HYDRATION PERFORMANCE OF CEMENT-BONDED WOOD COMPOSITES: COMPATIBILITY ASSESSMENT OF SIX PIONEER FOREST SPECIES

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ABSTRACT. *The presence of inhibitory substances such as starches, sugars, hemicelluloses and extractives in wood are known to affect and influence cement curing / setting time. The hydration heat measurement gives preliminary assessment in understanding hydration behavior of the wood-cement-water mixtures. The objective of this study was to assess the compatibility of six pioneer forest species with ordinary Portland cement (OPC). At the same time, analysis on starch and sugar content of each wood species was also conducted. Wood meal samples of Neolamarkia cadamba (kelempayan), Antidesma spp. (langian), Artocarpus spp. (terap), Macaranga gigantea (mahang gajah), Pterocymbium javanicum (melembu) and Sapium baccatum (memeh) were prepared and blended with OPC and water as per standard mixing method practiced by Wood Composite Branch, Forest Research Institute Malaysia (FRIM). Apart from that, the compatibility of the wood-cement mixtures with two different accelerators ($CaCl_2$ and $MgCl_2$) at dosage levels of 1.50%, 2.50% and 3.50% based on cement weight was also evaluated. The compatibility of the species studied was estimated from the hydration rate of the wood-cement-water mixture, corresponding to the maximum temperature and time. From the testing with or without accelerators, mahang (79.6ºC; 2.6 hours) was recorded as the most compatible species for manufacturing cement bio-based composite products. The least suitable species under natural condition were kelempayan, memeh and melembu. It was found that with addition of the accelerators, the compatibility of these species had much improved. However, even though the compatibility of the woods had improved, the hydration behaviour of the woods was affected based on the type and dosage level of the accelerators. For example; kelempayan, $MgCl_2$ @ 1.5%: 67ºC; 9.8 hours and $CaCl_2$ @ 1.5%: 71.3ºC; 5.3 hours. It was also observed that the treatment level at 3.5% accelerator was not improving the hydration rate compared with 2.5%. The results imply that the six species evaluated could be processed for the production of cement-bonded panels and to accelerate as well as improving the bonding process, accelerators at appropriate dosage could be added. For sugar and starch analysis, it was found that Autocarpus spp. the lowest sugar content, i.e. 0.01% and no starch presence was recorded for Macaranga gigantea.*

KEYWORDS. Compatibility, accelerators, cement-bonded particleboard, sugar starch analysis

INTRODUCTION

Some of the materials used to manufacture original Portland cement are limestone, shell, chalk or marl, combined with shale, clay, clat or furnace slag, silica sand and iron ore. Lime and silica made up approximately 85% of the mass (Anon, 1988). The name of this cement was derived from the name of its basic ingredient, Portland stone. Portland stone was first quarried in the early 19^{th} century.

The stone then was modified to Portland cement by a scientist, Joseph Aspdin in 1822, by using the same method that was usually used for making Roman cement (Tatum, 2009). The original formulation of making Portland cement was further refined by Aspdin's son, William in 1843 when he moved to Germany. His modified Portland cement was widely utilised as building materials in Germany, 30 years after it was introduced (Tatum, 2009). Since then, Portland cement (OPC) became one of the most common type of cements used around the world until now. This material is very popular to be used as a basic ingredient for concrete, mortar, stucco and grout as it cheap and available for utilisation (Anon, 2009). According to the Bureau of Indian Standards (BIS), there are three different grades of ordinary Portland cement, i.e. 33, 43 and 53 grade. The grade is classified based on the compressive strength of cement-sand mortar cube of face area 50 cm^2 composed of 1 part of cement to 3 parts of standard sand by weight with a water-cement ratio arrived at by a specific procedure. The grade number is referring to the minimum compressive strength of cement sand mortar in N/mm^2 at 28 days (ACC, 2009).

