DEVELOPMENT OF KAOLIN BASED GEOPOLYMER COATING FOR LUMBER WOOD APPLICATIONS

by

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LIST OF ABBREVIATIONS

Al	Alumina
Al_2O_3	Aluminum Oxide
ASTM	American Society for Testing and Materials
Ca	Calcium
CaO	Calcium Oxide
CO	Carbon monoxide
CO_2	Carbon Dioxide
CSH	Calcium Silicate Hydrate
K	Potassium
KOH	Potassium Hydroxide
M	Molar Ratio
mm	Milimeter
MPa	Megapascal
Na	Sodium
Na_2SiO_3	Sodium Silicate
NaOH	Sodium Hydroxide
OH	Hydroxide
OPC	Ordinary Portland Cement
S/L	Solid/Liquid
SEM	Scanning Electron Microscopy
Si	Silicon
SiO_2	Silicon Dioxide
XRF	X-Ray Fluorescence
XRD	X-Ray Diffraction
rpm	Rotation per minute
kX	Thousand times magnification
Ton	Tonnes

GPa	Giga pascal
g	Gram
Kg	Kilogram
cm	Centimeter
%	Percentage
wt%	Weight Percentage
+	Plus
^{o}	Degree
^{o}C	Degree Celsius
h	hours
WA%	water Absorption percentage
LOI	Loss in Ignition
LG	lumber grade
MK	metakaolin
θ	theta
=	equals
Cps	Cycle per second
ISO	International Standards Organization
pH	potential hydrogen

PEMBENTUKAN SALUTAN GEOPOLIMER BERASASKAN KAOLIN UNTUK APLIKASI KAYU JENIS LUMBER

ABSTRAK

Pengunaan simen Portland biasa (OPC) terbukti telah digunakan sejak berabad dahulu sehingga kini dalam industri sivil. Kekurangan pilihan bahan semulajadi dengan pencirian yang sama atau penambah baik telah menyebabkan pengguna mengabaikan kekurangan Portland biasa. Pengkajian geopolimer salutan yang berasaskan kaolin bukan hanya bertujuan untuk meningkatkan produktiviti dan aplikasi geopolimer tetapi juga supaya berpotensi untutk mengantikan pengunaan Portland biasa dalam industry salutan. Tujuan utama kajian ini adalah untuk menyediakan reka bentuk optimum geopolimer salutan yang terdiri daripada kaolin iaitu sebagai sumber bahan aluminosilikat dan larutan pengaktik alkali. Reka bentuk campuran optimum memainkan peranan penting terhadap kekuatan mekanikal, ketumpatan, kadar peratus penyerapan air, analisis fasa dan analisis morfologi sebagi parameter penting terhadap kepekatan, nisbah pepejal/cecair, dan nisbah sodium silikat/sodium hidroksida telah dikaji. Kaolin sebagai sumber bahan asas geopolimer telah diperincikan melalui ujian analisis saiz zarah, analisis fasa, komposisi kimia dam analisis morfologi. Pes geopolimer salutan dengan reka bentuk kepekatan 8 M sodium hidroksida, 0.9 nisbah pepejal/cecair, 0.40 nisbah sodium silikat/sodium hidroksida yang di uji pada suhu 70 °C selama 24 jam telah memberikan kekuatan mekanikal tertinggi iaitu 2.4 MPa (7 hari), 2.98 MPa (28 hari) dan 4.56 MPa (90 hari). Campuran rekabentuk ini telah membuktikan kebolehkerjaan yang baik, ketumpatan yang baik, serta menghasilkan fasa semikristal dan kepadatan sebatian geopolimer matrik telah dibuktikan melalui analisis morfologi yang juga sejajar dengan nilai keputusan kekuatan. Penambahbaikan berterusan pes geopolimer berasaskan kaolin sejuru dengan masa menunjukkan potensi penyediaan salutan geopolimer bersaskan kaolin. Berdasarkan pengetahuan terkini, tiada percubaan telah dibuat sebelum ini untuk menghasilkan salutan geopolimer kaolin untuk applikasi kayu. Justeru, menjadikan kerja penyelidikan ini novel. Pes geopolimer kemudiannya disalut pata substrat untuk mengkaji kekuatan kelekatan. Kekuatan ini dikaji melalui kekuatan perekat, kekuatan lenturan dan kajian antara lapisan imej mikro. Kaolin geopolimer salutan optimum telah Berjaya merekat pada substrat kayu pembinaan dan telah memberi kekuatan paling tinggi sebanyak 4.3 MPa (7 hari), 4.9 MPa (28 hari) dan 5.96 MPa (90 hari).Kekauatan awal mekanikal kaolin geopolimer salutan terhadap kayu pemibinaan berbeza dengan kekuatan mekanikal yang dipamerkan oleh pes kaolin geopolimer berasaskan kadar penyerapan lembapan terhadap substrat disokong dengan lapisan imej mikro. Kekuatan kekerasan telah dipersetujui dengan kekuatan lenturan kaolin geopolimer salutan pada kayu pembinaan dan analisi fasa dari pes kaolin geopolimer. Titik puncak zeolite telah menyumbang kepada peningkatan kekuatan awal tetapi telah menggangu struktur geopolimer dari masa ke semasa yang membawa kepada penurun kekuatan pada usia sample. Geopolimer salutan dalam tempoh masa yang berbeza terbukti penting untuk penggunaan jangka masa panjang. Secara keseluruhannya, kajian ini secara jelasnya merperlihatkan rekabentuk dan kebolehgunaan kaolin sebagai bahan geopolimer salutan.

