# The effect of four methods of surface activation for improved adhesion of wood polymer composites (WPCs)

Dimitriou, Athanasios; Hale, Michael; Spear, Morwenna

## International Journal of Adhesion and Adhesives

Version created as part of publication process; publisher's layout; not normally made publicly available

Cyswllt i'r cyhoeddiad / Link to publication

01. Jun. 2021

# Author's Accepted Manuscript

The effect of four methods of surface activation for improved adhesion of wood polymer composites (WPCs)

A. Dimitriou, M.D. Hale, M.J. Spear

Cite this article as: A. Dimitriou, M.D. Hale and M.J. Spear, The effect of four methods of surface activation for improved adhesion of wood polyme composites (WPCs), *International Journal of Adhesion and Adhesives* http://dx.doi.org/10.1016/j.ijadhadh.2016.03.003

# The effect of four methods of surface activation for improved adhesion of wood polymer composites (WPCs)

A. Dimitriou, M. D. Hale and M. J. Spear*

College of Natural Sciences, Bangor University, Bangor, LL57 2UW, UK

athdimitriou@hotmail.com

m.d.hale@bangor.ac.uk

m.j.spear@bangor.ac.uk

+44 1248 382029

*corresponding author

**Abstract**

Wood Polymer Composites (WPCs) have attracted a lot of interest in recent years as materials with a high renewable content. However the adhesion between WPC components is problematic because of low surface energy and the hydrophobic nature of the most widely used polymer matrices, i.e. polyolefins. Thus this paper has looked at four surface activation pretreatment methods to improve adhesion properties for bonding using epoxy adhesives, namely: hydrogen peroxide solution; hot air; a gas flame; and halogen heating lamps. The treatments were applied to WPC materials made from 60% wood flour in a polypropylene matrix, and lap joint shear strength was measured.

Shear strength values showed that all treatments except the halogen heating lamps increased the bond strength and the best results were achieved with hydrogen peroxide treatment at a pH of 7.5 (37% improvement); a two pass hot air treatment at a pass speed of 75 mm s$^{-1}$ (44% improvement); and a gas flame treatment at a pass speed of 175 mm s$^{-1}$(41 % improvement).The bond strength was increased to values that caused failure within the material, rather than at the interfaces of the bond line.

# 1. Introduction

Wood Polymer Composites (WPCs) are materials which are composed of lignocellulosic fibres and thermoplastic polymers in varying percentages [1]. Typically, solid profiles are formed by extrusion, and this material is suitable for mechanical jointing and bonding by traditional wood jointing techniques, but adhesion between components is currently poor. During recent years there has been special interest in developing plastics made using lignocellulose fibres as reinforcing fillers [2] and to reduce raw material costs. Although these improvements are not new developments the material is essentially renewable [3] and can be made employing recycled polymer and wood industry waste [4].

The most commonly used polymers for WPC production are the polyolefins – low density polyethylene (LDPE), high density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS) [1, 2, 5], which are suitable because of their low processing temperature. High processing temperatures (above 200°C) can cause thermal decomposition of the lignocellulosic reinforcement [3, 5]. Polyolefin wastes arise as a solid waste stream from cities, e.g. packaging materials, thus the re-use as matrix polymer for WPCs can provide green benefits [6]. The mechanical properties of recycled polymer WPCs are similar to those made from virgin polymers and in some instances, are better [7].

Jointing of WPCs normally uses mechanical fasteners which have been developed for wood, metal and polymeric materials [8]. On the other hand glued joints in WPCs can show poor adhesion properties, in the same manner as the native unfilled polyolefins. This is because of the limited wettability and low surface energy of the polyolefin matrix polymers [9] and this can be problematic in designing products such as furniture from WPC materials. In order to improve adhesion ability of polyolefins, surface activation treatments such as oxidative pre-treatments are essential [10, 11], but there are few studies in this area for WPC materials. The most common pre-treatments already used in the polymer industry are chemical, flame, corona, plasma and UV irradiation treatments [11, 12]. Other treatments like fluorination, oxy-fluorination and microwave irradiation are also effective for surface oxidation, which result in the improvement in adhesion [13, 14].

Moghadamzadeh *et al.* [15] clearly demonstrated that mechanical surface preparation by sanding altered the WPC surface from a polymer rich to a more wood-rich substrate. Sanding is a routine surface preparation technique in the wood furniture sector and has been applied to all samples including the control in this study. Also specifically for WPCs, Oporto and co-workers [10] performed a series of chemical, mechanical, energetic and physical pre-treatments (chromic acid, surface sanding, flame treatment, heat treatment, water, and combination of water-flame treatments). Almost all of the tested methods showed an improvement in the adhesion strength of their 50.4% pine, 39.6% PP composite. Heat treatment using a hot air gun was the only case where the adhesion strength was reduced. These improvements were recorded against unsanded control samples.

A greater number of studies have addressed the use of corona and plasma treatments for WPC materials [10, 12, 15, 16, 17, 18] than have investigated mechanical [10, 15] or flame [15, 19] or chemical [10, 12, 20] treatments for WPCs. Plasma and flame treatment options are suitable for use in factory production, however many WPC applications utilise relatively low investment technology, and a workshop scale of production. Flame and other sources of heat were considered to have potential for furniture manufacture within this context.