Pioneer forest species are currently considered as less-utilized species that normally grow wild in the gaps of selective felling, wind damaged areas, by the logging roads or thriving in abandoned shifting cultivation plots. Some of these potential pioneer forest species outlined by Yunus (2002), Cheah (1995) and Laurila (1995) are shown in Table 1:

Table 1. List of potential pioneer forest species for conversion to panel products.

Scientific name	Local name
Anthocephalus chinensis	Kelempayan
Macaranga gigantea	Mahang gajah
Macaranga Hosei	Mahang
Macaranga Hypoleuca	Mahang puteh
Macaranga triloba	Mahang merah
Acacia spp.	Akasia
Mallotus spp.	Balek angin
Paraserianthes falcataria	Batai
Octomeles sumatrana	Binuang
Bombax valetonii	Kekabu hutan
Trema spp.	Mengkirai
Mezzetia leptopoda	Mempisang
Pterocymbium javanicum	Mata lembu
Ficus spp.	Ara, A. Kelepong
Vitex spp.	Leban
Sapium baccatum	Ludai
Alstonia spp.	Pulai
Endospermum malaccense	Sesendok
Duabunga grandiflora	Berembang bukit
Maesa spp.	Gambir, G. hutan
Gironniera spp.	Hampas tebu / Kasap
Breynia spp.	Hujan panas, Meroyan
Eleocarpus spp.	Mendong
Memecylon spp.	Nipis kulit
Ptemandra spp.	Sial menaung
Randia scortechinii	Tinjau belukar
Glochidion spp.	Ubah
Schima wallichii	Gegatal
Sapium baccatum	Ludai
Antidesma spp	“Langian”

Source: Yunus (2002), Cheah (1995) and Laurila (1995)

" " : These species were recommended by a wood panel manufacturer (local name)

Earlier studies indicate that majority of the species are with low density thus could be converted into panel products bonded with synthetic resin adhesives or ordinary Portland cement and could be used in non-load bearing applications. In manufacturing process, wood fibre and the binder (OPC) must be compatible to establish a strong bonding between those mediums to form cement-bonded boards with superior strength performance. The term compatibility, when applied in the research area of wood cement composites, refers to the degree of cement setting after mixing with water and with a given wood in a fragmented form (Jorge *et al.* 2004).

One of the key issues in determining the compatibility of wood species for manufacturing wood-cement-bonded composites is the effect that the wood gave to the hydration (setting) performance of the ordinary Portland cement (OPC). Normally, when ordinary Portland cement is mixed with water, the result is viscous but still fluid mixture called cement paste. The mixture is able to stay between 2 to 4 hours before it is gradually cured which the process is also known as hydration (Jennings *et al.*, 2002). Based on the situation, logically, the Portland cement is cured due to drying of the mixture. However, this claim is wrong as according to a scientist, John Smeaton, the curing of Portland cement is caused by chemical reactions between water and calcium silicate which is readily available in the formulation of the cement (Jennings *et al.*, 2002). However, the hydration of the Portland cement would further be affected once it is mixed with wood for wood-based-cement-bonded composite. The presence of inhibitory substances such as starches, sugars, hemicelluloses and extractives may delay or accelerate the hydration of the Portland cement (Yi e*t al.*, 1999). Wood species that delay the hydration would be classified as not compatible with OPC. The compatibility property indicator is based on the maximum temperature reached at the minimum time. This level of suitability is expressed by the "hydration rate". There are three classifications on the degree of hydration rate (Yi, 2000): I) least inhibitory species the maximum temperature was >50ºC and the time was < 10 hour, II) the maximum temperature was >40ºC and the time was < 15 hour; and III) Highly inhibitory the maximum temperature was <40ºC and the time was >15 hour. If the wood recorded as highly inhibitory, this wood is considered as unsuitable for the manufacture of cement-bonded particleboard, as removal of the extractives from the wood is necessary prior to manufacturing.