PROCESSING AND PROPERTIES OF KAOLIN BASED GEOPOLYMER COATING FOR LUMBER WOOD APPLICATIONS

ABSTRACT

The use of ordinary Portland cement is evident for centuries now especially in civil industries. Lacking of greener option with equal or enhanced properties forced consumers to ignore the shortcomings of ordinary Portland cement. Investigation of kaolin based geopolymer coating was aimed to not only increase the productivity and applications of geopolymer but also to potentially replace the use of ordinary Portland cement in terms of coating technology. Initial aim for this study was to prepare an optimum geopolymer coating paste made up of kaolin, as the aluminosilicate source and alkaline activator solution. The optimum mix design was mainly judged by its mechanical strength, followed by physical, phase analysis and scanning electron microscopy micrographs as crucial parameters of sodium chloride concentration, solids-to-liquid (S/L) ratio and alkaline activator ratio was studied. Kaolin, the geopolymer source material was characterized by using particle size analysis, phase, chemical composition, and scanning electron microscopy testing. Kaolin geopolymer paste with 8 M sodium hydroxide molarity, solids-to-liquid (S/L) ratio of 0.9 and alkaline activator ratio of 0.40 cured at 70 °C for 24 hours, gave highest strength values of 2.4 MPa (7 days), 2.98 MPa (28 days) and 4.56 MPa (90 days). This mix design also proven to have good workability, density, semi crystalline phase, and homogeneous compacted geopolymer matrix through morphology micrographs, in agreement to strength values. Continues improvement of kaolin geopolymer paste over time showed promising potentials towards preparation of kaolin based geopolymer coating. To the best of our knowledge, no attempts have been made previously to produce kaolin based geopolymer coating for lumber wood application, thus making it a novel work. The geopolymer paste were then coated on most unlikely substrate to investigate the extent of its bonding capabilities. This was evidently studied through bonding, physical, mechanical and morphological results. Optimum kaolin geopolymer coating successfully adhered to lumber wood substrate and provided high strength value of 4.3 MPa (7 days), 4.9 MPa (28 days) and 5.96 MPa (90 days). Early mechanical strength of kaolin geopolymer coated lumber wood differs from mechanical strength exhibited by kaolin geopolymer paste due to moisture absorption into substrate as supported by interfacial layer micro images. Hardness value was in agreement with flexural strength of kaolin geopolymer coated lumber wood and phase analysis of kaolin geopolymer paste. Zeolite peak contributes to high early strength development but disrupts geopolymer structure over time that leads to drop in strength upon sample age. Investigation of sample over different time period is proven to be important for long term usage of geopolymer coating. Ultimately, this study clearly demonstrated the processing and feasibility of kaolin geopolymer coating material.