Due to its relatively high cost epoxy resin is a less commonly used adhesive for wood jointing in furniture industry [21], however it is widely used for polymer composites. Many researchers [10, 15,

19, 20] have used epoxy resin as the adhesion system for studies on WPC materials. A preliminary study on WPC materials [22] had compared lap shear strength of epoxy with the more traditional wood glues: PVA and polyurethane. In these tests epoxy outperformed PVA by 243.6% and PU by 198%, so was selected for use in the surface activation study.

In this study hydrogen peroxide and halogen heating lamps were chosen, alongside hot air and flame pretreatments. These were examined for surface oxidation and adhesion improvement. All the selected methods were chosen as safe and easily applicable procedures that could performed in a small workshop without the need of expensive, sophisticated equipment. The hot air gun, flame and halogen heating lamps were expected to activate the WPC surface as a result of the thermal degradation of the polymer. This mechanism is known to involve free radical formation by the action of heat. This may proceed by initiation and propagation as follows:

$$Polymer \rightarrow P\bullet + P\bullet$$

$$P\bullet + O_2 \rightarrow POO\bullet$$

$$POO\bullet + PH \rightarrow POOH + P\bullet$$
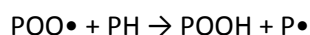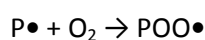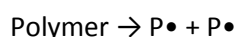
Peroxides are well known to form oxidative free radicals and are used for surface bleaching and surface colour modifications for wood [23] and thus suitable for surface application. Hydrogen peroxide forms a hydroperoxy radical $OOH\bullet$ which causes the surface oxidation as follows:

$$H_2O_2 \longleftrightarrow H\bullet + OOH\bullet$$

$$OOH\bullet + R\bullet \rightarrow ROOH$$

The $OOH\bullet$ radicals react with the material surface in the presence of an activator and significantly increases the surface polarity. NaOH is the most commonly used activator for controlling the pH of the treatment solution [23].

## 2. Materials and Methods

A WPC containing 60% Norway spruce flour, 35.5% polypropylene, 2% coupling agent, 1.5% UV-stabilizer and 1% colour pigment sourced from Kompetenzzentrum Holz GmbH Austria. This was produced under the European research project ERA-NET Cornet/2006/01. The WPC was an extruded square shaped pipe, allowing samples to be cut from the sides. 30% w/v $H_2O_2$ and 1M NaOH were obtained from Panreac Quimica SAU. A rapid curing two component commercial epoxy resin adhesive (Saldatutto mix) from Pattex was used for lap joint bonding.

WPC samples were cut with dimensions 40 mm X 20 mm X 5 mm and all samples were sanded with P220 glass paper to abrade the surface to produce a uniform surface for bonding. The samples were rinsed with deionised water after sanding to remove any remaining sanding dust on the surface, similarly, samples were rinsed after every surface activation treatment. Rinsing has been shown to have no significant effect on retention of surface activation for flame-treated polypropylene [24], and was required in this study to remove surplus treatment solution from the peroxide treated samples. They were conditioned at 20°C and 65% RH (relative humidity) to constant weight before treatment, and again afterwards. After each thermal treatment system (hot air, flame and halogen lamps), the samples were returned to the conditioning chamber to cool for 15 minutes before the rinsing step. The samples were then conditioned again to constant weight (2 weeks) before bond formation.

### 2.2. Surface pretreatments

Four pre-treatments were evaluated to determine whether they improved bond strength: a hydrogen peroxide treatment; a hot air treatment; a flame treatment and a halogen lamp treatment. Within each treatment, variables were examined to give different treatment intensities, and at each variable 20 replicates were included, giving a total of 560 samples tested. In addition 20 control samples were prepared by sanding and conditioning, and used as the untreated control set for each of the four pretreatment methods.

### 2.2.1. $H_2O_2$ treatments

A 30% $H_2O_2$ solution was used for the treatments at a series of pH steps from neutral, +/-2. The pH of the treatment solution was adjusted to 5, 6, 6.5, 7, 7.5, 8, 8.5 and 9 with NaOH. All treatments were performed at 20°C. The samples were soaked in the treatment solution for 30 minutes, then washed with deionised water and reconditioned to constant weight.

### 2.2.2. Hot air treatment

The samples were placed on a steel plate under the hot air gun. A DEWALT DW 340K 2000 W hot air gun was fixed to a STAUBLI TX90 robotic arm which was programmed to pass in a straight line direction over the WPC sample surfaces at a range of velocities (in mm $s^{-1}$: 18.5, 25, 31, 37.5, 67.5, 75, and 115). Thus more intense heating was achieved at slow pass velocities. A double pass at 75 mm $s^{-1}$ (2X75 mm $s^{-1}$) was also included for comparison. The distance between the sample surface and the hot air gun nozzle was fixed to 25 mm.

### 2.2.3. Flame treatment

In this treatment a MAPP gas flame torch was used, attached to the same robotic arm as for the hot air samples, but the vertical distance between the nozzle and sample surfaces was set to 50 mm and the pass velocities were faster (in mm $s^{-1}$: 125, 150, 175, 200, 225 and 250).

### 2.2.4. Halogen heating lamps treatment

For the halogen heating lamp treatment, three 400 W 20 cm long halogen heat lamps were combined to form a linear array with a spacing distance of 80 mm between them and the array unit was attached to the robotic arm 20 mm above the WPC sample surfaces. Samples were treated at slow pass velocities (in mm $s^{-1}$: 10, 20, 30, 40 and 50).

