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WOOD-CEMENT COMPATIBILITY REVIEW

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ABSTRACT

Wood-cement composites have been widely used for building construction in many countries. However, the wood-cement incompatibility limits the manufacture of this material for some wood species because of the wood extractives inhibiting the cement setting and hydration. This paper discussed the cause of incompatibility between wood and cement; investigated the influence factors of wood-cement compatibility including type of species, location, part of the tree, season during wood-cutting, wood/cement ratio, storage condition, type of cement and so on; and reviewed the current research situation of the assessment methods of wood-cement compatibility and the method of improving the wood-cement compatibility, involving the pre-treatment of wood, addition of cement curing accelerators, and CO_2 injection.

KEYWORDS: Wood-cement composites; compatibility; cause; influence factors; improving methods.

INTRODUCTION

Wood-cement composites are widely utilized in many countries for both interior and exterior applications because of their strength properties for building materials (e.g., siding, roofing, cladding, fencing and sub-flooring) and for acoustic properties such as in highway sound barriers (Moslemi 1999). The main problem is for producing wood-cement composites the incompatibility between cement and wood because some soluble chemicals of wood are found to hinder or stop the hydration of cement when they are attacked by the alkaline environment and diffuse into the cement paste, resulting in the lower mechanical strength of wood-cement composites compared with the neat cement (Zhengtian and Moslemi 1986; Thomas and Birchall 1983). The typical cement-hardening inhibitory components known up to now are divided into two groups. One is comprised of carbohydrates of sucrose in beech and arabinogalactan in larch and the other of phenolic compounds with a catechol unit of plicatic acid in western red cedar, teracacidin in *Acacia mangium*, and sequirin C in sugi (Yasuda et al. 2002).

Wood-cement incompatibility may limit the practical application of wood species in wood-cement composites. It has been found that the wood-cement compatibility varies within different wood species. Therefore, the level of compatibility should be evaluated to indentify whether the wood is suitable for producing wood-cement composites. At present, some assessment methods have been developed to classify wood of various species regarding its compatibility with cement based on hydration heat or temperature, strength, electrical conductivity and morphology.

To enhance the suitability of some wood species for manufacturing the wood-cement composite boards, it is common to add inorganic chemicals, known as accelerators, to accelerate the cure of cement or use pretreatments such as aqueous extraction to remove inhibitory substances from wood.

This paper has reviewed the current research situation of wood-cement compatibility, including the cause and improvement of incompatibility between wood and cement, the influence factors and assessment methods of wood-cement compatibility.

The cause of incompatibility between wood and cement

At present, although the precise mechanism of inhibition effect of wood on ordinary Portland Cement (OPC) hydration is yet to be fully understood, there are two possible mechanisms have been proposed. First is that the extractives composed of various organic compounds make complexes with the metal ions present in the cement solution. This decreases the concentration of Ca^{2+} ions in the cement and possibly disturbs the equilibrium of the solution, which delays the start of nucleation of $Ca(OH)_2$ and CSH gel (Miller and Moslemi 1991). Another suggested mechanism is that the organic compounds form a thin adsorption layer on the surface of cement grains and slow the hydration process (Gartner et al. 2002; Juenger and Jennings 2002).

Different components of wooden extractives cause the different inhibitory or retarding degree of cement hydration. The sugars and starches present in wood have been previously identified as the most critical compounds causing incompatibility between wood and cement, especially in softwoods (Bruere 1966). However, not all type of sugars has the same inhibitory effect. In hannoki (Japanese alder, *Alnus japonica* Steud.), glucose and sucrose are observed to be the main cement-hardening inhibitory components (Yasuda et al. 2002). Sucrose has greater retarding effect than glucose at the same concentration because the circular structure of sucrose causes stronger steric hindrance than the chain structure of glucose (Baoguo et al. 2005). The research conducted by Sandermann and Brendel (1956) showed some sugars, such as fructose, caused no essential effects on the cement hydration even at high concentrations of about 0.5 %, whereas, other sugars completely inhibited hydration at concentration of about 0.25 %. By contrast, some sugars, such as raffinose at concentrations up to 0.125 %, could even improve properties of cement. Miller et al. (1991) observed that small amount of hemicelluloses (0.1 %) could decrease significantly the curing strength of cement and had great influence on hydration properties of cement paste. They also assessed that pentoses (xylose, arabinose) had less effect on hydration of cement. Govin et al. (2005) found that water-soluble hemicelluloses were hydrolyzed and converted into carboxylic acid in alkaline cement paste and the alkaline degradation products were three times more efficient than wood extractives to inhibit the cement hydration. Boustingorry et al. (2005) investigated that the influence of separated extracts in poplar and forest pine on cement hydration and found that neither acetic acid nor the phenolic compounds were the principal retarding agents and the presence of trace elements such as tannins which were able to create complex ions with calcium may account for the cement hydration kinetic delays. Organic acids such as acetic tannic acid and other phenolics may not only inhibit cement hydration, they may also slowly attack and destroy the cement bond, resulting in a reduction in panel strength values and affecting the other panel properties (Blankenhorn et al. 1994). Tachi et al. (1988) found that the incompatibility between *A. mangium* and cement was due to the presence in heartwood of the flavonoid teracacidin which has a 7,8-dihydroxyl group in a leucoanthocyanidin structure.

