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A Novel Manufacturing Process of Paper Composite using recycled Old Newspaper Sheets and Phenol Formaldehyde Resin

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Abstract: *This novel research work is aimed at finding the beneficiated use of old newspaper as fiber source for environment friendly manufacturing of paper-phenol formaldehyde resin composites. Old newspaper has nearly 60 – 70 percent cellulosic material as natural fibers coming from various fibrous raw materials which have been used for paper manufacturing. The balance 30 -40 percent of news paper comprises inorganic fillers like kaolin clay, calcium carbonate and other additive chemicals like starch, colours, wet and dry strength resins etc. The news paper sheets are cut into proper size to fit in the mould and then both sides are coated with paste of phenol formaldehyde resins. The coated sheets are combined together and treated at high pressure and temperature for different curing times for composite formation. The different conditions have been explored for better properties and economical production. This study will provide a manufacturing process for news paper based composite products which can be used in electrical board sheet, kitchen housing, carcasing, kitchen table, and plastic housing roof. Various process parameters have been varied for getting better properties like the tensile strength of the sheet. The increase in the amount of PF resin increases the hardness of composite to larger extent.*

Key Words: *PF resin, Old newspaper, composites, Fiber, curing, and ash.*

Introduction

Phenol resins are mainly used in the production of circuit boards. They are better known,however, for the production of designed products including pool balls, laboratory countertops, and as coatings and adhesives. In the form of Bakelite, they are the earliest commercial synthetic resin. The P-F resin has various applications. It can be used in resin form as a bonding agent. Subsequently, the liquid resin can be dried and ground into moulding powder, which is usually used in moulding electrical fittings. Buttons, television and computer housing and the other household articles can be manufactured. The main research emphasis is given on the preparation, processing and characterization of P-F resin and moulding powder. For a long time, composite material as a structural concept has existed in various forms of straw-brick, paper and reinforced concrete. However as natural material, it has existed since almost the very beginning, for example, in form of wood and bone. But the polymer composites, presented here, belong to a relatively new group of industrial composites that have been in use since about half a century. By combining the two materials, fibre and matrix, unique properties are tailor made for the specific product. Strength and stiffness are obtained from the reinforcement, the fibres, which can be placed randomly or oriented in both continuous and discontinuous forms. The matrix situated between the fibres is meant to thoroughly surround and bind the fibres to transfer load and protect them against environmental influence. The interface between fibre and matrix is very important for the properties of the composite.

Wahab et al. have studied the adhesion properties of phenol formaldehyde-prepreg oil palm veneers that have potential for plywood manufacture. The veneers were soaked in each type of PF resin for 20 seconds, pressed between two rollers, and pre-cured in an oven maintained at 103 ± 2^{0}C for 24 hours. The bonding properties of plywood made from palm veneers were found to be superior to those of commercial palm plywood [1].

The results of the study done by Joseph et al. reveal that composite with good strength could be successfully developed using banana fibre as the reinforcing agent. The interfacial shear strength (ISS) values are calculated for banana fibre and glass fibre using single fibre pull out test. The interfacial shear strength value is much higher in banana/PF composites than for glass/PF composite. This is due to the hydrophilic nature of cellulose and PF resin. Hydrophilic quality of fibre arises from the hydroxyl groups of lignin and cellulose, which can easily form hydrogen bonds with methylol and phenolic hydroxyl groups of the resole resulting in a strong interlocking between the two. The tensile stress–strain behaviour reveals that the neat PF is brittle. Addition of fibres makes the matrix more ductile. The tensile, flexural and impact properties of the composites are found to be dependent on fibre length [2].

Advantages of the polymer composites are that they are easy in processing, high in productivity, and good in cost reduction. Agricultural waste and annual plants have an alternative of raw material for manufacturing of fibres composite products [3]. The physical properties (specific gravity and moisture content), mechanical property (3-point bending strength), and acoustical property (the sound absorption coefficients) of the composite were determined, to investigate the possibility of rice straw as a partial or complete substitute for wood particles in particleboard. The effects of rice straw size on mechanical properties of particleboards were examined by studying the mechanical properties of particleboards prepared according to rice straw sizes and those prepared by randomly mixing rice straw of different sizes. [4]. Different methods for separation of fibers from wheat straw and rice straw and making paper composites with use of fly ash filler and resins have been investigated [5,6,7,8]. Glass powder filled phenolic composite exhibited higher flexural strength and flexural modulus than the unfilled graphite fibres reinforced phenolic resin composite. Samples were examined using an optical microscope and Scanning Electron Microscope (SEM) [9]. The measurement of impact strength showed that regardless of the cure cycle used, the reinforcement of thermo sets by 30% (w/w) sisal fibres improved the impact strength by one order of magnitude. Curing with cycle 1 (150^{0}C) induced a high diffusion coefficient for water absorption in composites, due to less interaction between the sisal fibres and water [10]. Mechanical characterization (tensile and flexural strength) of woven sisal fabric composites with and without thermal treatment (at 60^{0}C for 72 hr.) on the fabric, thermal characterization by TGA and the manufacturing process by compression moulding had been investigated. Experimental results show that sisal/phenolic composites tensile strength and a flexural strength values 25.0 MPa and 11.0MPa, respectively, independent to the use of sisal fibres with or without thermal treatment [11].