### 2.3. Lap joint preparation

The adhesive resin and hardener were mixed at a 1:1 w/w ratio and applied to both sample surfaces of the lap joint under laboratory conditions at 20°C. Then the joints were gripped with pneumatic clamps under equal pressure for 15 minutes to ensure sample uniformity within and between treatment groups. The bonding area was 10 mm X 20 mm (Figure 1a). All samples then conditioned as above for 48 hours.
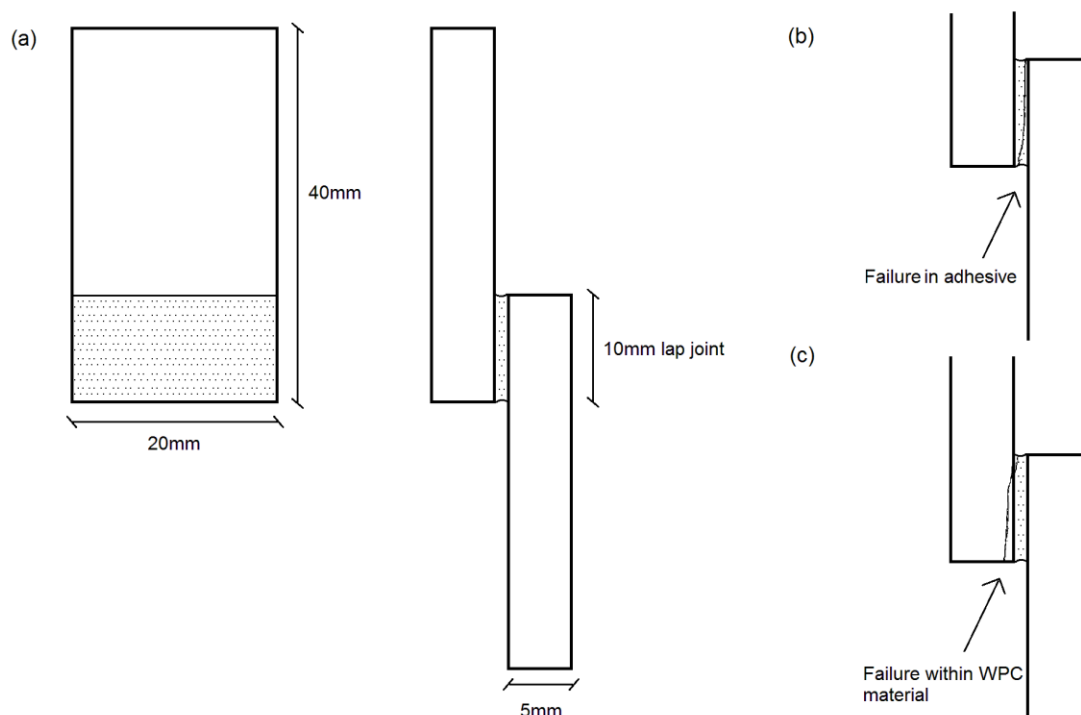
Figure 1. (a) Lap joint assembly (b) adhesive failure during shear test (c) material cohesive failure during shear test.

### 2.4. Shear testing of lap joints

For the bonding shear strength evaluation a Zwick/Roell Z020 universal testing machine was used. The mechanical properties were determined according to EN 205:2003 [25] using a 1.5 mm min$^{-1}$ cross head speed. The sample length was altered to accommodate the available dimensions of material (Figure 1a), but a lap joint area of 200 mm$^2$ was achieved as required. Mean lap-joint shear strength values were calculated using data from only samples where failure occurred in the glue joint. Analysis of the full sample set, including samples which showed material failure, was also conducted, and will be discussed where relevant in the text.

### 2.5. Statistical analysis

SPSS statistics version 17.0 software was used to carry out one way ANOVA treated and untreated control samples' shear strength values. The shear strength values were analysed with the treatment factors and untreated material (control) as the independent variable. Homogeneity variance, F-test and Tukey post hoc HSD multiple comparison test were performed to investigate the possible statistically significant differences within treatment factors compared to the control.

### 2.6. Scanning Electron Microscopy

Additional samples were prepared for each surface pre-treatment technique. Pieces of pre-treated WPC were cut to 20 x 20 mm for Scanning Electron Microscopy (SEM), mounted on stubs and carbon coated under vacuum (JEOL JEE-4X vacuum evaporator). A JEOL JMS 840A SEM was used to collect primary (backscattered) and secondary electron images.

## 3. Results

The shear strength results (Figure 2) showed adhesion strength improvements for all treatments as compared to the controls, except the halogen heating lamps, which gave lowered adhesion strength

at all speeds tested. The optimal treatment for each type of surface activation (hydrogen peroxide, hot air gun and flame) gave similar mean results (where all of the data is included) within the range of 801-836 N and gives proportional improvements in the order of 37-44%; all of these were considerable improvements. In some instances, where high increases in bond shear strength occurred, appreciable numbers of failures occurred within the material, although this was not statistically significant when tested by Tukey HSD test.