The influence factors of wood-cement compatibility

Wood-cement compatibility is affected by several factors such as type of species, type of cement, part of the tree, season during wood-cutting, wood/cement ratio, storage condition and so on (Karade and Aggarwal 2005). Generally, lower amount of inhibitory extractives diffuse into the cement paste is beneficial for the wood-cement compatibility.

Wood species

Different wood species contain different kind and amount of wooden extractives, so they have different effect on cement hydration with some completely preventing cement setting. In general, hardwoods are generally less compatible than softwoods because of the presence of large amount of soluble xylan in hardwoods (Weatherwax and Tarkow 1964). The compatibility of wood species grown in different regions has also been assessed by worldwide researchers. Oyagade (1994) evaluated the compatibility with cement of 8 tropical woods and among these species danta was observed to be the least inhibitory while gmelina was the most potent in its effect on cement setting. Fan et al. (2012) also investigated the compatibility and hydration of OPC with 15 commercially available tropical wood species, and the results showed that overall ranking order of the wood–cement compatibility at 5 % by weight wood content was mouvingui > nkanang> ngollon > sapel > tali > padouk > bibolo > ayous > eyong > frake > bête > bilinga > doussie > iroko >moabi. Hofstrand et al. (1984) calculated the inhibitory indexes for 9 softwood species and 12 hardwood species and found that lodgepole pine had a high level of suitability. Semple et al. (2000) tested the eight temperate *Eucalyptus* species grown at two sites in south-eastern Australia and observed that compatibility was significantly affected by species, and *E. badgensis* and *E. smithii* had the highest compatibility. Regardless of the assessment method, lodgepole pine was observed to be the most compatible North American wood species with cement (Defo et al. 2004). Wei et al (2000) studied the effect of 38 wood species mainly grown in China on the exothermic reaction of OPC and divided these species into two groups. The 24 species included in the first group showed a moderating influence on the hydration reaction of cement and the other 14 species inhibited cement hydration completely.

Part of the wood

Part of the wood is another factor influencing the wood-cement compatibility. Heartwood of radiata pine was found to severely inhibition cement hydration, and the wood-cement boards made with heartwood had little structural integrity, whereas boards made from sapwood have been made industrially and commercialized (Semple and Evans 2000). Heartwood had higher solubility than sapwood suggesting greater amounts of substances that could inhibit cement setting (Cabangon et al. 2000)

Storage condition of wood

Wood-cement compatibility is also affected by wood storage condition. A number of studies have found that storage of logs outdoors for 4 to 20 weeks reduces the wood sugar content and increases the suitability of the wood for the manufacture of wood–cement composites. Schwarz and Simatupang (1984) reported a 75 % decrease in the sugar content of temperate wood species such as beech (*Fagus sylvatica* L.) when the logs were stored outdoors for three months. The authors also found that air drying for 16 weeks increased the suitability of birch (*Betula* sp.) logs for cement-bonded particleboard manufacture. Sundi et al. (1989) found that after four weeks of natural storage without bark, sugar and starch content of rubberwood fell below 0.5 and 7.0 % respectively, and a longer period up to 12 weeks required for a similar effect when stored with bark. Boards from fresh rubberwood failed to set completely. The optimum strength of cement-bonded particleboard produced from rubberwood stored without bark could be obtained between four to eight weeks of storage while the other condition required about 12 weeks. *Acacia mangium* in its natural state is highly incompatible with OPC. While post-harvest storage of it for 6–32 weeks improves its compatibility with OPC (Cabangon et al. 2000). Lee et al. (1987) showed that cold storage of wood slightly increased hydration temperature and shortened hydration time of white oak and sweetgum but did not have any beneficial effect on southern pine, southern red oak, yellow-poplar, and hickory.