India led the world in terms of newspaper circulation with a massive pile of 374 million newspapers circulated daily (source: Registrar of Newspapers for India). According to the National Readership Survey, India has more daily newspaper than any other nation; out of the world's hundred largest newspapers, twenty are Indian. The demand for newsprint in the country is expected to grow at a rate of 9 percent and India's paper consumption is expected to increase from 2.1 MTPA in 2012-13 to 3.0 MTPA by 2015-16 (source: CRISIL). The Indian newsprint market is characterized with voluminous demand and a high growth rate[12].

Materials & Methods

Materials- In this research Phenol formaldehyde and old news paper samples of the times of India were used to manufacture the thermo set composite. Methanol used as a solvent to dissolve the phenol formaldehyde resin.

Preparation of Composite Sheet

We use the compression moulding machine for making the composite sheet. Newspaper samples impregnated with PF resin were taken in stack using the steel mould having the dimension 105 mm X 150 mm X 3.2 mm. First of all the mould surface was cleaned by toluene and then a fine layer of silicon oil was applied on it for easy removal of composite from the mould. Then the sample was placed in the compression moulding machine. The temperature of compression moulding machine was set around the 120 - 160^{0}C, and pressure applies between 16 - 24MPa (Mega Pascal) for 15minutes to 1 hour. The best conditions were analysed for making paper-resin composites with good strength quality.

The composites were prepared by different relative weight fraction of news paper and PF resin. After stacking the samples, the stack was placed in the mould and pressed in semi automatic hydraulic compression moulding machine at different process parameters.

Experimental steps includes the the following details:

First of all solution of PF resin & methanol is prepared and then coating of news paper sheets with PF resin solution was done. The sheets were dried up in air for one hr time to decrease the sticking quality and then coated sheets were placed in the mould under the different conditions of pressures and temperature by putting in hydraulic press/ Compression moulding machine. When temperature reaches to maximum, curing time was provided. The composite sheet was taken out after 38minutes of time.

Fig. 1: Hydraulic compression moulding machine

Measurements of Important Properties

1. Tensile strength- The test of Tensile Strength was carried out according to ASTM D-638. Tensile strength properties may provide the useful information of the composite material. Tensile properties are also useful in plastic engineering applications of composite, which is used for design purpose.

Fig.2: Tensile testing machine for polymer composites

The obtained results have been used to evaluate the tensile strength, and tensile modulus using the following equations.

Tensile strength = Force at yield / cross section area

Where: stress = force (Newton)/cross section area, strain = change in length/ original length of specimen.

2. Impact strength: Impact strength test was used to measure the impact force, which may be defined as toughness or ability of material to absorb energy during plastic deformation. The test has been carried out according to ASTM D-256.This type of impact testing measures a sample`s relative susceptibility to a pendulum type of loading. The notch depth measures 2.54 mm and must be cut in a carefully prescribed manner. The pendulum is relesed allowed to strike the sample and swing through. The sample must break completely in order for the data to be useful. An energy is taken from the pendulum swing and diveded by the sample thickness. Results are reported in joules.

Sample preparation for izod impact testing:

The dimension of specimen was 50.8 mm X 12.7 mm X 3.175 mm according to the ASTM D-256. The specimen must be notched to provide a stress concentration area that promotes a brittle rather than a ductile failure.

Hardness Test

To perform hardness test, durometer hardness tester of D-type was used. It consists of dial which is marked up to 0-100 scale and indenter. It is a test in which specimen is deformed temporally by indenter. As the indenter penetrates the surface of specimen, the needle on dial move and reading is noted down. The indenter has a specific geometry and resistance to movement. The resistance to movement of the indenter is accomplished through calibrated spring; ASTM D-2240 standards were followed.

Results and Discussion

Mechanical properties of natural fibres are depending on the physical and chemical bond between fibres and matrix. It means the better mechanical properties depend on the bonding between the fibres of newspaper and phenol formaldehyde resin.

The table 1 shows the hardness and thickness of composite samples prepared taking ten sheets of newspaper and coating with PF resin on both the sides of paper. PF resin used was 20% weight fraction of

Table1: Average weight, Thickness and hardness of paper samples

Serial No.	Weight of 10 sheets of paper (g)	Thickness of 10 sheets in (micrometer)	Hardness of composite sheet (0-100 calibrated scale)
1	10.8 ±0.2	800±32	69
2	10.8±0.2	800±34	69
3	10.9±0.2	800±30	71
4	10.8±0.2	800±28	68
5	11.5±0.02	850±36	69
6	11±0.02	850±34	70
7	11.1±0.02	850±30	70
8	10.8±0.02	800±48	68
9	10.5±0.02	800±30	69
10	10.8±0.02	800±28	68

total weight of composite. So 80% weight comprises of old newspaper and provides environment friendly product as news papers are biodegradable.