**a: Hydrogen peroxide**

**b: Hot air gun**

**c: Flame**

**d: Halogen heating lamps**
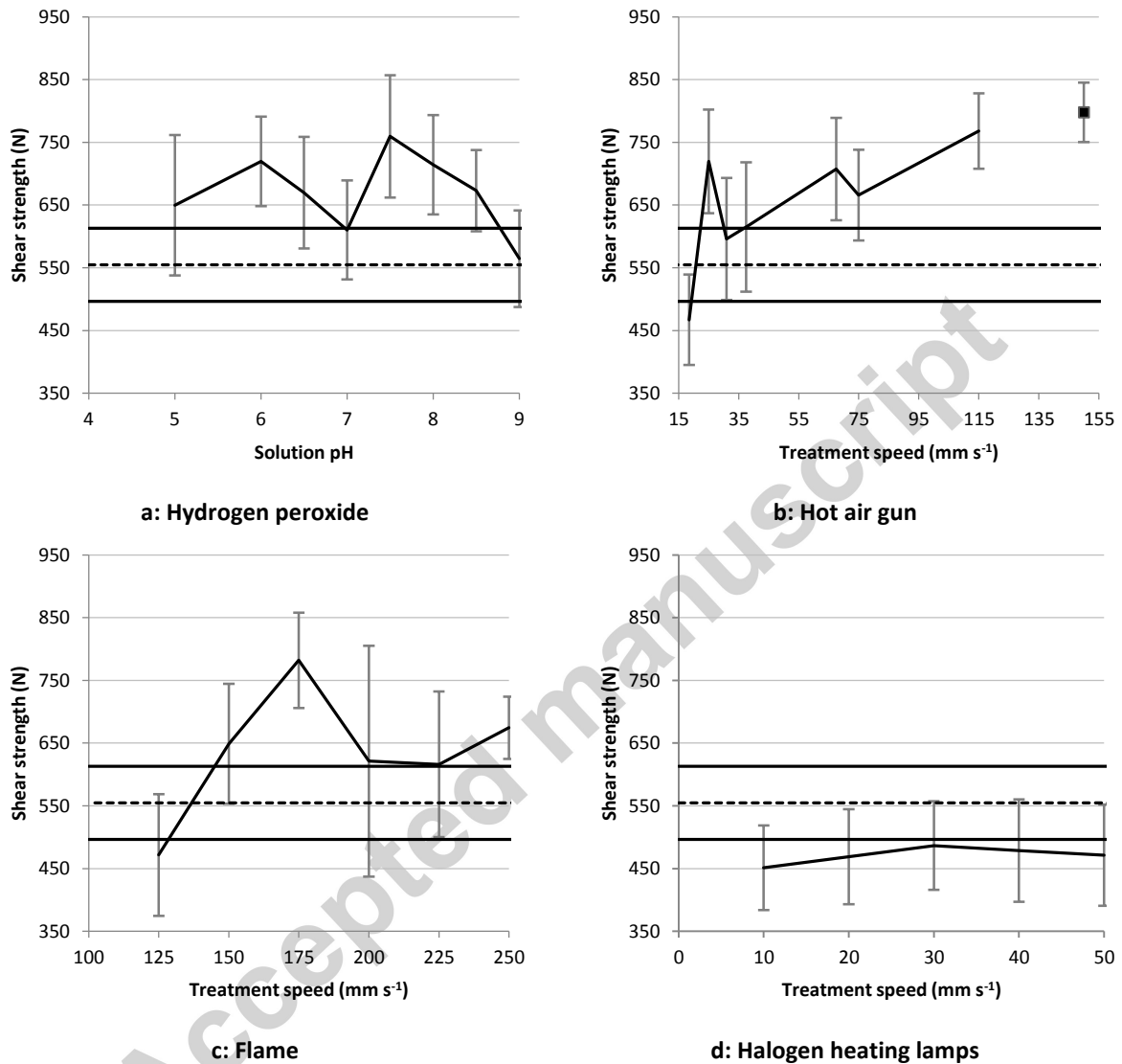
**Figure 2: Lap joint shear strength (N) of the four treatments. In each graph the horizontal dotted line presents the control mean value (555 N) and the solid horizontal lines the standard deviation (SD) of the control values. Error bars represent the SD (+/- 1). a) Hydrogen peroxide treatment. b) Hot air gun treatment. The mean value of the hot air gun at the 150 mm s$^{-1}$ treatment speed refers to the samples treated twice with the speed of 75 mm s$^{-1}$. c) Flame treatment and d) Halogen heating lamp treatment.**

### 3.1. Hydrogen peroxide treatment Lap-joint shear strength results

Hydrogen peroxide treatment significantly improved bond shear strength at some pH values (6, 7.5, 8 and 8.5, according to Tukey HSD test) but not at others, and at pH values of 7 and 9 they were

equal or worse than the controls. (Figure 2a). Those better than the controls, pH 6, 7.5, 8 and 8.5, had similarly high values of over 700 N (no significant difference, Tukey HSD test); overall the highest bond strength was seen at pH 7.5.