Adding an accelerator into a mixture of cement-water could encourage the hydration process to occur earlier. This was confirmed by previous studies Xu *et al.*, 2005, where they add shotcrete accelerator into a mixture of OPC-water and found out that the accelerator caused early hydration. The hydration rate was measured based on the ion concentrations and the pH value of the mixture. It was investigated that, the concentration of Ca^{2+} ions (a component of the accelerator) influenced the hydration rate in the liquid phase of the mixture. However, that study was conducted without wood as a part of the mixture proportion. Therefore, the objectives of this study are: 1) to determine the hydration rate of six secondary forest species; 2) to classify the selected secondary forest species in term of their compatibilities with OPC; and 3) to determine the influences of accelerators to mixture of wood-cement-water.

MATERIALS AND METHODS

Compatibility test

The compatibility test was done in order to determine the effects of the selected pioneer forest species to the hydration rate of OPC either with the addition of accelerators or not. This test is essential as it helps in selecting the best secondary forest species (out of the six selected pioneer forest species) that are compatible to be mixed with OPC prior to the fabrication of wood-cement-bonded particleboard. This method was implemented from Wood Composite Branch of FRIM and was employed by senior researchers of that branch.

The OPC was obtained from NS Cement from Negeri Sembilan. The six secondary forest species from Gua Musang, Kelantan. and sponsored by Duralite (M) Sdn. Bhd. From the billet, it passed through crusher to get particles. The particles were prepared into powder form using wiiley mill. The compatibility test was conducted at the Wood Composite Workshop of Forest Research Institute Malaysia (FRIM). The machine used for the test was hydration test kit. The OPC mixture for the test was divided into three different mixtures: 1) OPC neat (control sample); 2) OPC mixed with wood meal and water; and 3) OPC mixed separately with wood meal, water and accelerators ($MgCl_2$ or $CaCl_2$) at three different concentrations (1.5%, 2.5% and 3.5%).

The mixture of OPC-wood meal-water was prepared with standard ratio: 2.5:0.7:1. The components were mixed evenly about two minutes and then the mixture was placed in a plastic bag which was then stored in a thermoflask. A thermocouple was soaked into the mixture which was placed in a thermoflask covered with a lid and the thermocouple was connected to the hydration test machine using Type J wire (Figure 1). The temperature at every 5 second was recorded automatically by the machine as the mixture was left overnight.

Figure 1. Hydration test

Same procedures were applied to the OPC-wood wool-water-accelerators mixture, with the intention of investigating the accelerator effects to the hydration of OPC-wood wool mixture at three different concentrations.

Starch and sugar analysis

The method of analysing the starch content of the pioneer forest species was implemented from Humphrey (Humphrey and Kelly, 1961) and the sugar content was analysed using method of Simatupang (Simatupang, 1979). The intention of this experiment was to investigate the starch and sugar contents of each selected pioneer forest species. The analysis of starch was conducted by mixing 5g wood meals of each species with 4.7 ml $HClO_4$. Then the wood meals-HClO4 mixture was left to react for 15 minutes before the mixture was diluted to 100 ml with distilled water. The diluted mixture was then spin using Hettich Universal at speed of 2000 rpm for 10 minutes to form a mixture called aliquot. Five ml of the aliquot was taken and mixed with 2-3 drops of phenolphtalein and 2N NaOH. As the NaOH was added, the colour of the aliquot changed to pink. Two N acetic acid was added into the mixture, which made the aliquot, became colourless. Further amount of acetic acid (2.5 ml) was added with 0.5 ml KI (at 10% concentration) and 5 ml KLO_3 (0.01 N) into the aliquot. The mixture once again was left to react for 15 minutes before it was diluted again to 100 ml solution and analysed using UV spectrophotometer at 650nm wave.

As for the sugar content analysis, the wood meals were first mixed with 5 ml MeOH at 75% concentration in a volumetric flask. And the volumetric flask was placed in a water bath and was shaken for 24 hours. Ten ml of MeOH at 80% concentration was added into the mixture to form aliquot. The aliquot was then filtered and dried at a temperature between 40ºC – 50ºC before it was further mixed with 3ml distilled water to dilute it to 100ml. The diluted aliquot was stored in a bottle and was analysed using High Performance Liquid Cromatography (HPLC).