So such type of composites have potential to replace non biodegradable composite materials and can be used for making boxes, boards and other packaging materials. The figure 3 shows the surface finish and texture at magnification of fifty and hundred times. The smooth and flexible texture is obtained at lower percentage of PF resin.

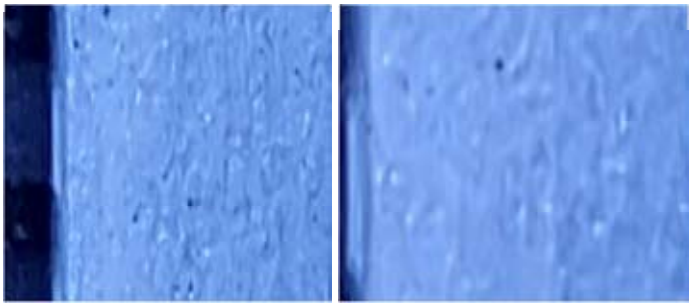

Figure 3: Surface images (with 50X and 100X magnification) of composites with 20% wt fraction of PF resin

Effect of temperature variation on tensile strength of composite: Temperature was varied from 130^{0}C to 180^{0}C, which resulted in tensile strength of the composite increasing from 2.8MPa at 130^{0}C to 3.90MPa at 160^{0}C temperature and then decreased to 3.2 MPa at 180^{0}C. The results showed that maximum tensile strength properties was obtained at 160^{0}C temperature, so this is better temperature condition for manufacturing the high strength composite sheets.

Effect of temperature variation on impact strength of composite: Other experiments showed that with variation of temperature from 130^{0}C to 180^{0}C, the impact strength of composite sheets increased from 8.3 kJ/m2 at 130^{0}C to16.65 kJ/m2 at 160^{0}C temperature and then decreased to 13.49 kJ/m2 at 180^{0}C. The results showed that maximum impact strength properties were obtained at 160^{0}C temperature, so this was better temperature condition for manufacturing of the high strength composite sheets.

Effect of time variation on tensile strength of composite: Again in another set of the experiments, the temperature was fixed at 160^{0}C and time was varied from 15 minutes to 60 minutes to see the effect of time on the strength properties of composite sheets. The effect of time on the properties of the composite sheets manufactured have been studied, Tensile Strength of composite sheets increased and then decreased. We got optimum value at 45 minutes of time.

Effect of time variation on impact strength of composite: The experiments showed that when temperature was fixed at 160^{0}C and time was varied from 15 minutes to 60 minutes, the Impact Strength of the composite sheets increased from 22.85 kJ/m^2 for 15 minutes setting time, to 31.65 kJ/m^2 for 45 minutes time, and then decreased to 28.3 kJ/m^2 for 60 minutes time.

Effect of pressure variation on tensile and impact strength of composite: Now we fixed temperature and time at 160^{0}C, and 45 minutes, and varied pressure from 5 MPa to 28 MPa to see the effect on strength properties of composite sheets. The effects of pressure on the strength properties of composite sheets have been studied; one of the strength properties of composite sheets, According to the linear experiments 160^{0}C temperature, 45 minutes of setting time and 25 MPa is the suitable process conditions for manufacturing composite sheets.

The hardness shown above in table 1 is good for different applications as kitchen and electrical appliances.

Conclusion

The mechanical properties of the composites are influenced mainly by the adhesion between matrix and news paper sheets. Phenol formaldehyde resin is used as flame retardant. Its water absorption property decreases as the percentage of phenol formaldehyde resin increases in the composite. Phenol formaldehyde-paper compositesamples show the good strength properties, so itcan be also used in the interior panelling in the automobile industries. Phenol formaldehyde resin is a good electrical insulating material so the composite can be also used in electrical industries. The composite samples exhibited the good mechanical strength properties like tensile strength, and Impact strength at the 160^{0}C temperature, 25 MPa pressure and 45 minute curing time.

Taking all the parameters into the consideration we came to the conclusion that if we increase the amount of PF, the hardness can also be increased up to the larger extent.

It was also found that the PF comes in the glue form after 100hrs if not stirred. And if it is constantly stirred then it will not be dissolved completely in the solvent.

Future scope: Further scope is there to make the composite sheets which completely recycle the paper waste. Because this is shown experimentally that the sheet can be formed under various combinations of the temperature and pressure. The sheets can be used in the applications as Kitchen table, Kitchen Housing, car casing, Electrical Board sheet, Plastic Housing Roof etc having good strength, recycling quality and thermal and electrical insulation quality.

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