able to withstand at high temperature and also resistant to salt and acid [3]. Self-cleaning concrete or photocatalytic concrete will keep the concrete surface free of dirt and keeps its colour, even in polluted condition and industrial areas. This concrete also called as green concrete due to its self-cleaning properties [1].

Self-cleaning technology is commercialized for indoor and outdoor applications. There are various applications of self-cleaning technology, such as in paints, glasses, textiles, tiles, etc. Glasses windows or tiles are modified with the addition of TiO$_2$ as photocatalytic thin films for their self-cleaning functions, which have been applied in the glass surface of the National Opera Hall, China. In Japan, eco-friendly, self-cleaning windows and roof tiles are widely used. Japan and Europe country were using this technology to make their polluted cities keep-clean and reduce the effect of air pollution [1]. Nanoparticles of TiO$_2$ are also applied on the construction materials especially in concrete in order to enhance the self-cleaning properties. Addition of titania in the concrete materials can contribute to the air-purifying and self-cleaning surfaces due to the photocatalytic process that has been activated by the UV light radiation. Thus, the building can maintain their aesthetical appearance and improve the quality of air by the degradation of airborne pollutants [4]. Self-cleaning is an automatic cleaning process on buildings or concrete [5]. This can be seen through the example of the white cement containing TiO$_2$ nanoparticles has been coated on the roof of Dubai Sports City’s Cricket Stadium and also used for designing church at Rome, Italy. From the result, it was proved that the buildings could keep their original colour over the years. While in automotive industries, self-cleaning coatings with antifogging properties have been used in order to develop the clean and glare free windows [5]. In latest research, ethanol suspension of perfluorooctyltriethoxysilane (C$_{14}$H$_{19}$F$_{13}$O$_3$Si) coated with anatase TiO$_2$ nanoparticle is used to produce paint that can create the self-cleaning surfaces [5]. There are several reviews found on TiO$_2$ coating, where they can improve the mechanical properties of cotton fibers and also high stain removal of bacteria [5]. Thus, the purposes of applying the self-cleaning materials such as titania onto the geopolymer concrete are, to enhance the aesthetic durability and depollution effect of the products.

2. Approaches to self-cleaning

In 1973, the principle of self-cleaning was discovered by the botanist Wilhelm Barthlott. The products from the reaction of self-cleaning are easily removed by rain or simple rinsing due to the phocatalytic actions with the addition of TiO$_2$ due to the hydrophilic surface as shown in Figure 1[5]. The application of titania as a photocatalyst to the concrete are focusing on the self-cleaning effect and air purifications [6]. Self-cleaning concrete involved photocatalytic reaction, which contribute to the elimination of volatile organic compounds (VOCs) such as formaldehyde. Photocatalytic reaction occurs with the presence of UV radiation from the sun. When light and heat strike the surface of concrete, TiO$_2$ use the energy to break the dirt into the basic oxygen, water, CO$_2$, nitrate and sulphate molecules. Photocatalyst convert air pollutants that contain toxic to lesser toxic forms in the presence of UV light [7]. Thus, depollution removed contaminants and impurities from the surrounding environment [8].
2.1. Photocatalyst

There are a lot of materials that act as photocatalyst. It can be seen through a few researches that have been carried out in order to investigate the influence of photocatalyst in cementitious materials [9]. Nanoparticles are used as photocatalyst in order to improve the performance of the cement properties. Nanoparticles that being added to cement can function as either an admixture in cement paste or to replace part of cement. These nanoparticles act as fillers in the empty space, well distributed, increasing rate of hydration and tendency to agglomerate during mixing. Nanoparticles that most common used in cement products are silicon oxide (SiO2), zinc oxide (ZnO), titania or titanium dioxide (TiO2), aluminium oxide (Al2O3) and carbon nanotubes [9]. Research reported that the addition of nano-silica into the cement paste influenced the mechanical properties of the products [9]. A few studies showed that by adding nanoparticles of TiO2 are mostly performed with photocatalytic activity due to its chemical stability and high catalytic activity. In concretes that contain nano-TiO2, the resistance to penetration of chloride is higher than concretes that contain the same amounts of nano-SiO2. This is due to the refinement of pore structure increases while the chloride penetration decreases with the content of nano-particles [3]. The addition of nano-TiO2 shows a positive result with fewer cracks in compact microstructure and enhances the formation of geopolymer [3]. The compressive strength can be improved by increasing the amount of nano-TiO2 in the modified cement [10].

