Development of Rubber Pressure Molding Technique using Butyl Rubber to Fabricate Fiber Reinforced Plastic Components based on Glass Fiber and Epoxy Resin

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ABSTRACT: A rubber pressure molding (RPM) technique is developed to prepare fiber reinforced plastic components (FRP) using glass fiber and epoxy resin. The technique is based on the matching die set, where the die is made of hard metal like steel and the punch from flexible rubber like materials. The use of flexible rubber punch helps to intensify and uniformly redistribute pressure (both operating pressure and developed hydrostatic pressure due to the flexible rubber punch) on the surface of the product. A split steel die and rubber punch were designed and fabricated to prepare the FRP components. The same split die was also used to cast the rubber punch. Butyl rubber was used to prepare a rubber punch in this investigation. Burn test, coin test, scanning electron microscopy and mechanical tests like interlaminar fracture toughness, interlaminar shear test, tension test, etc were carried out to know the fiber content, void content, presence of delamination, bonding between fiber and resin, microstructure, and mechanical properties of the composite materials. These properties were also compared with FRP components made by the conventional technique to evaluate its performance in the structural applications. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 1095–1102, 2006

Key words: fiber reinforced plastic; rubber pressure molding; epoxy resin; glass fiber; butyl rubber; mechanical properties

INTRODUCTION

Fiber reinforced plastic (FRP) products are recognized as high tech materials when compared with the conventional engineering materials. These are now regularly used in the stress critical applications, as more and more designers are realizing its high specific strength and stiffness properties. However, the widespread use of these materials is still limited, because of its high cost considerations and partially due to the designers choosing to stick with what they know best. But the casting of FRP components is more difficult than the casting of metal, because the liquid metal has good flowability and can easily flow into the gating channel for filling the mold cavity. During the manufacture of FRP components, polymer being liquid and low viscosity has good flowability, where as the fibers have high stiffness and do not take shape easily over the high curvature of FRP components. Therefore, application of pressure is an important parameter to provide shaping of material before solidification of polymer. Several methods have been developed to manufacture FRP products.[1–12] Some of these are filament winding, pultrusion method, vacuum bagging technique, autoclave technique, matching die set compression molding, resin transfer molding, etc. Among these methods, the autoclave technique is the best method but an expensive process due to the requirement of expensive tooling and disposable bagging materials. At the present scenario, the development of new process for fabrication of FRP components for high tech application is a challenge for scientists/engineers. The objectives of this article are to develop a new process for fabrication of FRP products having a complicated shape and its characterization. In this context, the authors would like to mention that the silicone rubber mold is known for the last few decades and used to make a non-structural item like toys. Again it is costly material. Its use in structural application is not known. Keeping this in mind, to fulfill the afore-mentioned objective, a rubber pressure molding (RPM) technique based on low cost butyl rubber (compared to silicone rubber) is developed. The FRP components were made by both RPM and conventional techniques to evaluate its performance in the structural applications.

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TABLE I
Formulation of Butyl Rubber (phr)

Raw materials	Phr
Butyl rubber	100
Zinc oxide	5.00
Stearic acid	3.00
Carbon black (N330)	45.0
Sulphur	5.00
TMTD	2.50

EXPERIMENTAL

Raw materials

Glass fiber (E-glass, 4-satin) was used for this study, supplied by Harsh Deep Industries, India. Epoxy resin (matrix) and hardener (HY951, curing agent for epoxy resin) used to make FRP components were supplied by Resinova Chemie Ltd. India. Exxon chemicals, USA, supplied butyl rubber (Exxon Butyl 065). The oxidation resistance of butyl rubber is more compared to the natural rubber (or synthetic polyisoprene rubber) and again it is cheaper than silicone rubber. That's why it is used to make a rubber mold in RPM technique. Other rubber chemicals like zinc oxide, stearic acid, accelerator (TMTD), sulfur (curing agent), and carbon black (N330) used to make the rubber mold were received from Avadh rubber limited, India.