A large number, 80% of the of the pH 7.5 samples, failed by fracture within the WPC material rather than at the bond line, leading to their exclusion from the calculated mean value and standard deviation, according to the chosen EN 205 protocol. In the WPC samples the failure within the material occurred as a cohesive failure within the material immediately adjacent to the bonded area as shown in Figure 1c. This is similar to the failure which would be expected in the beech wood substrate commonly used in this test protocol. The EN 205 test [25] is predominantly used to evaluate the performance of adhesives, so failure in the material indicates the adhesive to be stronger than the substrate. Analysis of the shear strength values for the samples which showed material failure, revealed that failure occurred at a higher stress (809 N) than the samples which failed in the bond line (760 N) and the high failure rate (80%) within the pH 7.5 $H_2O_2$ treated WPC contrasts with only a 5% failure with control samples. This emphasises the good bonding with peroxide surface treatment at pH 7.5. Other peroxide treatments which showed modest increases of the within material failures (25%, 40%) were at the slightly higher pH values of 8 and 8.5 respectively and indicates little or no deterioration in the strength of the WPC substrate at these pH values as a result of the treatment.
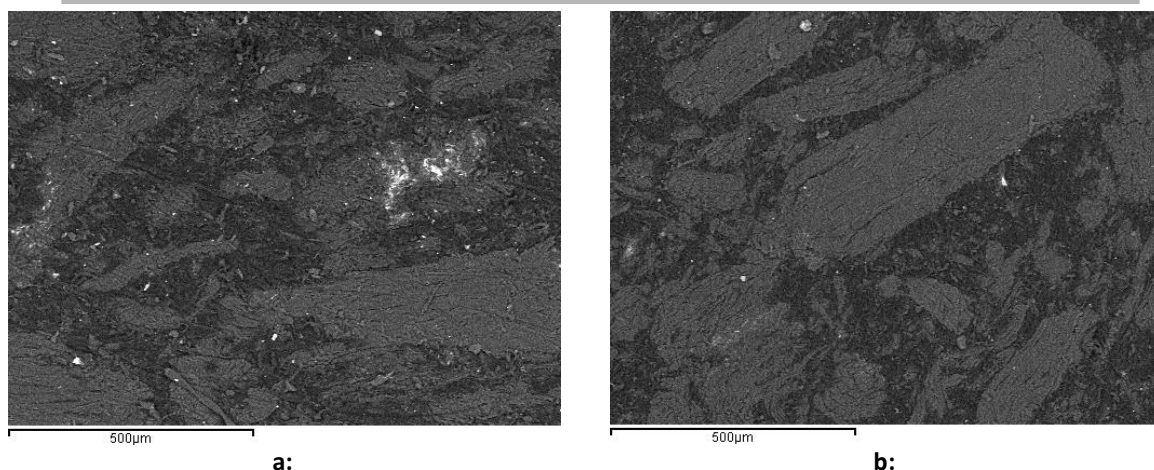
### 3.2. Hot air treatment

The hot air treatments at the faster pass velocities of 67-115 mm s$^{-1}$ and the double pass of 2 x 75 mm s$^{-1}$ also had a positive effect on the adhesion shear strength (Tukey HSD multiple comparison test). In general the slower speeds gave less positive results, but only the 25 mm s$^{-1}$ was significantly better than the controls (Figure 2.b).

It is interesting to note that the samples treated by two passes of the hot air at treatment pass speed of 75 mm s$^{-1}$ (75 x 2, Table 1) showed an improvement over those treated with a single pass at the same speed. The double pass of 75 mm s$^{-1}$ also gave the highest bond shear strength amongst all of the surface treatment investigated in this study, and the use of multiple passes warrants further investigation.

The double pass and the fast treatment speed of 115 mm s$^{-1}$ showed higher failure rates within the WPC (50%) which was, as with peroxide treatment at pH 7.5-8.5, associated with high adhesion shear strength values, in contrast to lower values at lower treatment speeds (Table 1). Conversely the samples treated at 18.5 mm s$^{-1}$ had lower bond strengths than the control samples and higher material failure, which is an indication of material degradation. In scanning electron microscopy studies distinct differences were observed in the surface for the heat treated samples (Figure 3b). These were smoother than the control, possibly indicating spreading of the polymer under the heat, reducing the prominence of sanding marks seen in the control (Figure 3a).

**Table 1: Shear strength (N) of samples surface modified at different pass velocities of hot air and percentages of samples which failed within the material rather than at the glue-line.**

| Property | Control | Treatment velocity (mm s$^{-1}$) | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | 18.5 | 25 | 31 | 37.5 | 67 | 75 | 115 | 75 (X2) |
| Shear strength (N) (EN205) | 555 | 467 | 719 | 596 | 615 | 707 | 666 | 768 | 798 |
| Within material failure (%) | 5 | 10 | 5 | 30 | 30 | 25 | 25 | 50 | 50 |
| Shear strength all samples (N) | 566 | 493 | 730 | 673 | 674 | 743 | 686 | 789 | 836 |

**Figure 3: Backscattered electron image of a) rough texture in sanded control sample and b) hot air treated sample (18.5 mm s$^{-1}$) where polymer surface (darker areas) appears smoother due to the action of heat.**

### 3.3. Flame treatment

Of the six different flame treatment pass speeds, from 125 to 250 mm s$^{-1}$, only two clearly and significantly (Tukey HSD test) improved the adhesion strength (Figure 2c, 175 mm s$^{-1}$ and 250 mm s$^{-1}$) but all treatments except the slowest velocity (125 mm s$^{-1}$) had a positive effect. High variation in the data gave insignificant differences for the other velocities. As well as giving the highest bond strengths, the samples treated at 175 mm s$^{-1}$ also had a higher percentage of material failure (65%) rather than at the bond line. This further indicates that 175 mm s$^{-1}$ is the optimal treatment velocity (Table 2). As expected, the samples with decreasing lap shear strengths showed progressively lower percentage material failures.

