Part 1  Studies on wood-to-wood bonding adhesives based on natural rubber latex.

Adhesives are used in construction application for bonding wood pieces\(^1\). The mechanism of wood adhesion is related to the complex physical structure and chemistry of the substrates. Wood is largely composed of cellulose and lignin and to a lesser degree of extractables which differ greatly in their reaction to adhesives\(^2\). Some wood adhesives promote strong forces of covalent bonding, while in the case of others, weaker adhesion forces such as van der waals forces and hydrogen bonding operate and/or mechanical interlocking may occur\(^3\)\(^,\)\(^4\).

Generally, latex based adhesive offer a great advantage over other adhesive systems for joining porous substrates. The water phase of these adhesives tend to penetrate into the material and hence results in faster drying of the adhesives\(^5\).

Natural rubber latex and synthetic latices once produced as water-dispersed adhesives cannot be redispersed in water after they have been dried out. A forced drying occurs while drying latex adhesives\(^6\)\(^,\)\(^7\). The most consistent theory for the film formation of water dispersed adhesives is the one proposed by Vanderhoff et al\(^8\).

Water based adhesives are more advantageous than solvent based adhesives because organic solvent emitted into air are pollutants and are toxic in nature, and may cause explosion hazard. Furthermore, solvent recovery and recycling require more expensive equipment, compared to water\(^9\).

In this part of the work we propose to prepare NR latex based adhesives and to study their efficiency as a bonding agent for wood-to-wood bonding. Different formulations based on NR latex are proposed to be used for wood bonding. The effect of degree of prevulcanisation on bond strength is also proposed to be studied. The effect of the amount of carboxymethyl cellulose, ammonium caseinate, phenol formaldehyde resin(PF), cooked
starch, etc. on bond strength also will be investigated. The changes in bond strength with aging of the adhesive are also proposed to be studied.

Experimental

Fevicol was supplied by Pidilite Industries, Bombay, India. Wood used was a kind of soft wood of carpentry grade.

The centrifuged latex was deammoniated to 0.2% ammonia content. All water insoluble ingredients were added as dispersions in water. The compounding ingredients were added as per two formulations, L1 and L2 (Table 1), with formulation L1 used as base formulation.

Wood pieces were cut into 25x300x3 mm strips. These wood pieces were dried and polished using sand paper No. 60 (250μm), and then the adhesives were applied to a thickness of 0.2 mm on the two wood pieces and bonded as a lap joint, applying a load of about 1 kg for 24 h. The lap joints were conditioned for 7 days at room temperature. The tension shear strength (N/cm²) of the bonded joints was tested on a Zwick UTM model 1445 at ambient temperature according to ASTM D 903-49 (72).

Table 3.1.1 Formulation

<table>
<thead>
<tr>
<th>Ingredients, Phr</th>
<th>L1</th>
<th>L2</th>
</tr>
</thead>
<tbody>
<tr>
<td>NR latex (60% DRC)</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>KOH (10%)</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Dispersol VL</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>CMC</td>
<td>0.0</td>
<td>3.0</td>
</tr>
<tr>
<td>Ammonium caseinate</td>
<td>3.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Sodium silicate</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>Starch</td>
<td>3.0</td>
<td>3.0</td>
</tr>
<tr>
<td>Sulphur (50%)</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>ZDC (50%)</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>
Latex compound L2 was subjected to prevulcanisation for different times varying from 12 to 45 min at a constant temperature (70°C). The prevulcanised compounds were then used for bonding wood pieces and the lap shear strength was determined as described earlier. The optimum prevulcanisation time for different compounds were determined. The extent of prevulcanisation was determined by chloroform coagulation test. Latex was stirred with a fixed proportion (equal volumes) of chloroform and degree of vulcanisation was judged from the appearance of the coagulum formed. When the degree of crosslinking increases, the rubber particles coalesce together less rapidly to form a coherent coagulum. As the degree of vulcanisation increases the coagulum changes from soft to a crumbly material. Here the coagulum is a weak lump which breaks easily when stretched.

The effect of varying the amount of thickening agent, carboxy methyl cellulose, from 1 to 5 phr in formulation L2 (Table 1) on the bond strength was studied. The latex compounds were prevulcanised for 30 min at 70°C prior to joint strength determination.

The lap shear strength of bonded specimens with amounts of ammonium caseinate varying from 1 to 5 phr was also determined as above. These latex compounds were also prevulcanised for 30 min at 70°C.

Similarly, the variation of PF resin from 10 to 60 phr and cooked starch from 2 to 5 phr in formulation L1 (Table 1) was studied. The bond strength of lap joints using prevulcanised latex compounds was also measured as described above.