RESULTS AND DISCUSSION

Compatibility Test

Hydration temperature and time

The maximum hydration temperatures achieved by each sample against the OPC-neat is demonstrated in Figure 2. The OPC neat was found to achieve the maximum temperature (78.8ºC) within 5.7 hours. The pioneer species was found to delay the hydration rate of the OPC by 20 to 180% and lowered the hydration temperature by 5.21% to 52.19% compared to the data for the OPC-neat. The OPC-mahang mixture was found to achieve the maximum temperature 78.7ºC within 6 hours followed with the OPC-langian (66.7 ºC within 8 hours) and the OPC-terap (61.3ºC within 8 hours). The less active mixtures were the OPC-kelampayan, OPC-memeh and OPC-melembu with the maximum temperatures and the times to achieve those temperatures for the three less active mixtures were 50.33ºC within 14 hours, 46.67ºC within 13 hours and 37.7ºC within 14 hours respectively.

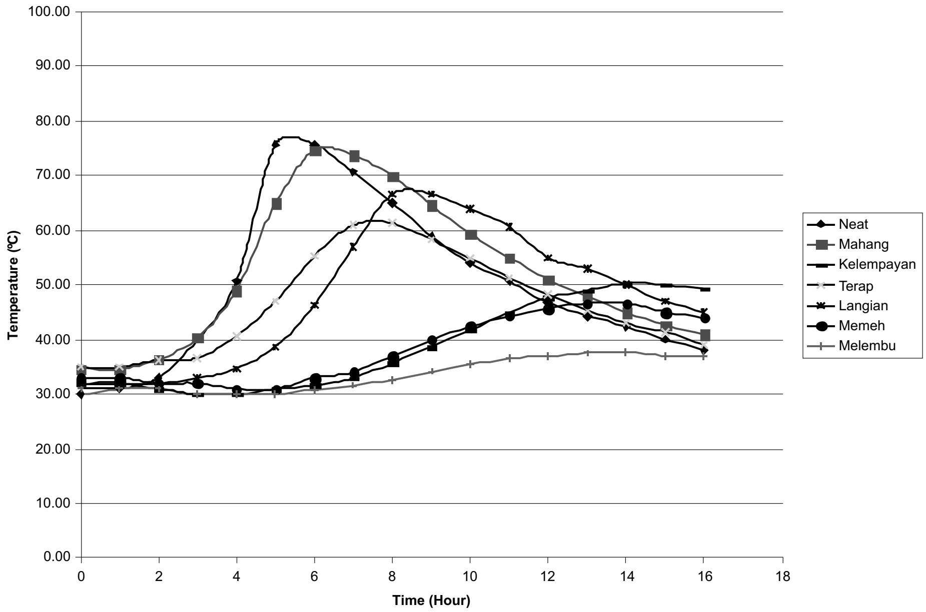

Figure 2. Hydration temperature of OPC with six pioneer forest species

Effects of accelerators to the hydration of the OPC-pioneer species mixtures

Regarding relationship of the accelerator types and concentrations to the hydration rates of the OPC, it was found that each accelerator at the three different concentrations affected the hydration of the OPC. The hydration rates of the OPC-accelerator-pioneer species mixtures are presented in Figure 3 to 8. Based on the figures, $CaCl_2$ acceleration was superior to $MgCl_2$. The hydration rate of the OPC-Mahang was increased higher than that of the OPC neat when $CaCl_2$ at 1.5% concentration was added into the mixture, whereas $MgCl_2$ at 2.5% concentration was needed to achieve similar result. Amazingly, each OPC-pioneer species mixture achieved maximum temperature faster than the OPC neat when $CaCl_2$ at 3.5% concentration was added (Figure 5).