Curing tests of epoxy resin for rubber and its other chemicals

There is a possibility of chemical reactions between epoxy resin and butyl rubber including other rubber chemicals like zinc oxide, stearic acid, accelerator (TMTD), sulfur (curing agent), and carbon black (N330). Before making FRP components it is necessary to study the curing behavior of epoxy resin in presence of these earlier mentioned chemicals. These chemicals are generally used to enhance the properties of rubber product. To check, if any of these chemicals is preventing the epoxy resin to cure in contact with the butyl rubber and its other chemicals, a curing study was carried out for the epoxy resin in presence of all these chemicals at a temperature of 25°C for 24 h. It is observed that epoxy resin cures well when in contact with all these rubber chemicals. Similar tests were also conducted to find out the compatibility of butyl rubbers (before curing, i.e., in green form but after mixing of all these rubber chemicals in butyl rubber, and also after curing) with epoxy resin. Epoxy resin cures nicely in all these conditions.

Fabrication of rubber punch

The RPM technique is based on the matching die set, where the die is made of hard metal like steel and punch from flexible rubber like materials. To fabricate an FRP product using RPM technique, a rubber punch is needed and prepared from butyl rubber. The strength of raw rubber is not sufficient to withstand the load applied during the fabrication of FRP components. To enhance its strength, other rubber chemicals like zinc oxide, stearic acid, sulfur, accelerator (TMTD), and carbon black, N330 (reinforcing filler) were used. The formulation used for this study to make the rubber mold is given in the Table I.

Mixing of rubber chemicals

The compounding ingredients (rubber chemicals) were mixed with butyl rubber on a two roll mixing mill at a temperature of 25 to 50°C and friction ratio of 1 : 1.1 according to the ASTM D 3182–89.

Curing and molding

The curing characteristics of the mixed butyl rubber were evaluated at a temperature of 150°C with a Rheometer *R*-100S according to ASTM D 2084–93. Subsequent molding for rubber punch was carried out in a hydraulic press at a temperature of 150°C for 40 min under a pressure of 5 MPa. The butyl rubber punch with split steel die and steel cone is shown in Figure 1.

Fabrication of FRP product

The complicated product selected in this study, which is a component of cooler pump, is usually made of steel sheet of thickness 1 mm. But it usually gets rusted and it is felt that a component of composite pump might be a more appropriate material choice. This component has three important geometry elements: (i) cylindrical, (ii) conical, and (iii) flat surface, as shown in Figure 2. The cylindrical part has an outer diameter of 120 mm and thickness of 1.5 mm; the

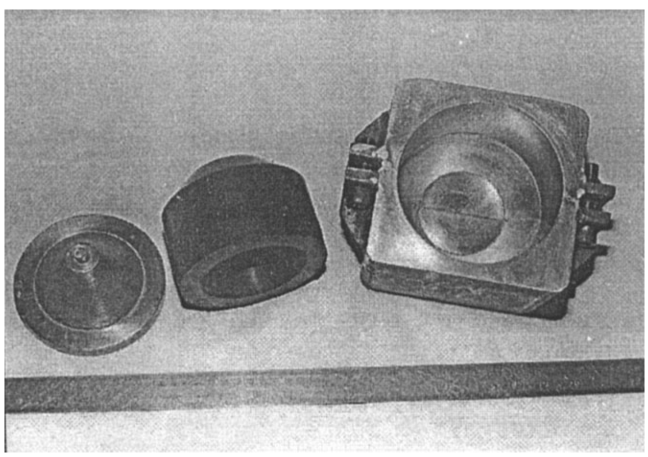


Figure 1 Rubber punch made of butyl rubber.