2.1.1. Role of nanoparticles TiO2 in self-cleaning.

Titania is widely used to produce concrete pavements by mixing the nano-particles within the pavement surface. TiO2 has three different crystal structures, which are anatase, rutile and brookite. The efficiency of photocatalytic reaction was influence by TiO2 structure where anatase nanoparticles shows more efficiency in crystal condition and transparent when applied on the substrate. The cleanibility of the substrate can be enhanced by nanostructured titania [11]. The main reason TiO2 was loaded with cementitious materials because only water, oxygen and solar light required for photocatalysis reaction. In addition, TiO2 is a photocatalyst that can break down almost any organic compound with the presence of sunlight. If any dirt present on the surface, the water will penetrates...
under the dirt and removes it as illustrated in Figure 2 [12]. TiO$_2$ photocatalyst are usually are coated on a surface as thin films. Most photocatalytic coatings are prepared by post-deposition of the catalyst, and then apply onto substrate. They’re various method of coating TiO$_2$ with the cementitious materials such as sputtering, spray coating, sol-gel dip coating, electrodeposition and etc. These techniques of coating are used to incorporate the photocatalysts into the building materials [7]. When the properties of nanoparticles are transferred to the surface of substrates, it will become functionalization for self-cleaning application. TiO$_2$ are widely applied in different field such as air purification, anti-bacterial, food industry, production of paper, cosmetics, building materials and etc. TiO$_2$ are widely used because of its features, which is high catalysis efficiency, good chemical and thermal stability, low toxicity and low cost materials [11].

![Figure 2. Self-cleaning effect of n-TiO$_2$ at concrete surface [12]](image)

### 2.2. Methods of sample preparations

There are a lot of methods to prepare the sample by adding nano-TiO$_2$ onto the cement paste. There is a study about nano-TiO$_2$ with several of particles size (ground and sieved for 60 minutes in the sieve shaker), which have aims to investigate the effect of addition of nano-TiO$_2$ on fluidized bed fly ash based geopolymer. [3]. There is a recommendation to use TiO$_2$ as a solution by mixing water for better dispersion [3]. In another approach, the TiO$_2$ nanoparticles were soaked in water before mixing with cement, then, vibrated ultrasonically in 30 minutes for well distribution. After 3 minutes, the water was mixed with cement under rate of stirring 800 rph [14]. For sample preparation, ball are milling is used to mixed cement with various TiO$_2$ nanoparticles content (0%, 2.5%, 5.0% and 7.5%). Before mixing process, the TiO$_2$ powder was added to dry cement and subsequently water was added [15-16].

### 3. Self-cleaning testing

The self-cleaning test is conducted in order to evaluate self-cleaning abilities and depollution effects of the material. Photocatalytic performance is measured by photo degradation of methylene blue (MB)
solution or rhodamine B (RhB) in the concrete surface in order to evaluate self-cleaning activities. The photo-induced bleaching of MB dye to evaluate photocatalytic materials can be measured by using the UV/visible spectrophotometric. There are a rapid and inexpensive method to test photocatalytic activity of a film, which is using digital scanner and indicator ink such as resazurin (Rz), basic blue 66 (BB66) and acid violet 7 (AV7) [5]. RhB is extensively used in the laboratory test method of photocatalytic degradation because it can simulate particulate pollutants due to its similar structure as the airborne particulate compounds. RhB also water soluble with reddish colour, which are stable in alkaline environments such as mortars and make it good for testing [5]. The experimental study on the ability of silicate coating material, which contains various dosages of TiO2 to clean the surface by itself and for degradation of particulate pollutants. Based on the result of the experiment, an optimum dosage of TiO2 in the silicate coating is 15% TiO2 by mass of solid silicate are most effective in photocatalytic degradation of RhB [5]. The investigation proved that building materials which is coated with TiO2 have an ability in soot removal. These are based on changes in sample colour in RhB and photocatalytic degradation was tested by closed chamber experiment [4]. Cement plates that are prepared using Portland cement are co-modified with TiO2-N, C are tested by Multi N/C carbon analyzer in order to measured total carbon and nitrogen content. The degradation rate of Monoazo dye Reactive Red is determined by UV/vis spectrophotometer. The result shows that, the cement plates containing TiO2-N,C photocatalyst exhibit higher photocatalytic efficiency than unmodified cement plates [5-6].