The water resistance of the bonded samples was determined by accelerated aging methods involving sample exposure to cold water (30°C), and hot water (100°C). Some bonded pieces were placed in cold water for 1 day at room temperature and then dried prior to lap shear testing. Similarly, some bonded pieces were placed in hot water for 1 hr and lap shear strength was determined. The water resistance test was conducted according to ASTM D1151 (72) procedure.

Latex compound L1 was aged for 30 and 45 days at room temperature and subsequently used to make lap joints of wood, and lap joint shear strength was determined.
Results and Discussion

The lap shear strength is found to increase with the prevulcanisation time, attain a maximum value and then decrease (Figure 3.1.1). This shows that prevulcanisation time increases, good bonding develops, but as the extend of prevulcanisation increases the inter particle bonding decreases, resulting in a reduction in the lap shear strength. This trend shows that there is an optimum level of prevulcanisation time which results in maximum bond strength. Conventional sulphur vulcanisation occurs in each rubber particle.

![Graph showing lap shear strength vs prevulcanisation time](image)

**Fig. 3.1.1 The effect of prevulcanisation time on lap shear strength**

The lap joint strength increases as the carboxy methyl cellulose (CMC) content increases to 2 - 3 phr and then decreases (Figure 3.3.2). This shows that CMC can be used to get excellent wet adhesion strength. It can be used as core binder. This trend may be due to the increase in the viscosity of the latex compound as the CMC acts as a thickening agent. This further shows result shows that there is an optimum viscosity for a latex based adhesive for obtaining maximum bond strength. The bond strength of prevulcanised latex compounds
show a trend similar to that for unvulcanised latex compounds (series 1), with prevulcanised being (series 2) slightly better in strength.

As the amount of ammonium caseinate increases, the lap joint strength increases (Figure 3.3.3). This shows that ammonium caseinate can act as a tackifier in NR latex based adhesives. Ammonium caseinate also acts as thickener for NR latex, which increases the viscosity thus preventing streaming when applied to vertical surfaces. The bond strengths of prevulcanised

![Graph 3.1.2: Variation of lap shear strength with amount of CMC](image)

![Graph 3.1.3: Variation of lap shear strength with ammonium caseinate content](image)
latex (series 2) and unvulcanised latex (series 1) show the same pattern of joint strength. The slight reduction in bond strength in the case of prevulcanised latex may be due to over cooking of the ammonium caseinate while prevulcanising the latex. Ammonium caseinate enhances the durability of wood joints because of its higher molecular weight. The ammonia present will escape easily, resulting in insoluble casein. The active amino acid group is CONH, which has higher molecular weight and superior water resistance.

Phenol formaldehyde (PF) resin modifies the viscoelastic properties of the rubber components so that immediate wetting and bonding occur. The lap joint strength increases as the amount of PF resin increases up to 50 phr, after which the bond strength decreased with increasing amount of PF resin (Figure 3.1.4). This result may be due to the phase change, i.e. the PF resin phase becoming the continuous phase, resulting in higher modulus, due to poor wetting. Series 1 unvulcanised and series 2 prevulcanised in figure. The bond strength of the prevulcanised latex shows a pattern similar to that of unvulcanised latex compounds but displays only a lower joint strength.

![Figure 3.1.4 Variation of lap shear strength with amount of PF resin.](image)

---

58
In the dispersed form, starch has a higher degree of spreadability and filmness and is capable of wetting polar surfaces such as cellulose, penetrating crevices and pores, and forming strong adhesive joints.

It is the result of both mechanical interlocking and the interaction of secondary bond forces (van der Waal's and hydrogen bonding)\textsuperscript{12}. The variation of lap shear strength of prevulcanised (series 2) and unvulcanised (series 1) latex compounds with the amount of starch is shown in figure 3.1.5. This result shows that starch can increase the bond strength of latex-based adhesives, and there is an optimum amount of starch which can give the maximum bond strength.

![Graph showing lap shear strength variation with starch content](image)

Table 3.1.2 shows the water resistance of the bonded samples using a commercial adhesive (Fevicol) and NR latex-based adhesive (L1). The NR latex based adhesive shows higher retention in bond strength, both in cold and in hot water. In hot water, the unvulcanised latex-based adhesive shows better results. This may be due to the crosslinking efficiency of the latex film in between the wood pieces in hot water.
Table 3.1.2 Percent retention of dry strength vs accelerated ageing shear strength.