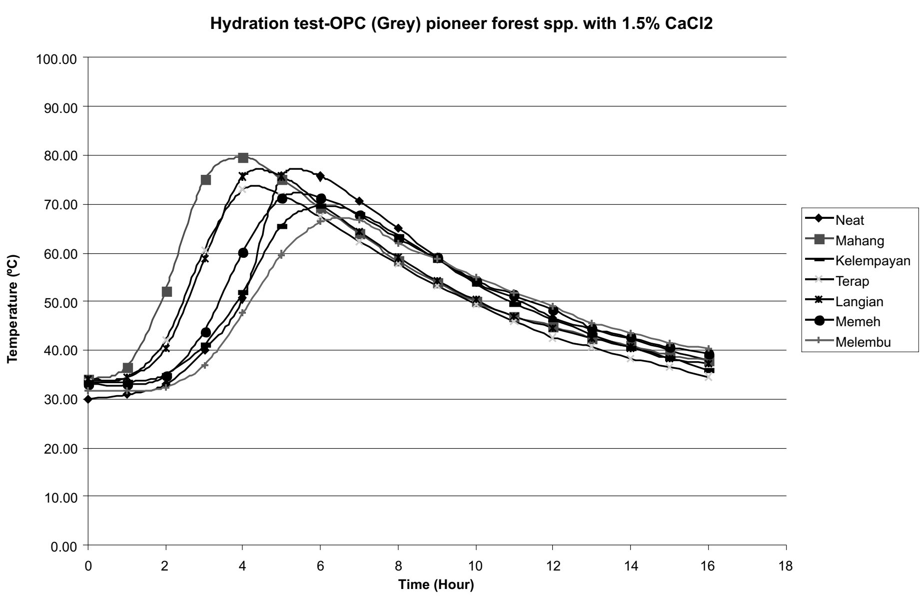

Figure 3. The hydration rates of the OPC-pioneer species at 1.5% concentration of $CaCl_2$

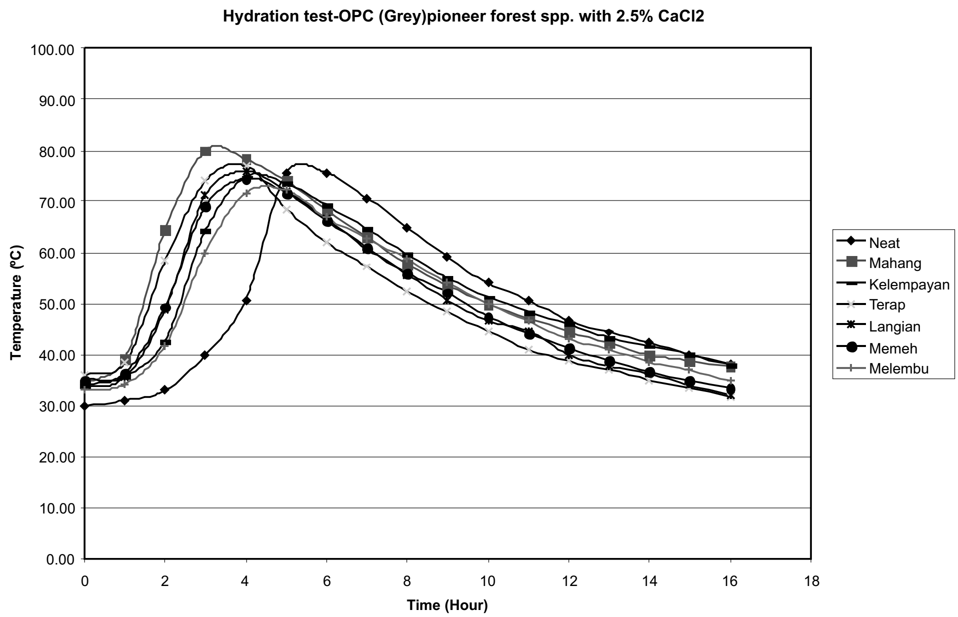

Figure 4. The hydration rates of the OPC-pioneer species at 2.5% concentration of $CaCl_2$