Type of cement

Besides OPC, in recent years, magnesium oxychloride cement (MOC) is usually used to manufacture wood-cement composite products, including MOC-wood fiber board, straw-MOC thermal insulation wall materials, bamboo-MOC particleboard and so on (Zhou and Li 2012; Zhang et al. 2010; Wang et al. 2012). MOC cement has many properties superior to OPC such as lower carbon emission, higher fire resistance, higher abrasion resistance, higher temperature resistance, lower thermal conductivity, lower alkalinity, lower shrinkage and creep and better durability. Compared to the OPC, MOC cement presents a perfect match with wood, so MOC can be a substitute for OPC to manufacture wood-cement particleboard especially for the wood species that have high incompatibility with OPC (Huang and Ma 2010). For example, Mei et al. (2013) found that the presence of the external bamboo wall powder increased exothermic amount of MOC and the presence of various bamboo powders increased the flexural and compressive strength of the MOC.

Other factors

In addition, a faster reaction time and higher hydration temperature was observed at higher wood/cement ratios (Lee et al. 1987). Winter-cut sapwood was found to be have a higher compatibility than Spring-cut sapwood (Biblis and Lo 1968). It is found that when bark was present, the compatibility of *M. uncinata* increased compared with that without bark, in contrast, the bark of *E. loxophleba* strongly inhibited cement hydration, resulting from the chemical composition of bark varying greatly among wood species. Consequently, the higher proportion of bark in the younger (2.5-year-old) *E. loxophleba* trees reduced its compatibility with cement (Semple and Evans 2004).

Improvement of the compatibility between wood and cement

In order to improve the compatibility between wood and cement, several methods are employed including pre-treatment of wood, addition of cement set accelerators and injection of CO_2 gas. The elimination of cement hardening inhibition by these methods can be ascribed to either reducing the amount of inhibitory substances diffuse into cement paste or accelerating the cement hydration by external force to counteract the retarding effect of wood addition.

Pre-treatment of wood

Aqueous extraction is an effective method to remove soluble deleterious compounds before wood mixing with cement, and cold or hot water and NaOH solution are mostly used. In a lot of researches (Sutigno 2000; Ma and Wang 2012; Ye et al. 2002), the treatment of incompatible wood with cold water was verified to be less efficient than with hot water and the treatment of NaOH solution was most effective. Bamboo has a strong inhibitory effect on cement setting, making it unsuitable for wood-cement composites manufacture, while after aqueous extraction the incompatibility reduces greatly (Sulastiningsih et al. 2000). Fan et al. (2012) found that compatibility was significantly improved by prior extraction of the wood, with an overall ranking order of efficacy of treatments being $Ca(OH)_2$ > MeOH > H_2O > $CaCl_2$ although depending on the wood species. In addition, it was found that copper-chromium-arsenate (CCA) treatment more effectively increased wood-cement compatibility than hot water extraction, which resulted from the extensive reduction of water extractive contents and the change of available sugars after wood was treated by CCA preservative. However, there were small amounts of chrornium leached, and the addition of $FeSO_4$ and silica fume could reduce the leaching of chromium and Cr^{6+} in some cases (Qi 2001).

The coating of wood surface can isolate the wood from the cement paste and enhance the resistance to the alkali or water attack so improve the compatibility between wood and cement. Hou and Zhu (2010) modified the rice straw surface with polymer emulsions and found that the film-forming treatment could promote the hydration of straw cement mixture, thereby increase the mechanical strength of hardened cement paste compared with the original rice straw. In another research (Han et al. 2009), surface of rice hull was treated with the synthesized carboxymethylcellulose methyl methacrylate (CMC-g-PMMA) and the compatibility between treated rice hull and cement was improved due to the sealing effect of CMC-g-PMMA for inhibitory wood chemicals. Monreal et al. (2011) treated the beet pulp with linseed oil and the oil successfully covered on the beet pulp surface, resulting in the better resistance to water over time.