CHAPTER 1: INTRODUCTION

1.1 Research background

Coating technology are used in a form or another since the very beginning of civilization. Starting from Chinese artifacts or the Greek statues, coating had been used for decorative and functional purposes (Rajdev, Yadav & Sakale, 2013). Functional coating is when applied on a substrate to improve or change the original properties of the substrate in terms of adhesion, wettability, corrosion resistance, or wear resistance. With the current developments, most engineered products are coated to be protected from usage wear, reduce maintenance cost and harmful environments (Diamanti, Brenna, Bolzoni, Berra, Pastore & Ormelles, 2013).

Coating can be classified as solid, liquid or gas; metallic or non-metallic; organic or inorganic. Organic coatings include paints, resins, lacquers and varnishes. Inorganic coatings includes cementitous, geopolymer, porcelain enamels, glass linings and metallic coatings. Recently, inorganic coating is argued as a better alternative as compared to organic coating due to its harmful processing nature and exhibit lower resistance properties which also limits their end application (Kishan & Radhakrishna, 2013)

Ordinary Portland Cement (OPC) coating, an existing cementitous inorganic coating faced multiple environmental issues and exhibits mechanical properties that is not suitable for long term end usage (Muttashar et al., 2014). The use of OPC generates carbon dioxide (CO_2) through calcinations of raw materials and fuel consumption. The manufacturing of OPC and the combustion of fossil fuels involved the process of de-carbonate of limestone in the furnace which brought to the production of 1 ton CO_2 in making of 1 ton of OPC (Lellan et al., 2011). Besides, the production of the OPC has increased the emission of greenhouse gas which is estimated around 1.35 billion tonnes per year and it is expected to be increase up to 3,500 million tonnes by the year 2019 (Malhotra, 2002). As per the carbon emission between nations 2013 report review, Malaysia stands at 8.1 ton per capita per year (Figure 1.1). It is also noted through the Malaysia's Biennial Update Report (BUR) 2011, the energy sector is the highest contributor to carbon emission at 76% and the subsectors are constructions, electricity, oil and gas manufacturing industries and transport (Jaafar, 2011).