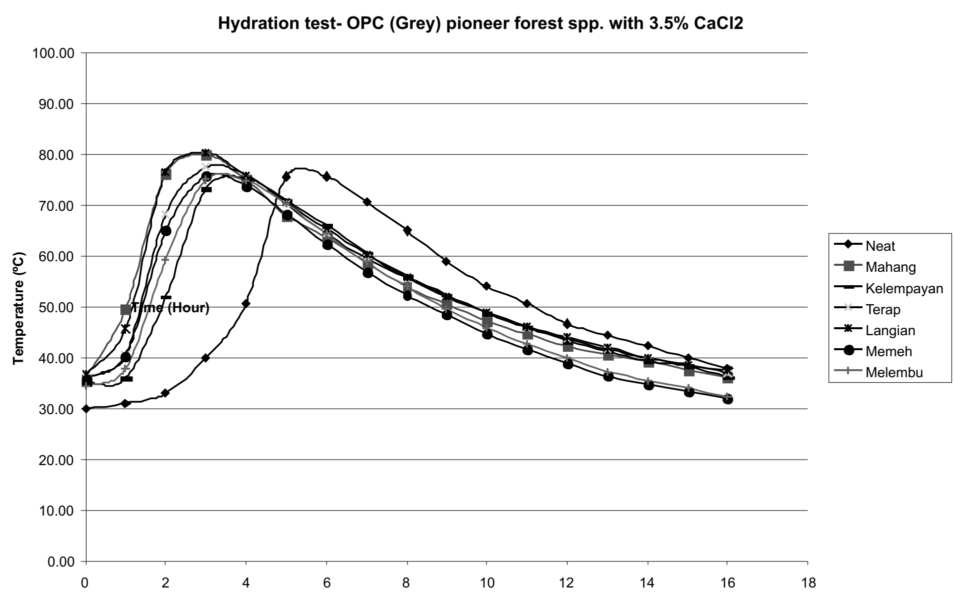

Figure 5. The hydration rates of the OPC-pioneer species at 3.5% concentration of $CaCl_2$

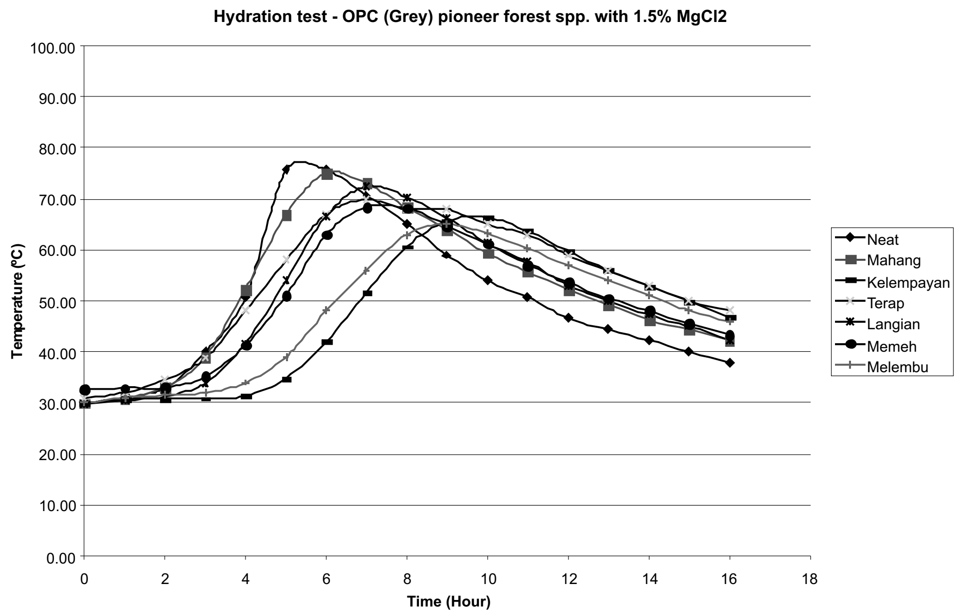

Figure 6. The hydration rates of the OPC-pioneer species at 1.5% concentration of $MgCl_2$