Soaking in Na_2SiO_3 can either remove part of the inhibitory substances of wood or prevent the action between inhibitory substances and cement by forming a layer on wood surface. However, improvement of the mechanical properties as well as migration of thickness by the use of Na_2SiO_3 is weak compared to water- or NaOH-extraction (Kavvouras 1987).

Olorunnisola and Adefisan (2008) found that peeling had negative effect on the setting time of the *Calamus* species but enhanced those of the *Lacosperma* species. Higher quantities of extractives (alkaloid, terpenes and saponins) in the unpeeled *Lacosperma* might have resulted in higher setting time than in the peeled *Lacosperma*-cement composite. This observation again suggested that cement inhibitors were concentrated at the periphery in the *Lacosperma* species.

Addition of cement curing accelerators

The addition of some chemicals that act as cement curing accelerators has usually the effect of improving the compatibility of a wood-cement compoites, and that addition can also be made after an extraction of the furnish to further improve the cement setting (Jorge et al. 2004).

A number of metal salts, such as $CaCl_2$, $FeCl_3$ and $Al_2(SO_4)_3$ can be used to accelerate cement setting, and $CaCl_2$ with low cost in particular has been successfully used to improve the strength properties of wood-cement composites (Ma and Wang 2012; Defo et al. 2004). The accelerating effect of 137 inorganic compounds addition on cement hydration mixed with *Acacia mangium* which comprised chlorides, sulphates, nitrates, acetates, oxides, carbonates and fluorides of the cations Al, Ba, Ca, Co, Cr, Cu, Fe, K, Li, Mg, Mn,Na, Ni, Pb, Sr and Zn were studied. The results showed that compounds with the ability to strongly accelerate cement hydration and form insoluble chelates with inhibitory heartwood tannins were the most effective at reducing the inhibitory effect of *A. mangium* heartwood on cement hydration. Most of the compounds were chlorides and nitrates, including $SnCl_4$, $AlCl_3$, $(NH_4)_2$ $Ce(NO_3)_6$ and $FeCl_3$ (Matoski et al. 2013). In some cases, combination of chemical accelerators can make better improvement effect, for example, combination use of Na_2SiO_3 and $CaCl_2$ was found to confer better strength to the rice straw-cement board than the chemical additives used singly (Fernandez and Taja-on 2000).

Some mineral additives added to a mixture of cement and wood improve its quality. According to Sellevold and Nilsen (1987), one of the most promising additives for the wood-cement boards industry is silica fume. The rapid growth of the silica fume use in cement-based materials is attributed to its beneficial effects on physical-mechanical properties of cement based composites, such as increased strength and decreased permeability. The replacement of cement with 10 % of silica fume eliminates the inhibitory effect of wood on cement setting (Del et al. 2007). Addition of opoca to cement mixtures decreases the harmful retarding effect of wood extractives. Because the specific surface of opoca is much bigger than of cement and the sorption ability is higher, adsorption of water-soluble wood materials first of all takes place on the surface of opoca and the concentration of extracts decreases (Vaickelionis and Vaickelioniene 2006). Alpar et al. (2012) added new additives, the montmorillonite nano particles and poly (diallyldimethylammonium chloride) (PDDA), to improve the wood-cement compatibility and the results showed that montmorillionite-PDDA additive combination gave a better results of strength of polar wood - cement composites than $MgCl_2$. Fly ash addition was observed to improve the mechanical properties of rice straw-MOC composites (Liu et al. 2010).