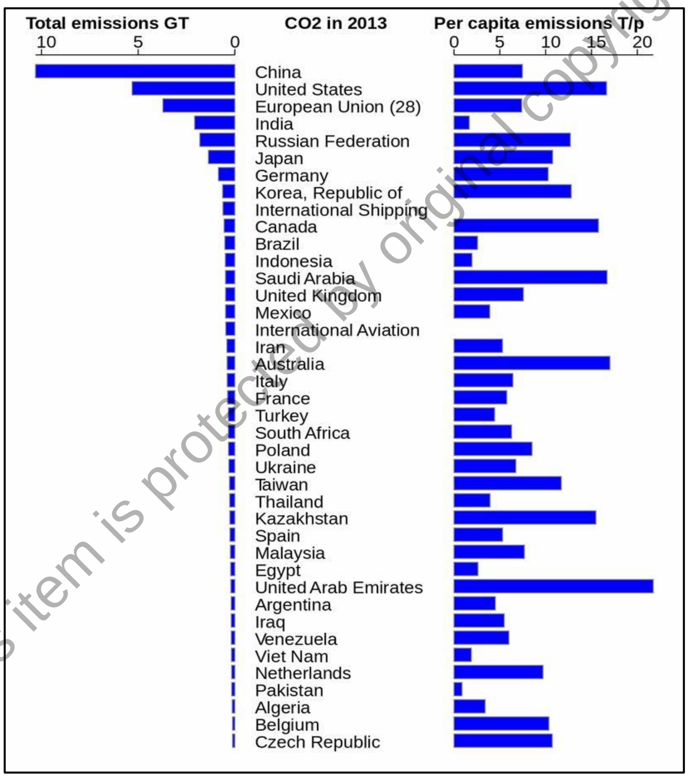

Figure 1.1: Carbon emission between nations on per capita basis (Jaafar, 2011).

Other common problems faced by OPC coating is an inability to bridge cracks and shrinkage that develop in substrates after application. OPC coating that is meant for water proofing also requires high degree of expertise for installation as OPC coating has fairly high water absorption rate as an inorganic coatings. OPC coating is also not recommended for application over wood or metal substrates due to its poor bonding properties towards these substrates (Diamanti et al., 2013).

Whereas, inorganic geopolymer based coating promises advantages that exhibits equivalent or enhanced physical, mechanical, bonding properties, and environmental friendly (Duxson, Mallicoat, Lukey, Kriven & Deventer, 2007). Therefore, geopolymer as inorganic polymer or alkali activated binder (Davidovits, 1994b) has expanded international interests as coating material. The term 'geopolymer' is always misunderstood as a type of polymer. However, properties of geopolymers completely differs from polymers except for the fact that it has a monomer unit.

Geopolymerization is expected to be one of the most in demand technology that will be essential in multiple fields such as aerospace, foundry, construction, building, automobile, medical and coating ((Provis & Deventer, 2005, Duxson et al., 2007; Hajimohammadi, Provis & Deventer, 2008). Geopolymer synthesis uses aluminosilicate source raw material, which is an abundant resource on earth. Geopolymers have special features such as fast setting, long durability, chemical, fire resistant and good volume stability as these materials have lower shrinkage ability and resistance to volume change. The most attractive point of geopolymer technology is that it does not emit any greenhouse gases and its completely environmental friendly (Duxson et al, 2007). Geopolymers are said to be greener construction materials especially as a replacement for concrete and cement.

Plenty of research works has been done lately on geopolymers as a green brick, concrete, mortar or even aggregates (Chindaprasirt, Chareerat & Sirivivatnanon, 2007; Aguilar, Diaz & Garcia, 2010). Nonetheless, countable attempts have been made to study on geopolymers as coating or repair material. This is because when geopolymers are investigated as a coating material, it is much more critical to obtain an optimum formulation as coating applications requires repeatable mechanical, bonding and physical properties as an end product. As evident by literature, most of the attempts and works on geopolymer coating is mainly on fly ash based geopolymer coating (Liyana, Bakri, Kamarudin, Ruzaidi, Azura, 2014; Khan, Azizli, Sufian Man, 2015; Norkhairunnisa & Fariz, 2015).

Commonly known geopolymer source materials are fly ash, metakaolin, blast furnace slag, kaolin and white clay. Fly ash, also known as pulverised fly ash is a by-product of coal combustion process. Whereas kaolin are materials that are rich in kaolinite. Metakaolin, also known as sintered kaolin is a cured form of clay mineral kaolinite. Among these three famous geopolymer raw materials, fly ash had been widely studied as repair or coating material as compared to kaolin and metakaolin (Liyana et. al., 2004). This work chose kaolin as a raw material instead of fly ash or metakaolin to be used as geopolymer repair material. This is because as a by-product, chemical composition of fly ash differs almost in each batch of collection which makes it not practical to obtain a repeatable end product worldwide. Whereas, for metakaolin, it is produced as raw instead of natural occurrence for industrial usage. Thus, makes it an expensive source material that also involves high thermal curing process which is not a green option (Palomo, Alonso, Jimenez, Sobrados & Sanz, 2004). Kaolin is arguably low reactive compared to fly ash and metakaolin but to our benefit, this characteristic allows us to have a more detailed understanding that occurs pre and post desired geopolymerisation (Wiyonoa, Antonia & Hardjito, 2015). Hitherto, kaolin as geopolymer raw material, leads to a more stable formulation establishment to obtain an ideal geopolymer based coating repair material.