<table>
<thead>
<tr>
<th>Property</th>
<th>Commercial % Retention</th>
<th>Unvulcanised(L.1) % Retention</th>
<th>Vulcanised(L.1) % Retention</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lap shear strength(dry), N/cm²</td>
<td>181.5</td>
<td>90.9</td>
<td>66.7</td>
</tr>
<tr>
<td>Shear strength after 1 day in cold water, N/cm²</td>
<td>45.8</td>
<td>44.7</td>
<td>41.0</td>
</tr>
<tr>
<td>% Retention</td>
<td>25</td>
<td>49</td>
<td>62</td>
</tr>
<tr>
<td>Shear strength after 1 h in hot water, N/cm²</td>
<td>0</td>
<td>33.7</td>
<td>23.5</td>
</tr>
</tbody>
</table>

The effect of room temperature aging on bond strength is shown in Table 3.1.3. The slight reduction in bond strength observed may be due to the natural destabilization of the latex during storage.

Table 3.1.3 Lap joint strength using the room temperature-aged NR latex

<table>
<thead>
<tr>
<th>NR latex type</th>
<th>Fresh</th>
<th>After 30 days</th>
<th>After 45 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unvulcanised (L.1) lap shear strength(N/cm²)</td>
<td>90.9</td>
<td>68.4</td>
<td>58.8</td>
</tr>
<tr>
<td>Vulcanised (L.1) lap shear strength(N/cm²)</td>
<td>66.7</td>
<td>38.4</td>
<td>30.5</td>
</tr>
</tbody>
</table>

Conclusions

1. A natural rubber latex compound can be used as an adhesive for wood-to-wood bonding.
2. The bond strength of NR latex adhesives can be improved by prevulcanising the latex compound to an optimum level.
3. Modification of NR latex compounds using ammonium caseinate and carboxy methyl cellulose improves the lap shear strength.
4. The addition of PF resin enhances the bond strength.
5. The NR latex adhesives show higher resistance both in cold and in hot water, compared with a conventional latex based adhesive, Fevicol.


References

Part 2 Use of styrene butadiene vinyl pyridine copolymer latex based adhesives for bonding wood.

Experimental:

Wood pieces were bonded using VP latex compound as per the procedure described in part 1. The formulations for VP latex compound is given below in Table 3.2.1.

Table 3.2.1 Formulations of VP latex compounds, \( \rho_{\text{VR}} \)

<table>
<thead>
<tr>
<th>INGREDIENTS</th>
<th>V</th>
<th>V1</th>
<th>V2</th>
<th>V3</th>
<th>V4</th>
<th>V5</th>
</tr>
</thead>
<tbody>
<tr>
<td>VP LATEX</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>AMMONIUM</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CASEINATE</td>
<td>0</td>
<td>30</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>STARCH</td>
<td>0</td>
<td>0</td>
<td>30</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>CMC</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>30</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>RF1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>RF2</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>SULPHUR</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>ZDC</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

The latex compound was prevulcanised for different times ranging from 10 to 40 min. at a constant temperature (70°C). The prevulcanised latex compounds were used for bonding wood pieces and strengths were determined as in part 1 of this chapter.

The effect of varying the amount of thickening agent ammonium caseinate and carboxy methyl cellulose and modifiers like cooked starch, RF1 (slightly alkaline) and RF2 (slightly acidic) resins in the VP latex compound on joint strength was determined. These latex compounds were prevulcanised for 30 min. at 70°C and joint strengths were studied as in the case of NR latex in part 1.

A mixed system with different combinations of ammonium caseinate and cooked starch were prepared and added to the latex and lap shear strength was determined. The effect pH change on lap shear strength was also studied. The pH value was changed by the addition of
dilute ammonia solution. The compounded VP latices were kept for one month and lap shear strength was determined after equal intervals of time.

The wood joints with the VP latex adhesives were kept immersed in cold water (30°C) for 24 hours, in hot water (100°C) for 2 hours, in acid (80°C) for 2 hours and in alkali (80°C) for 2 hours and joint strength was determined after taking out the specimens and drying as per ASTM 1151 (72). Same tests were conducted using commercial adhesive also.

Optical photographs of the wood surfaces before adhesive application and after peeling were taken.

Results and Discussion

Figure 3.2.1 shows the effect of lap shear strength of wood joints bonded with VP latex compound VI (Table 3.2.1) with varying time of prevulcanisation. Lap shear strength increases, reaches a maximum value and then decreases as prevulcanisation time increases which may be due to lower film strength of the latex after attaining a required prevulcanisation.

![Graph showing effect of prevulcanisation time on lap shear strength](image)

Figure 3.2.2 shows the prevulcanisation time for different VP latex compounds. For the compounds with CMC the lap shear strength increases when prevulcanisation time increases. In the case of ammonium caseinate, cooked starch and resorcinol formaldehyde resins (RF1 &
RF2) the joint strength increases, reaches a maximum and then decreases. The increase in lap shear strength with prevulcanisation may be due to the increased film strength, but after attaining a state of prevulcanisation the particle coalescence becomes poor, resulting in decreased lap shear strength.