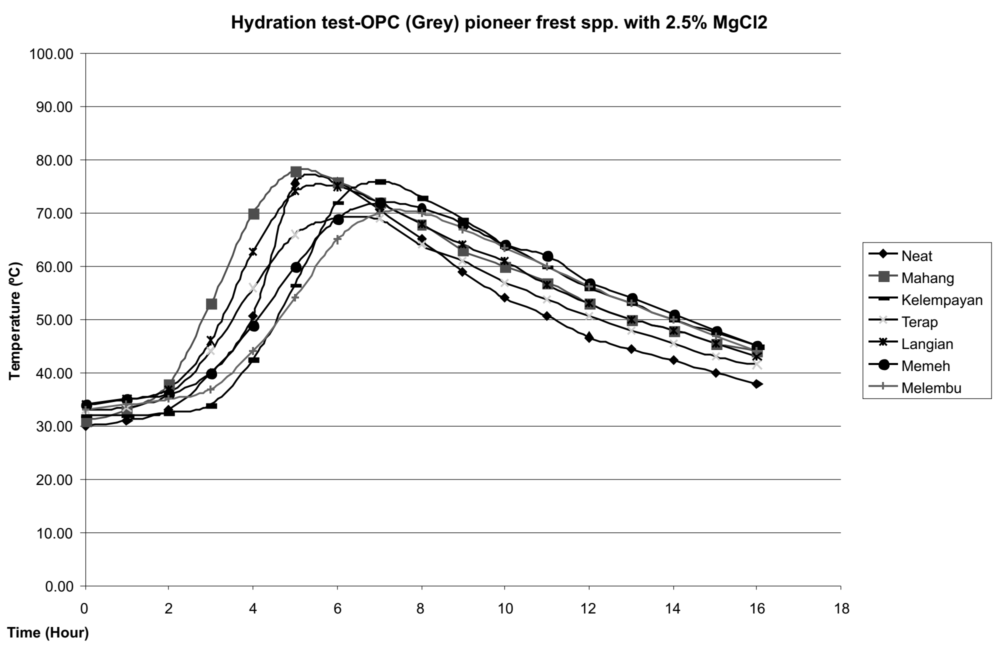

Figure 7. The hydration rates of the OPC-pioneer species at 2.5% concentration of $MgCl_2$

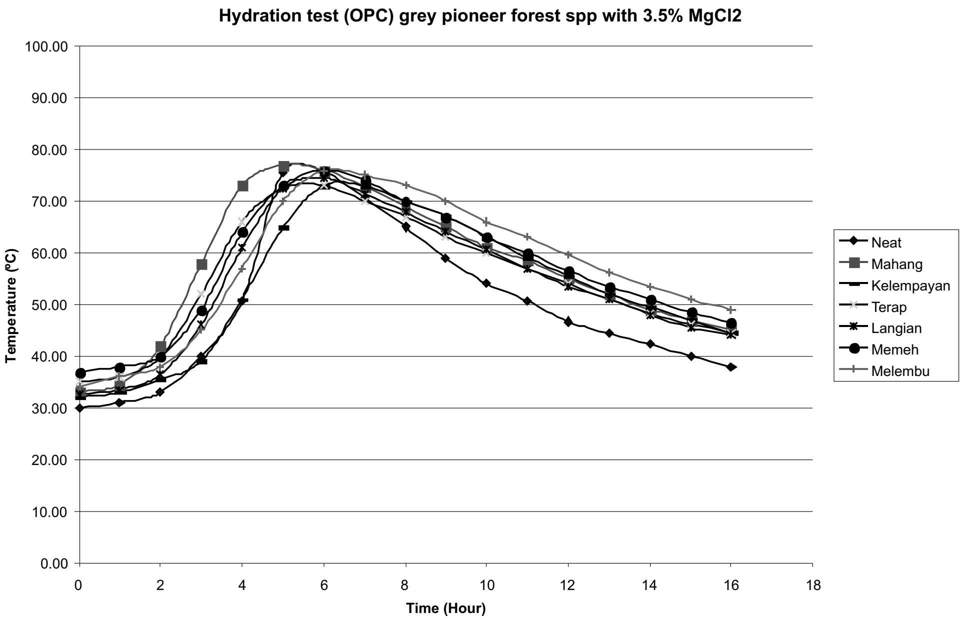

Figure 8. The hydration rates of the OPC-pioneer species at 3.5% concentration of $MgCl_2$