**Table 2: Shear strength (N) of samples surface modified at different pass velocities of a flame and percentages of samples which failed within the material rather than at the glue-line.**

| Property | Control | Treatment velocity (mm s$^{-1}$) | | | | | |
| --- | --- | --- | --- | --- | --- | --- | --- |
| | | 125 | 150 | 175 | 200 | 225 | 250 |
| Shear strength (N) (EN205) | 555 | 472 | 649 | 782 | 621 | 616 | 674 |
| Within material failure (%) | 5 | 0 | 20 | 65 | 10 | 5 | 25 |
| Shear strength all samples (N) | 566 | 472 | 666 | 841 | 641 | 631 | 704 |

### 3.4. Halogen heating lamps treatment

All of the halogen heat lamp treated samples had reduced bond strengths and high variability (Figure 2d), with mean values ranging from 451 to 487 N, some 12-19% less than the controls. No material failure was observed among the treated samples, probably due to the weakness of the bond.

According to the Tukey HSD multiple comparison test, all the samples treated with the halogen heating lamp were significantly statistically different to the untreated control and there were no statistically significant differences among the treatment speeds.

### 4. Discussion

From the micrographs (Figure 3) it is clear that the sanded WPC surface has both wood and polypropylene components exposed. Both components will interact with the adhesive in different

ways, relating to their surface energy, level of roughness and chemical functionality of the surface. The surface energy of polypropylene is typically reported as 30.1 mN m$^{-1}$ and for wood is 40 to 60 mN m$^{-1}$, resulting in values of around 31 mN m$^{-1}$ being recorded for WPCs with PP matrices [26, 27]. Reported values for the surface energy of epoxy resins vary with resin chemistry, but range from 39.1 to 51.6 mN m$^{-1}$ [28, 29], all of which are higher than the surface free energy of the substrate material, and indicate that poor wetting will occur. In the control samples of this study, which received sanding only, it is believed that this exposed the wood particles, and provided surface roughness for mechanical keying of the adhesive. Previous work [15] showed that there was a significant increase in adhesion after sanding compared with sample surfaces direct from the polymer moulding process. Sanding is common practice in wood adhesion, so was adopted throughout this study, prior to additional treatments.

The adhesion shear strength tests reported here have shown that hydrogen peroxide, hot air and flame treatment all show an improvement in adhesion strength when used as surface pre-treatments for WPCs. Only the halogen lamp treatment failed to have a beneficial effect under any of the parameters studied. For the four treatments evaluated, further work to observe changes in surface chemistry and surface texture development has also been undertaken but will be reported in paper 2.

Hydrogen peroxide solutions proved an effective treatment for WPC surface activation in order to improve its adhesion ability at specific pH values, particularly at pH 6 and 7.5 and 8 but less significantly at other values tested. This method has been previously reported for bamboo [14], but not in WPC material. The mechanism is believed to relate to free radical formation on the surface, which is likely to increase surface energy. WPC materials typically have low surface energies, due to the matrix polymers used. At pH 7.5 hydrogen peroxide gave an increase in shear strength of 37% but at this value high variation was encountered (SD=97.53). Curiously there was some activation either side of neutral pH values but not at neutral (pH 7), and towards the extremes of the pH values tested the values also decreased. At the optimal value the adhesion was sufficiently strong that shear failure occurred within the material rather than at the adhesive bond line. Further study of the different chemical mechanisms acting in the mild acidic and mild alkaline treatment solutions have been undertaken by FTIR spectroscopy and contact angle analysis, and appear to show significant differences. The effect of the treatment solution on the wood flour component and on the polypropylene component within the WPC surface, and resulting effects on the adhesion strength, will be reported in greater depth in a second paper.

Hot air treatment is also a quick and easy pretreatment to improve WPC adhesion and it is much faster and easier than hydrogen peroxide treatment because it does not require any chemical preparation and much less time is needed (few seconds) to obtain similar results. The 115 mm s$^{-1}$ and 2X75 mm s$^{-1}$ treatment velocities provided the best surface modification, giving an increase in bond strength of 38% and 44% respectively, resulting in 50% of the samples showing failure in the material rather than at the bond line. Low velocities resulted in low bond strengths and little within material failure which highlights the importance of amount of treatment. The improvement seen in this study contrasts with the results of Oporto and co-workers [10], who showed a 6% strength loss in their study which also used epoxy resin. They used a pass speed of 25 mm s$^{-1}$, but a different heat gun and the heat was applied from a greater distance (5 cm) from the surface. In the current study, the use of pre-sanded samples for heat treatment, and the shorter distance from the nozzle of the heat gun gave a significant increase in bond strength (30%).