The lap joint strength increases as the carboxy methyl cellulose (CMC) content increases to 7.5 phr and then decreases (Figure 3.2.3). This may be due to the increase in the viscosity of

![Graph](image_url)

**Fig. 3.2.2** Variation of Prevulcanisation time with lap shear strength

![Graph](image_url)

**Fig. 3.2.3** Variation of lap shear strength with CMC content.
latex compound as CMC acts as a thickening agent. This result shows that there is an optimum viscosity for latex based adhesives for obtaining maximum bond strength. Bond strength of prevulcanised latex compounds (prevulcanised for its optimum prevulcanisation time) show almost similar trend as that of unvulcanised latex compounds but show higher joint strength. This may be due to the increased film tensile strength of prevulcanised latex film.

As the amount of ammonium caseinate increases, the lap joint strength increases (Figure 3.2.4). This shows that ammonium caseinate can act as a thickener in VP latex based adhesives. The bond strength of prevulcanised latex and unvulcanised latex show the same pattern. Ammonium caseinate increases the durability of wood joints because of its higher molecular weight. Ammonia present will be lost easily, resulting in insoluble casein. The use of thickener is to increase viscosity which can prevent streaming when applied to vertical surfaces. It may also suppress any tendency for the aqueous phase of the adhesive to separate out of the adhesive when applied to porous surfaces. The separation of aqueous phase may prevent the development of optimum bond strength cause staining of the adherends.

In dispersed form, starch has high degree of spreadability and filminess and is capable of wetting polar surfaces such as cellulose, penetrating crevices and pores, and forming strong adhesive joints. It is the result of both mechanical interlocking and the interaction of secondary
bond forces. The variation of lap shear strength of prevulcanised and unvulcanised latex compounds with the amount of starch is shown in Figure 3.2.5. This shows that starch can increase the bond strength of latex based adhesive and when prevulcanised there is a decrease in bond strength which may be due to the over cooking of starch while prevulcanising.

Fig. 3.2.5 Variation of lap shear strength with starch content.

Fig. 3.2.6 Variation of Lap shear strength with RF1 resin content
Figure 3.2.6 and Figure 3.2.7 show the effect of variation of RF1 and RF2 resins in VP latex compound respectively. Both resins show similar trend in prevulcanised and unvulcanised state.

There is an optimum amount of RF resin which gives maximum joint strength.
Figure 3.2.8 shows the effect of pH value on lap shear strength. As pH increases from 7.2 to 8.2 the lap shear strength increases. The pH value in the alkaline range gives more adhesion strength.

Fig. 3.2.9 Variation of lap shear strength of mixed systems

Fig. 3.2.10 Variation of Lap shear strength with days of storage
Figure 3.2.9 shows the lap shear strength of different combinations of ammonium caseinate and starch in VP latex. The different combinations do not give a higher strength compared to when they are used individually.

Figure 3.2.10 shows the variation of lap shear strength with the number of days of storage. Even after 30 days of storage the lap shear strength of the adhesives are not changed much. This shows that the VP latex based adhesives possess good shelf life.

Table 3.2.2 shows the resistance of VP latex based adhesives in cold water, hot water, acid and alkali compared to natural rubber latex and commercial latex based adhesive, Fevicol. VP latex based adhesives show much higher retention in strength compared to natural rubber based wood adhesive and Fevicol.

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>Original</th>
<th>Water</th>
<th>Hot water</th>
<th>Acid</th>
<th>Alkali</th>
</tr>
</thead>
<tbody>
<tr>
<td>V1</td>
<td>260.2</td>
<td>259.2</td>
<td>145.7</td>
<td>244.4</td>
<td>234.8</td>
</tr>
<tr>
<td>V2</td>
<td>109.2</td>
<td>28.6</td>
<td>27.7</td>
<td>97.1</td>
<td>96.9</td>
</tr>
<tr>
<td>V3</td>
<td>148.3</td>
<td>118.6</td>
<td>45.2</td>
<td>109.6</td>
<td>108.6</td>
</tr>
<tr>
<td>V4</td>
<td>174.1</td>
<td>119.7</td>
<td>57.8</td>
<td>151.0</td>
<td>150.5</td>
</tr>
<tr>
<td>V5</td>
<td>176.59</td>
<td>100.23</td>
<td>50.6</td>
<td>181.31</td>
<td>180.6</td>
</tr>
<tr>
<td>FEVICOL</td>
<td>200.6</td>
<td>58.5</td>
<td>0.0</td>
<td>6.3</td>
<td>0.6</td>
</tr>
<tr>
<td>NR LATEX</td>
<td>90.9</td>
<td>44.8</td>
<td>33.7</td>
<td>30.8</td>
<td>31.2</td>
</tr>
</tbody>
</table>

Photograph 3.2.1 shows the roughed wood surface used for adhesive application. Photograph 3.2.2 and 3.2.3 shows the peeled off wood surface with laboratory adhesive and commercial adhesive(Denrite) respectively. From the photograph it can be observed that the rubber film adhering to the surface is more in the case of laboratory adhesive so we get more adhesion strength. Here adhesive failure occurred during separating the adherends.
Photograph 3.2.1 Roughed wood surface before adhesive application.