In order to evaluate the depolluting effect, degradation of nitrogen oxide was determined by using continuous gas flow reactor under UV exposure (as shown in Figure 3). The study was designed a stirred flow reactor in order to measure photodegradation potential at the surface of photocatalytic cementitious materials. The result of the study shows that, the oxidation rate increased with the order of benzene, toluene, ethyl-benzene and o-xylene. Moreover, another study tested the photocatalytic activity of TiO2 by using a 2-propanol as a reactant of VOCs in the mortar. Different amount of 2-propanol are mineralized to acetone after a certain time under UV irradiation, which are analyzed using gas chromatography (GC) [7].

Figure 3. Continuous gas flow reactor [16]
4. Importance of self-cleaning

Air pollution has given negative implication for human health. Major pollutants that are produces from human activity are nitrogen oxide (NOx), sulphur dioxide (SO₂), volatile organic compound (VOCs), etc. Therefore, when photocatalytic reactions absorb UV from the sun, the hydroxyl radicals and superoxide anions are formed which have the ability to react with pollutant molecules such as SOx, NOx, and etc. Nowadays, atmospheric pollution also causes the external decay of buildings due to the deposition of organic matter and contaminants. Self-cleaning concrete has a potential to keep the city clean by reducing the air pollutants. Due to its properties, self-cleaning can save maintenance cost, thus extent the lifetime of the products. Moreover, self-cleaning also can reduce the heat that builds up on building with reflect more light, therefore, help in keeping city become cooler during hot season. Photocatalysts will remove odours and indoor air quality, so that, the city will become clean and beautiful [17]. The photocatalyst that involved in self-cleaning are also being used to purify water and fight the disease by reducing the spread of germs. The developments of photocatalytic cements have given an impact to depollution effect [5].

Self-cleaning are widely used in cementitious materials because it improve the mechanical properties and performance due to its properties. There are a study reported that mechanical properties of TiO₂ nanoparticles that added partially into cement are increase in tensile and flexural strength of the cement mortar [10]. There are a review about efficiencies of self-cleaning when TiO₂ are added to mortars or concretes. Most of TiO₂ mixed in mortars and concretes are remain unutilized because TiO₂ only active when exposed to light irradiation. There are also a few research studied about the addition of TiO₂ as coatings because coating layer are direct contact and exposed to pollutant. The result shows the concrete or mortar, which is covered by TiO₂ are less affected by cement hydration products. TiO₂ mixing with nano-particles are widely utilized to produce concrete pavements, thus contribute to mitigate urban air pollution [7]. White cement with addition of TiO₂ is used at the surface of buildings that can attribute to the durability of the visual aspect of the building. The whiteness of building will remain and maintained due to photocatalytic action.

5. Conclusions

This review aims to give an overview on self-cleaning concrete. This applications is promising to be utilized as a self-cleaning building materials. With these development of technology, air pollutions can be reduced because self-cleaning contribute to the elimination of VOC’s that present in indoor and outdoor atmosphere. Nowadays, air pollution is a serious problem in urban environment that can affect human health. Thus, self-cleaning concrete is an effective alternative to provide cleaner environment and maintain building appearance. Through the development of cementitious materials, UV radiation from the sun may represent one of the solutions to the pollution issues. In conclusion, self-cleaning can be used in various application, especially cementitious materials which contribute to sustainability and towards green environment. Further research need to be focused on producing a functional geopolymer concrete with self-cleaning behavior.
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