Another important factor about the heat treatment is the case of the treatment repetition. It seems that two passes of the gun with the speed of 75 mm s$^{-1}$ was a more effective procedure than the

single pass at any speed. It could be expected that the treatment exposure would be the key factor for the treatment effectiveness but effectively a 2X75 mm s$^{-1}$ treatment is equivalent to a speed of 37.5 mm s$^{-1}$ in terms of overall treatment time. However the slower treatment pass speed resulted in much lower shear strength than both the 75 mm s$^{-1}$ and 2X75 mm s$^{-1}$ samples. It is likely that the exposure time of the 37.5 mm s$^{-1}$ causes higher temperature on the surface, which results in lower adhesion strength than the 2X75 mm s$^{-1}$. Therefore the repetition might ensure that a more optimal temperature is achieved for a short duration twice, which is most probably more important for priming the WPC surface in order to improve the adhesion strength. Initially it was thought that slow velocities would result in greater surface activation, but the opposite, in terms of bond strength, was observed. Adhesion mechanisms are a complex system which involves mechanical, chemical and energetic factors, which interact to affect the adhesion strength. In WPC materials these interactions are further complicated by the action of heat on two different substrates, with complex results. It is likely that at faster speeds the beneficial effect of the flame ionisation of the PP surface contributes to increased lap shear strength, with minimal change to the wood flour component. These factors have been further investigated using contact angle analysis to determine surface energy, and will be presented in another paper.

Although the flame treatment gave high mean bond strength values, particularly at velocities slower than 125 mm s$^{-1}$, the results were inconsistent, showing high standard deviation at many of the treatment velocities. At the optimal velocity of 175 mm s$^{-1}$ it gave a 41% improvement over the controls, similar to, but slightly lower than, that of the optimal hot air treatment flame (44% improvement). In terms of variation at these optimal values the flame treatment fared worse, i.e. flame 782 N at 175 mm s$^{-1}$ (SD 76.14); hot air 798 N at 2X75 mm s$^{-1}$ (SD 47.45) and thus is less stable. The higher variation could be explained by the effect of the flame on the wood flour component of the WPC. Despite this, the flame treatment is also an effective and quick method to improve the adhesion strength of the WPC. It does however also require accuracy in selecting the treatment speed, as incorrect speeds may result to adhesion strength decrease. In industrial flame treaters the control of feed speed can be well maintained, with Strobel et al [24] reporting typical values of 100 to 300 m min$^{-1}$ for their experimental studies. In this study the feed speed was slower (7.5 to 15 m min$^{-1}$), which may have contributed to higher levels of oxidation and greater variability in properties. In a workshop preparing small joint areas where the speed is controlled manually there may be a further increase in the levels of variability seen. In addition, the flame treatment has a medium level of risk, as it involves applying a flame on flammable materials and there is a greater risk of volatile emissions. Although the decomposition products of polypropylene itself are relatively harmless; care should therefore be taken if transferring the technique to other polymers.

Overall the heat treatments applied by hot air gun and flame showed that they were both effective treatments for improving the adhesion ability of the WPC and also too slow a treatment, i.e. excessive heating, led to lowered bond strength. Concerning the treatment method efficiency, Oporto's study [10] agrees in the case of flame treatment but disagrees with the heat air gun treatment. It is likely that parameters such as treatment pass speed, proximity of the flame or hot air source to the substrate surface, and the width of the area receiving flame or hot air treatment, vary from machine to machine and have significant influence on the level of ionisation which occurs in the polypropylene component of the WPC material. The same parameters may also influence the level of thermally induced change in the wood flour component, albeit small in many cases, and conflict between the beneficial and adverse effects seen in the two components may explain the suboptimal results observed in some treatment speeds. This area is worthy of greater investigation and discussion to seek an optimal treatment.

The halogen heating lamps treatment does not appear to have any advantageous effect on the WPC adhesion ability. It produces a modified surface that has a reduced adhesion strength, with values lower than the untreated WPC. This treatment is possibly useful to help us understand the factors that negatively affect the adhesion strength of the WPC.

## 5. Conclusions

This study has demonstrated three effective surface pretreatment methods for WPCs: hydrogen peroxide solutions, hot air and flame. A further treatment using halogen heat lamp failed to show an improvement in adhesion at any of the speeds studied (10 to 50 mm s$^{-1}$).

The hydrogen peroxide solutions which showed greatest benefit were pH 6, 7.5, 8 and 8.5. Hydrogen peroxide at these close to neutral pH values is easily applied and controlled, and offers a simple system for use in the workshop. A timer and a pH meter is all the equipment required, and the amount of NaOH which is added into the $H_2O_2$ solution for controlling the pH is in very small quantities that are considered safe [23].

The use of heat for surface activation provided by a hot air gun or from a flame, also improved adhesive bonding. The majority of conditions studied used single passes of the heat gun or flame torch. Optimal treatment speeds for the two systems were different – with 115 mm s$^{-1}$ heat gun and 175 mm s$^{-1}$ for the flame torch. In the heat gun an additional experiment using two passes at a slower speed of 75 mm s$^{-1}$ also showed beneficial results of a similar level.

**6. References**

1. Stark M.N. and Matuana M.L. 2007. Characterization of weathered wood-plastic composite surfaces using FTIR spectroscopy, contact angle and XPS. Polym Degrad Stabil 92:1883-1890.

2. Son J., Kim H. and Lee P. 2001. Role of Paper Sludge Particle Size and Extrusion Temperature on Performance of Paper Sludge-Thermoplastic Polymer Composites. J Appl Polym Sci 82:2709-2718.

3. Rowell R.M. 2007. Challenges in Biomass-Thermoplastic Composites. J Polym Environ 15:229-235.

4. Cui Y., Lee S., Noruziaan B., Cheung M. and Tao J. 2008. Fabrication and interfacial modification of wood/recycled plastic composite materials. Compos Part A-Appl S 39:655-661.