Photograph 3.2.2 Peeled off wood surface with laboratory adhesive.

Photograph 3.2.3 Peeled off wood surface with commercial adhesive.
Conclusions

1. VP latex based adhesives can be used for bonding wood pieces.
2. An optimum amount of ammonium caseinate, CMC, starch and RF resin in VP latex adhesives gives maximum bond strength in wood joints.
3. Prevulcanisation of the latex improves bond strength.
4. VP latex based wood adhesives have good shelf life.
5. The resistance of VP latex based adhesives to water, acid and alkali are found to be superior to that of commercial adhesive.

References

Part 3 Rubber solution adhesives for wood to wood bonding

The performance or behavior of wood adhesive system is dependent on a wide range of variables, such as surface smoothness of wood substrate, pH, presence of extractables, amount of debris present and others which are related to the environment conditions such as temperature and relative humidity. The bonding mechanism of wood adhesives is related to the complex chemistry of the substrates. In some cases, strong forces of covalent bonding may occur which in others weaker forces such as Van der waal’s forces and Hydrogen bonding may operate or mechanical interlocking may occur.

Adhesives based on urea formaldehyde (UF) and phenol formaldehyde (PF) are the major adhesives used for bonding wood. But these adhesives are very sensitive to hydrolysis and stress scission. The UF and PF adhesives produce health hazards due to formaldehyde release. Isocyanate type adhesives have disadvantages like shorter pot life, higher cost, limited durability and lack of tack etc.

In this part of the study development of solution based wood adhesives based on chloroprene rubber and natural rubber and their evaluation are proposed.

Experimental

Adhesives used:
A - Adhesive prepared in laboratory.
B - Dunlop adhesive supplied by Dunlop India Ltd, Calcutta.
C - Superbond adhesive supplied by Superchem MIDC, Thane.
D - Dentrite adhesive supplied by Chandra’s chemicals enterprises, Calcutta.
E - Fevibond adhesive supplied by Pidilite Industries Ltd, Bombay.

Adhesive preparation:

Rubber compounds were prepared on a laboratory two roll mill as per the formulations in Table 3.3.1. The compound was sheeted out from the mill and cut into small pieces and dissolved in toluene to make a 40 percent solution. The solution was kept for two days and stirred vigorously for getting a uniform solution using a high speed mechanical stirrer.
### Table 3.3.1 Formulations of rubber compounds, Pk-1

<table>
<thead>
<tr>
<th>Ingredients</th>
<th>CR AD type</th>
<th>CR W type</th>
<th>NR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neoprene AD</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Neoprene W</td>
<td>0</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>NR</td>
<td>0</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>MgO</td>
<td>5</td>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>Wood rosin</td>
<td>10</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>PF resin</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Accinox TQ</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

#### Wood preparation:

Wood pieces were cut into 25x300x3 mm strips and polished using sand paper grit no.60 (250μm).

#### Wood bonding and testing:

Using a brush the adhesive solution was applied on both pieces to a thickness of 0.1 mm and lap joined for 25x30 mm area of overlap. A load of about 1 kg was placed over the joint and kept for 24 hours. Then the wood joints were kept at room temperature (30°C) and relative humidity (RH) of 50±5 for 7 days.

These wood joints were tested for lap shear strength on a Zwick UTM model 1445 as per ASTM D 906-49 (72).

#### Effect of CNR and PF resin:

The effect of chlorinated natural rubber (CNR) on lap joint strength was measured by varying its amount from 10 to 60 phr in the formulations in Table 1. Similarly the amount of PF resin was varied from 20 to 45 parts and change in lap shear strength was determined.

#### Replacing CR AD type with W type CR and NR:

Adhesive grade neoprene was partially replaced by W type neoprene and lap shear strength was determined. Similarly neoprene AD was replaced by natural rubber and its effect on lap shear strength was studied.
Shelf life of the adhesive:

The neoprene based adhesive was kept for two months and lap shear strength was determined at equal intervals of time in order to determine the shelf life of the adhesive.