The addition of $CaCl_2$ and $MgCl_2$ increased the temperature of the mixtures and encouraged the activation of cement hydration which later gives a more dense paste structure with smaller pores compared with the OPC neat (Maltese *et al.*, 2007). Therefore, as the concentration of $CACl_2$ and $MgCl_2$ were increased, the activation became more rapid and dense paste structure could be achieved earlier. The accelerating performances can be evaluated by determining the final setting time of cement pastes admixed with the alkali-free accelerator: the shorter the final setting time, the more efficient the accelerator (Maltese *et al.*, 2007). During the first hydration stage, calcium and sulphate ions, supplied by the setting regulator, react with aluminium which normally present in the cement formulations and water to give an amorphous layer of aluminates. The aluminates layer gradually slows down the hydration to dormant stage (Scrievener *et al.*, 1984). The accelerators are usually added after the formation of the aluminates layer. It was found that the lower the instantaneous solubility rate of the setting regulator the more efficient the accelerator. The reason for this behaviour could be connected with the different morphology of ettringite formed during the first minute of the reaction between the hydrating cement and the alkali-free accelerator. Same result was recorded by Maltese *et al.*, (2007)

Sugar and Starch Analysis

The intention of this experiment was to investigate the starch and sugar contents of each selected pioneer forest species. Table 2 represents the total sugar and starch available in the selected pioneer species.

Table 2. Total sugar and starch percentage of the six pioneer species

Species	Total sugar (%)	Total starch (%)
Memeh	0.05	0.11
Langian	0.04	0.02
Kelempayan	0.70	0.02
Melembu	0.53	0.03
Terap	0.07	0.01
Mahang	*	0.03

*unavailable

Based on Table 2, the total sugar percentages for memeh, langian, kelempayan, melembu and terap were 0.05, 0.04, 0.70, 0.53 and 0.07% respectively. No percentage was recorded for mahang as the the sugar content was so low and undetectable. The total starch percentage was 0.11, 0.02, 0.02, 0.03, 0.01 and 0.03% respectively, which also indicated that the lowest starch content was recorded from terap. The low content of sugar and starch in mahang might be the cause of the highest hydration rate of the OPC-mahang compared to the other mixtures (Figure 2). It has been shown by earlier researchers that wood extractives (particularly wood sugars) even present at low percentage inhibit cement setting and thus loss of board strength. According to Yi *et al.* (2000), Simatupang *et al.* (1993), Azizul & Rahim (1989) sugars in wood significantly inhibit cement hydration (setting and hardening). Razali and Hamami (1993) and Milligan (1995) in the review of Growing and Utilization of *Acacia Mangium* cited the works of Tachi and co-workers (1988, 1989) that the extractive component in mangium called teracacidin in the heartwood could cause cement hardening inhibitory. Generally, hardwoods have a lower compatibility with cement than softwoods, partly due to the inhibitory properties of hydrolyzable hemicellulose and other extractives present in hardwoods (Yi *et al.* 2000). The hydration of the OPC mixed with the six pioneer species could be improved if the extractives are removed prior to the mixing stage.

CONCLUSIONS

The results obtained in this study have identified that the extractives available in the pioneer species wood affected the hydration rate of the OPC. In the study, the compatibility was determined by observing the maximum temperature achieved by the mixtures. The mixtures with temperatures more than 50°C were considered as compatible. Mahang showed the highest rating, since only small extractives were detected with and without enhancer. On the other hand, kelempayan was identified as the highly inhibitory species without accelerator. When accelerator was added, it gave good reaction and improved the setting time of the mixture. Based on the results, it was found that the species have the potential to be further researched on to convert into panel products such as cement-bonded particleboard.

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