5. Clemons C. and Stark N. 2007. Use of Saltcedar and Utah Juniper as Fillers in Wood-plastic Composites. Forest Products Laboratory Research Paper 641. 17 pp.

6. Selke E.S. and Wichman I. 2004. Wood fiber/polyolefin composites. Compos Part A-Appl S 35:321-326.

7. Adhikary B.K., Pang S. and Staiger P.M. 2008. Dimensional stability and mechanical behaviour of wood-plastic composites based on recycled and virgin high-density polyethylene (HDPE). Compos Part B-Eng 39:807-815.

8. Farid S.I., Kortschot M.T. and Spelt J.K. 2002. Wood-Flour-Reinforced Polyethylene: Viscoelastic Behavior and Threaded Fasteners. Polym Eng Sci 42(12):2336-2350.

9. Ritter G.W. 2000. Surface Preparation IV: Treatment of Plastics II. ASI (Adhesives & Sealants Industry Magazine) http://www.adhesivesmag.com/articles/85846-surface-preparation-iv-treatments-for-plastics-ii (Accessed online 19/1/15)

10. Oporto S.G., Gardner J.D., Bernhardt G. and Neivandt J.D. 2007. Characterizing the mechanism of improved adhesion of modified wood plastic composite (WPC) surfaces. J Adhes Sci Technol 21(11):1097-1116.

11. Pinto A.M.G., Magalhaes A.G., Silva F.G. and Baptista A.P.M. 2008. Shear strength of adhesively bonded polyolefins with minimal surface preparation. Int. J. Adhes. Adhes. 28:452-456

12. Gupta B.S. and Laborie M.-P.G. (2007) Surface activation and adhesion properties of wood-fiber reinforced thermoplastic composites. J. Adhes. 83(11):939-955.

13. Du Toit F.J. and Sanderson R.D. 1999. Surface fluorination of polypropylene 1. Characterization of surface properties. J Fluorine Chem 98:107-114

14. Mirabedini S.M., Hamid R., Hamedifar Sh. and Mohseni S.M. 2004. Microwave irradiation of polypropylene surface: a study on wettability and adhesion. Int J Adhes Adhes 24:163-170.

15. Moghadamzadeh H., Rahimi H., Asadollahzadeh M. and Hemmati A.R. (2011) Surface treatment of wood polymer composites for adhesive bonding. Int J Adhes Adhes 31(8):816-821.

16. Wolkenauer G., Avrimidis E., Hauswald H.M., Militz H. and Viöl W. 2008. Plasma treatment of wood-plastic composites to enhance their adhesion properties. J. Adhes. Sci. Techn. 22(16):2025-2037.

17. Liu Y., Tao Y., Lv X., Zhang Y. and Di M.W. 2010. Study on the surface properties of wood/polyethylene composites treated under plasma. Appl. Surf. Sci. 257:1112-1118.

18. Tao Y. and Di M.W. (2011) Study on plasma treatment and adhesion of wood/polyethylene composites. Appl. Mechan. Materials 66-68:911-915.

19. Gramlich W.M., Gardner D.J. and Neivandt D.J. (2006) Surface treatments of wood-plastic composites (WPCs) to improve adhesion. J. Adhes. Sci. Technol. 20(16):1873-1887.

20. Teng X. and Di M. (2011) Effect of liquid oxidation surface treatment on bonding properties of wood/polyethylene composites. Appl. Mechan. Materials. 66-68:907-910.

21. Stoeckel F., Konnerth J. and Gindl-Altmutter W. (2013) Mechanical properties of adhesives for bonding wood – a review. Int J Adhes Adhes 45:32-41.

22. Dimitriou A. (2015) Surface pre-treatment methods for improving adhesion ability in wood polymer composite for frame and furniture applications. PhD Thesis, Bangor University.

23. Lu K.T. 2006. Effects of hydrogen peroxide treatment on the surface properties and adhesion of ma bamboo (*Dendrocalamus latiflorus*). J Wood Sci 52:173-178.

24. Strobel M., Jones V., Lyons C.S., Ulsh M., Kushner M.J., Dorai R. and Branch M.C. (2003) A comparison of corona-treated and flame-treated polypropylene films. Plasmas and Polymers 8(1):61-95.

25. BSI (2003) BS EN 205: 2003 Adhesives – wood adhesives for non-structural applications – determination of tensile shear strength of lap joints.  14pp.

26. Mohamed-Ziegler I., Oszlánczi Á., Somfai B., Hórvölgyi Z., Pászli I., Holmgren A. and Fosling W., (2004) Surface free energy of natural and surface modified tropical and European wood species. J. Adhes. Sci. Technol. 18(6): 687-713.

27. Gupta B.S., Reiniati I. and Laborie M-P.G. 2007. Surface properties and adhesion of wood fiber reinforced thermoplastic composites. Colloid. Surf. A: Physicochem. Eng. Aspects. 303:388-395

28. Wu S. (1989) Part VI, in: Polymer Handbook, J. Brandrup and E.H. Immergut (Eds), pp.414-426.

29. Occhiello E., Morra M., Morini G., Garbassi F. and Johnson D. (1991) On oxygen-plasma treated polypropylene interfaces with air, water, and epoxy resins. II. Epoxy Resins. J. Appl. Polym. Sci. 42(7):2045-2052.