Comparison with commercial adhesives:

The adhesive showing maximum bond strength was compared with the commercial adhesives for lap shear strength. The solid content of the adhesive prepared in laboratory was reduced 25 from 40% by adding more solvent to the solution as the commercial adhesives are having 25% total solid content only.

Resistance of the wood joints:

Wood pieces bonded with the adhesive prepared in laboratory and some of the commercial adhesives were kept in cold water (30°C) for one day. Then they were taken out, dried at room temperature (30°C and RH 50±5) for one day and the lap shear strength was determined. Similarly bonded pieces were immersed in hot water at 100°C, in acid (pH 2) and in alkali (pH 10) both at 80°C for one hour. Then the lap shear strength was determined as described earlier. The bonded wood pieces were kept for one month at room temperature and lap shear strength was measured for determining the ageing resistance of the wood bonds were determined.

Direct shear method:

Wood pieces were cut into 60x60x20 mm size, dried and polished using emery paper no.60 (250μm). Adhesives were applied on both pieces to full area to a thickness 0.1 mm. A load of 1kg was placed over it for one day and then it was kept for seven days at room temperature. These wood joints were tested on a direct shear machine and the load required for shearing one piece over the other was measured and the result was expressed as N/cm² by considering the area of the wood pieces. The test was conducted as per IS 2720 (1972) part XIII.

Relationship between solid content and viscosity:

The effect of solid content and viscosity on the adhesive with varying amounts of resin content and rubber content was studied. To the adhesive solution the amount of resin and rubber content varied and the effect of viscosity and solid content was estimated.
Adhesive with different solvents:

The rubber solution with different solvents were also tried. Solvent mixtures were also used to make the adhesives. These adhesive solutions were tried on wood joints and lap shear strength was estimated.

Results and Discussion

Figure 3.3.1 shows the effect of variation of chlorinated natural rubber in the adhesive solutions on the lap shear strength. Lap shear strength increases as the amount of chlorinated natural rubber increases, reaches a maximum and then decreases. In the case of natural rubber solution the highest strength is obtained at 50 phr CNR. This shows that there is an optimum concentration of NR which gives maximum bond strength. At higher concentrations CNR may form the continuous phase and the bond joint loses its elastomeric nature. There is no visible phase separation. The solution is homogeneous in nature. In the case of W type CR based adhesive also the lap shear strength initially increases in reaches a maximum and then decreases similar to that of Neoprene AD type adhesive. But W type Neoprene based adhesive shows lower lap shear strength relative to Neoprene AD. This may be due to the slower crystallizing nature of Neoprene W type. NR based adhesive shows only a marginal increases in lap shear strength.

![Figure 3.3.1 Variation of lap shear strength with CNR content](image-url)
even at 50 phr CNR. This may be due to the low compatibility of polar CNR and nonpolar NR.

In Figure 3.3.2 shows the effect of variation of PF resin in the adhesive solutions on lap shear strength. Lap shear strength increases initially and then decreases. This may be due to the
phase change in the solutions when PF resin becomes the continuous phase. CR AD type, CR W type and NR based adhesives show the same trend when the amount of PF resin is varied.

The decrease in lap shear strength when part of adhesive grade CR is replaced by W type CR is shown in Figure 3.3.3.

![Graph showing variation of Lap shear strength with days of storage](image)

**Figure 3.3.4 Variation of Lap shear strength with days of storage**

![Bar chart showing effect of water on wood joints](image)

**Figure 3.3.5 Effect of water on wood joints**
CR AD type crystallizes much faster than CR W type and so W type CR gives lower strength compared to CR AD type. This figure also shows that the decrease in lap shear strength is more pronounced when adhesive grade CR is replaced by natural rubber. This may be due to the nonpolar nature of NR. Polar adhesives are likely to give higher bond strength with polar adherends like wood.

![Figure 3.3.6 Effect of hot water on wood joints](image)

Figure 3.3.4 shows the variation of lap shear strength of the adhesive with days of storage. The joint strength is found to increase with the length of storage of adhesive solution. It may be

![Figure 3.3.7 Effect of acid on lap shear strength](image)
due to the increase in solid content due to the slight evaporation of solvent and partial precuring of the solution during storage.

The water resistance of joints prepared with the commercial adhesives and the adhesive prepared in the laboratory is shown in Figure 3.3.5. Compared to commercial adhesives the water resistance is found to be superior for the adhesive prepared in the laboratory.

![Fig.3.3.8 Effect of alkali on wood joints](image1)

![Fig.3.3.9 Effect of ageing lap shear strength of wood joints](image2)
Figure 3.3.6 shows the hot water resistance of the adhesives. There is a slight increase in bond strength for some of the joints. This may be due to the slight crosslinking of adhesive film in boiling water. The adhesive prepared in laboratory is found to be comparable to that of commercial adhesives.