CO_2 injection

Another promising method to accelerate the setting of cement mixed with wood and improve the wood-cement compatibility is the use of gaseous or supercritical CO_2. CO_2 is neutralized by calcium silicate in cement, resulting in highly insoluble calcium carbonate. The carbonation reaction is confirmed to be a diffusion-controlled process. It occurs very quickly in the first two minutes of reaction. After that, its rate decreases drastically, due to the increasing difficulty of transporting reactants to reaction sites (Qi et al. 2010). Rapid carbonization might accelerate formation of the hydration products (e.g., calcium carbonate and calcium silicate) and the scanning electron microscopy (SEM) analysis showed that interaction of calcium silicates in cement with carbon dioxide resulted in the rapid hardening of wood-cement-water mixture (Hermawan 2002). Qi et al. (2006) found that significant strength development of wood cement composites containing 14 or 20 % fiber occurred after 1-3 min CO_2 injection, which would allow composites to be removed from the press in a few minutes rather than after several hours, which is common when composites harden by normal hydration processes. Soroushian et al. (2003) observed that a lower CO_2 concentration (25 %) yielded immediate flexural performance characteristics which were generally comparable to those obtained at 100 % CO_2 concentration. They hypothesized that the excess rate of reactions and rapid heat generation associated with 100 % CO_2 concentration, which could compromise any gains associated with the greater extent of reactions at higher CO_2 concentrations.

The supercritical fluid (SCF) technique might allow more rapid penetration and uniform CO_2 distribution, which cause more rapid curing of cement - hence the board strength properties (Hermawan et al. 2000). The optimum properties of the supercritical CO_2-cured wood-cement boards could be achieved earlier than those with gaseous CO_2 curing (Hermawan et al. 2001). However, it was considered that supercritical CO_2 treatment over a longer time span leaded to degradation of cement-bonded particleboard (Maail et al. 2011).

Soroushian et al. (2004) develpoed a process of CO_2 curing cement-bonded boards with agricultural residues. The curing process mainly involved the application of alternate vacuum and CO_2 on the top or bottom of boards pressed to a thickness of 10 mm before curing. They also found that the wood-cement boards with this CO_2 curing process had higher flexural strength and stiffness and better longevity and weathering resistance because calcium carbonate presence reduced the capillary porosity of wood cement mixture and enhanced the wood fiber matrix interfaces (Soroushian et al. 2012; 2013)

Assessment methods of wood-cement compatibility

Various attempts have been made to accurately assess the compatibility between wood and cement. The most commonly used method is the measurement of hydration temperature because of the simplicity of the test. The maximum temperature (T_{max}) of hydration and the time (t_{max}) required to reach the temperature were used directly to evaluate the index of compatibility between wood and cement. Sandermann and Kohler (1964) classified the compatibility as 'compatible' (T_{max} >60°C), 'moderately compatible' (50°C< T_{max} <60°C) and 'not compatible' (T_{max}<50°C) according the T_{max}. Wei et al. (2000) divided the compatibility of 24 wood species into three levels: least (T_{max} >50°C and t_{max}<10.2 h), intermediate (T_{max} >40°C and t_{max}<14.7 h), and highly (T_{max} <40°C or t_{max}>14.7 h) inhibitory species for wood-cement panels production based on the combination of T_{max} and t_{max}. They insisted that T_{max} and t_{max} could be considered the most important parameters and indicators for estimating the compatibility of given species. The inhibitory index (I) calculated from T_{max}, t_{max} and the maximum slope of the exothermic curve(S) for evaluating and classifying the compatibility of wood-cement-water mixtures wood species and cement was developed by Hofstrand et al. (1984). The effect of the inhibited cement setting can be classified into four grades according to the inhibitory index (I), that is, 'low inhibition'(I<10), 'moderate inhibition'(I=10-50), 'high inhibition'(I=50-100) and 'extreme inhibition (I>100) (Okino et al. 2005). However, Hachmi et al. (1990) found that these compatibility evaluation methods mentioned above lacked consistency in the classification of species. Therefore, they proposed there alternative indicators: C_T (weighted maximum temperature rate ratio), C_H (maximum heat rate ratio) and C_A (the ratio of the amount of heat released from a wood cement mixture in 3.5–24 h interval). It is recommend by them that C_A factor over the C_T and C_H factors, and three classes of compatibility based on C_A had been suggested: 'compatible', C_A>68 %; 'moderately compatible', 28 %<C_A<68 %; and 'not compatible', C_A<28 %.