Kaolin, soft earthy fine powder that is bright white in colour, makes it convenient to be amended as per desired beneficial industrial application. In general, kaolin also has low shrink-swell capacity and a low cation exchange capacity (Khan et al, 2015). In terms of current end application of kaolin is majorly in paper industry as a coating layer that enhances appearance, brightness, glossiness, smoothness and printability of papers. Kaolin in paper also used as filler to reduce cost and use of tree base materials. Kaolin is also part of pioneer materials used in China porcelains and tableware. Other uncommon usage of kaolin is in cable insulation, fertilizers, cosmetics, and paint industries (Nkoumbou, Njoyo, Grosbois, Njopwouo, Yvon & Martin, 2009). Important factors that are required in coating or repair material will be the durability, water absorption percentage, compression and shearing properties. Adhesiveness and bond strength is also the crucial demeanour in a coating material as it determines the possibility for this application purpose (Khan et al., 2015).

Kaolin based geopolymer coating possess excellent possibilities as bonding material especially in terms of bonding with concretes. However, if kaolin based geopolymer coating that is able to adhere with organic material subsrates such as wood or polymer is establish, it will rather be a breakthrough in cementitious coating as well as geopolymer technology.

Another important factor in geopolymerization process is its alkaline activator solutions. Most commonly used alkaline activator solution is potassium hydroxide (KOH) and sodium hydroxide (NaOH) with sodium silicate (Na_2SiO_3) solutions. In our work, we have chosen to use NaOH and Na_2SiO_3 solutions, as it is a more economical option (Naganathan, Razak & Hamid 2012). Combination of Na_2SiO_3 and NaOH solution is proven by previous works to improve the reaction between source materials and alkaline activators. Combination of these two alkaline boost the reaction rate as compared to usage of single alkaline hydroxides (Davidovits, 2002). NaOH solution acts a crucial provider of Na^+ ions and together with Na_2SiO_3, it becomes the alkaline activator solution needed for geopolymerisation. In my research work, kaolin is chosen as the aluminosilicate source material with alkaline activator solution of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3).

1.2 Problem Statement

Existing cementitious coating or repair material is mainly ordinary Portland cement (OPC) based for almost all available cementitious coating. The flexibility of OPC for onsite casting or prefabrication made most ignore the danger of OPC itself (Rajdev et al., 2013). Apart from contributing to CO_2 emission, its harmful and hazardous materials used in the pre and post production of this inorganic coating might also volatilize into the atmosphere. Producing one ton of OPC emits nearly one ton of CO_2 into the atmosphere (Lellan et al., 2011). As a consequence, partial or full replacement of OPC from the cementitous coating industries will help overcome this detrimental environmental impact.

Existing inorganic cementitous coating also has issues as final product that requires settlement. An alternative coating material that does not suffer from shrinkage, high porosity, poor adhesiveness and durability over time is crucially required (Diamanti et al., 2013). Common issues faced in existing inorganic coatings are lack of stability of coating layer that leads to deterioration of mechanical properties over time. Cementitous coating final applications demands reliable adhesive strength and excellent mechanical properties that is not influenced by sample age.Thus finding a greener alternative that can present equivalent or enhanced performance towards traditional cementitious coating in several main applications becomes a necessity (Gartner, 2004).

Considering all above issues, geopolymer is a popular alternative since the last three decades to work on replacing OPC based applications. As coating material, geopolymer coating would provide corrosion resistance, protect structural integrity, and