Acid and alkali resistance of the adhesives are shown in Figure 3.3.7 and Figure 3.3.8 respectively. The adhesive prepared in laboratory gives better resistance compared to other adhesives.

![Graph showing direct shear strength of different adhesives.](image)

**Fig.3.3.10 Direct shear strength of different adhesives**

![Graph showing variation of Lap shear strength with solid content.](image)

**Fig.3.3.11 variation of Lap shear strength with solid content**
Figure 3.3.9 shows the results of room temperature ageing of the wood joints for one month. Results show an increase in joint strength which may be due to the crosslinking of the adhesive film during storage.

Direct shear strength of different adhesives are shown in Figure 3.3.10. Adhesive prepared in laboratory shows better direct shear strength compared to other commercial adhesives.

![Viscosity variation with solid content](image)

**Fig.3.3.12 viscosity variation with solid content**

![Lap shear strength variation with solid content](image)

**Fig.3.3.13 variation of Lap shear strength with solid content**

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Figure 3.3.11 shows the effect of total solid content on lap shear strength as resin content increases. Lap shear strength increases reaches a maximum and then decreases. The resin forms the continuous phase when used in higher amounts.

![Graph showing the effect of total solid content on lap shear strength.](image)

**Figure 3.3.14 change in viscosity with solid content**

Figure 3.3.12 shows the effect of viscosity change when resin content increases. As the amount of resin increases viscosity increases, reaches a maximum and then decreases and levels off to a particular viscosity. Due to the lower molecular weight of PF resin there will not be any change in viscosity after a particular level of resin. Molecular weight is a factor that directly affects the solution viscosity of polymers.

Figure 3.3.13 shows the effect of lap shear strength of wood joint when the amount of rubber content is varied in solution adhesive. As the rubber content increases the strength increases reaches a maximum and after that slight reduction in lap shear strength is observed. This may be due the large increase in viscosity which decreases the spreadability of the adhesive film and hence makes it more difficult to wet the wood surfaces.

Figure 3.3.14 shows the effect of rubber content in the viscosity of rubber solution adhesive. Viscosity increases significantly as the rubber content increases, as expected, since molecular weight of rubber is very high.
Table 3.3.2 shows the lap shear strength of rubber solutions with different solvents. Results from solvent mixtures are also shown in this table. Toluene/EDC mixer shows the best results.

Table 3.3.2 Lap shear strength (N/cm^2) of adhesives with different solvents.

<table>
<thead>
<tr>
<th>Xylene/Naptha(%)</th>
<th>25/75</th>
<th>28.2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>50/50</td>
<td>29.27</td>
</tr>
<tr>
<td></td>
<td>72/25</td>
<td>34.53</td>
</tr>
<tr>
<td>Toluene/Naptha(%)</td>
<td>25/75</td>
<td>28.16</td>
</tr>
<tr>
<td></td>
<td>50/50</td>
<td>57.22</td>
</tr>
<tr>
<td></td>
<td>75/25</td>
<td>162.68</td>
</tr>
<tr>
<td>EDC/Toluene (%)</td>
<td>25/75</td>
<td>114.5</td>
</tr>
<tr>
<td></td>
<td>50/50</td>
<td>168.71</td>
</tr>
<tr>
<td></td>
<td>75/25</td>
<td>198.65</td>
</tr>
<tr>
<td>Xylene(%)</td>
<td>100</td>
<td>20.23</td>
</tr>
<tr>
<td>Naptha(%)</td>
<td>100</td>
<td>Not soluble</td>
</tr>
<tr>
<td>EDC(%)</td>
<td>100</td>
<td>147.53</td>
</tr>
</tbody>
</table>

Conclusions

1. Adhesive solution prepared with Neoprene AD type is found to give superior bonding than commercial wood adhesives.
2. Optimum amounts of CNR (30%) and PF resin (35%) improves adhesive strength of neoprene based adhesives.
3. Neoprene AD based adhesive solution is found to have better resistance to cold water, hot water, acid and alkali compared to the commercial adhesives.
4. Shelf life of the adhesive is found to be comparable to that of commercial adhesives.

References

Part 4 Novel adhesives based on thermoplastic elastomers for wood to wood bonding

Thermoplastic elastomer shows properties of plastic as well as elastomer\(^1\). Thermoplastic elastomers are widely used in different mechanical applications due to its toughness and also its flexibility\(^2\). They are used in many hot melt adhesives\(^3\)\(^4\). Use of hot melt adhesives need complex mechanism for heating and cooling the adhesive joints. In the case of wood joints heating of the joints is not usually practised.