The existing methods to qualify wood-cement compatibility are based on the $T_{max}$and t_{max}or the heat evolved in a specific period. However, Olorunnisola (2008) found that the assessment results of rattan-cement compatibility based on t_{max}, T_{max} and C_A respectively were inconsistent because addition of wood inclusions in cement effectively dilutes the system with respect to cement hydration, and maximum hydration temperature or the area under the hydration heat rate curve could not reflect the actual compatibility. He suggested that TR, which is the ratio of setting time of wood-cement composite to that of neat cement to evaluate the compatibility, was more effective and applicable to evaluate the compatibility. Karade et al. (2003) insisted that one of the limitations of T_{max} and t_{max} indicators was that they are effective only for the comparison of compatibility of different wood species when using the same wood:cement:water ratio and laboratory conditions, and C_A factor, in some cases, might not be effective because it represented gross heat evolve rather than the intensity of hydration and therefore did not represent the true hydration behavior. In order to overcome these problems, they proposed a new compatibility index (CI) expressed as:

$$CI = \sqrt{\left(\frac{Q_{emax}\, t'_{emax}}{Q'_{emax}\, t_{emax}}\right)} \cdot 100 \qquad (1)$$

where: Q_{emax} = maximum heat evolution rate of wood-cement mixture;
Q'_{emax} = maximum heat evolution rate of neat cement;
t_{emax} = the 'equivalent time' required to reach Q_{emax};
t'_{emax} = the 'equivalent time' required to reach Q'_{emax}.

The new index took into the consideration the capacity of the system, cooling rate, the varying temperature during the hydration process and the intensity the reaction. The initial results indicated that the *CI* values reflected the hydration behavior reasonably well. However, Pasca et al. (2010) believed that accelerator agents could alter the *CI*, since the agents only reduce the time of setting of cement; and therefore they artificially increase the value of the index, meant to be an overall compatibility assessor. Hence, he explored another new index *CX* expressed as:

$$CX = \sqrt[3]{\frac{HR_{max} H_{3.5-24}\, t'_{max}}{HR'_{max} H'_{3.5-24}\, t_{max}}} \qquad (2)$$

where: HR_{max} = maximum heat rate of wood-cement mixture;
HR'_{max} = maximum heat rate of neat cement;
t_{max} = time to reach ;
t'_{max} = time to reach ;
$H_{3.5\text{-}24}$ = total heat released in 3.5–24 h interval of wood-cement mixture;
$H'_{3.5\text{-}24}$ = total heat released in 3.5–24 h interval of neat cement.

This new index took into consideration most of the exothermic characteristics of wood-cement mixtures hydration: Maximum heat rate, time to reach that maximum heat rate, and total heat released during the chemical process, and diminished the artificial 'high' compatibility given especially through *CI* approach.

In addition, some physical tests have been utilized as indicators of wood-cement compatibility. Tchehouali et al. (2013) found that the compressive strengths values of wood-cement composites were linearly proportional to C_A factors so it is conclude that physical test could be a real indicator of wood-cement compatibility. Good correlations were also found between T_{max} and the compressive strength of wood-cement mixtures of various fiber–cement compositions. However, the correlations were low between t_{max} and the compressive strength (Wei et al. 2003). Electrical conductivity measurement was used to evaluate the compatibility of wood-cement composites in some studies (Juenger and Jennings 2002; Boustingorry et al. 2005).

CONLUSIONS

Wood-cement composites are now being investigated and made industrially in many countries in the world, mostly in the form of panels because of its excellent exterior properties. The main difficulty for wood-cement composites manufacturing is the chemical incompatibility between wood and cement (mainly OPC) which inhibits cement setting and hardening. The inhibitory substances mainly include some sugars, part of hemicelluloses and their degradation products. The inhibitory degree is affected by many factors including wood species, location, part of the tree, season during wood-cutting, wood/cement ratio, type of cement, storage condition, etc.

Before manufacturing wood-cement composites board, the wood-cement compatibility must be assessed. The main assessment methods involve the measurement of hydration characteristic of a cement-aggregate matrix and the comparison of the mechanical properties of cement-aggregate mixes. However, these methods are not effective for all test conditions or all wood species, and the assessment results based on these methods may be inconsistent. So the researchers have been making efforts to develop a more effective and accurate method that take into consideration more possible factors influence the assessment results.

In order to improve the wood-cement compatibility, many measurements can be taken such as aqueous extraction of wood before mixing with cement paste, addition of cement curing accelerators like $CaCl_2$,$MgCl_2$,Na_2SiO_3, $Al_2(SO_4)_3$ and some mineral chemicals, and gaseous or supercritical CO_2 injection. These methods are effective for the wood species that seriously hinder the cement hydration, and therefore promote the application of these species in manufacturing wood-cement composites products.

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