In this study solutions of thermoplastic elastomers are proposed to be used for bonding. Use of solutions avoid the need for heating and cooling and thus make the process simpler and comparatively cheap.

Experimental

The adhesives were prepared from thermoplastic based on rubber plastic blends. This part of the study is being patented and author request to excuse for not providing the exact details.

Commercial Adhesive used for comparison was Dentrite adhesive supplied by Superchem.

Optical microscopy photographs of the wood adherends were taken before applying the adhesives and after peel off.

Results and discussion

Figure 3.4.1 shows the effect of thermoplastic content in the adhesive on lap shear strength of wood joints. When thermoplastic content increases lap shear strength increases. But when the thermoplastic or the elastomer is used alone the lap joint strength is lower. This shows there is an optimum amount of the thermoplastic and elastomer in the adhesive to
get maximum joint strength for wood pieces. In both types of thermoplastic elastomer studies the same trend is observed.

![Graph showing the effect of thermoplastic content on lap shear strength.](image)

**Fig. 3.4.1 Effect of thermoplastic content in lap shear strength**

Figure 3.4.2 shows the effect of tackifier resin in the thermoplastic elastomer based adhesive for wood joints. When the tackifying resin content increases lap shear strength increases reaches a maximum and then decreases. When the resin content is above an optimum level there may be phases change of the adhesive film to a more brittle nature thus reducing the lap shear strength.
Figure 3.4.3 shows the effect of plasticiser on lap shear strength of thermoplastic elastomer based adhesives on wood bonding. As the amount of plasticiser increases lap shear strength reduces due to the lowering of film strength of the adhesives. When the amount plasticiser increases it reduces the inherent strength of thermoplastic elastomer.

Figure 3.4.4 shows the variation of lap shear strength by changing the viscosity of the adhesives. As viscosity increases lap shear strength increases but after a particular viscosity spreadability of the adhesive film decreases and hence it cannot be handled easily.
Figure 3.4.5 shows the effect of storage of the adhesives for 5 weeks on lap shear strength of wood joints. On storage the lap shear strength of both the adhesives increases slightly which may be due to the evaporation of solvent leading to higher solid content. From this it is clear that adhesives have enough storage life.
Figure 3.4.6 shows the lap shear strength of the wood joints made of two types of thermoplastic elastomer based adhesives and commercial adhesive before and after soaking in cold water. There is slight reduction in lap shear strength in all cases, but commercial adhesives show much more reduction in strength compared to the adhesives based on thermoplastic elastomers.
Figure 3.4.7 shows the effect of hot water on lap shear strength of the adhesives. Adhesives based on thermoplastic elastomers show better retention in strength compared to commercial adhesives. Thermoplastic elastomer-based adhesives show better properties due to the hard domains present in the molecule making it resistant to different environments.

![Figure 3.4.8 Effect of acid on lap shear strength](image)

![Figure 3.4.9 Effect of alkali on lap shear strength](image)
Figure 3.4.8 shows the lap shear strength of the wood joints before and after soaking in hot acids. The adhesives prepared in laboratory show better strength retention compared to commercial adhesives. The chemical resistance of thermoplastic elastomer based adhesives is much better than ordinary elastomer based adhesives.

![Graph showing lap shear strength](image)

**Fig.3.4.10 Effect of ageing on lap shear strength**

Figure 3.4.9 shows the effect of hot alkali on lap shear strength of wood joints. Thermoplastic elastomer based adhesives show superior resistance to alkalis compared to commercial adhesives.

Figure 3.4.10 shows the ageing resistance of the adhesive joints. Commercial adhesive shows lower resistance than thermoplastic elastomer based adhesives. Thermoplastic elastomer based adhesives have better heat resistance compared to other elastomers due the presence of hard domains.

Photograph 3.4.1 shows the roughened wood surface before adhesive application. Photographs 3.4.2 and 3.4.3 show the peeled off surfaces of adhesive prepared in laboratory and one commercial adhesive respectively. From the photographs it is evident that former leaves more polymer residue over the wood surface which may be the cause for more adhesion strength.
Photograph 3.4.1 Roughed wood surface before adhesive application.

Photograph 3.4.2 Peeled off wood surface with laboratory adhesive.
Conclusions

1. Thermoplastic elastomer based adhesives can be used for wood to wood bonding.
2. Optimum amount of tackifier resin and plasticiser can improve the adhesion strength of wood joints with thermoplastic elastomer.
3. The thermoplastic elastomer gives very good storage life and so it can be commercialized.
4. The resistance of this adhesive in different environment like water, acids, alkalies and high temperature etc. is found to be superior compared to commercial adhesives.
5. Optical microscopic studies give evidence for superior strength to thermoplastic elastomer based adhesives.

References