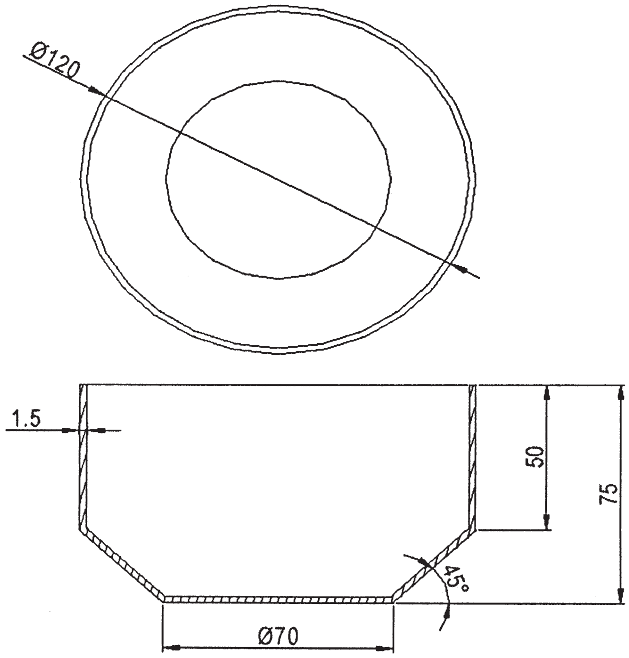


Figure 2 Dimensions of FRP component.

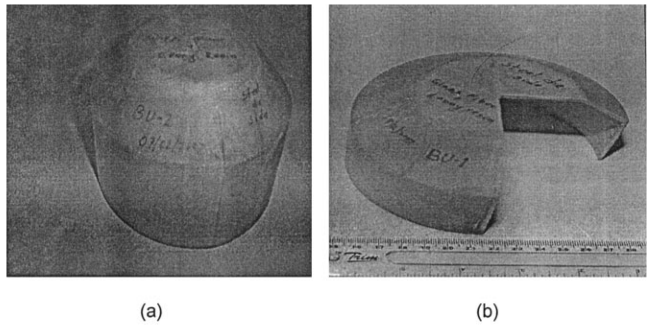


Figure 4 An FRP component (a and b) made of RPM technique using butyl rubber.

conical portion has a half cone angle of 45° and thickness of 1.5 mm; the flat portion has a diameter of 70 mm and thickness of 1.5 mm. The total height of the pump cap is 75 mm. The glass fiber and epoxy resin were used to fabricate this component. Suitably cut pieces of glass fabric were stacked over the steel die by hand lay up technique. The steel die with preform and rubber punch was then loaded into a hydraulic press at a temperature of 25°C and a pressure of 0.5 MPa. After 24 h, the product was taken out and tested. This setup is shown in Figure 3. Figure 4 shows an FRP component made by the RPM technique. Five FRP components were made from each technique (RPM and conventional processes) to evaluate its performance in the structural applications.

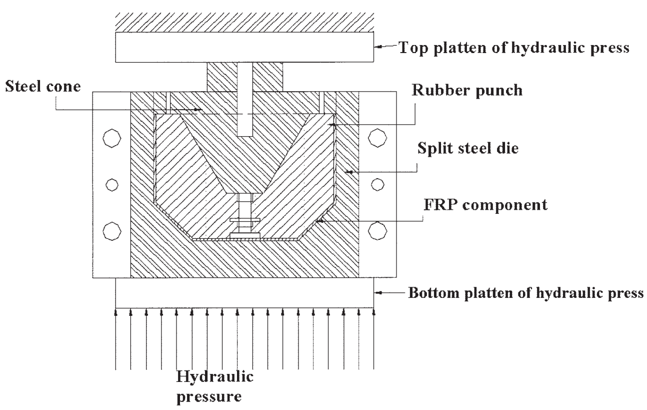


Figure 3 A schematic diagram of rubber molding technique.

Characterization of FRP component

Burn test

In burn test, the volume fraction of fiber and matrix materials and void content of cylindrical, conical, and flat parts of the FRP component were calculated using eqs. (1)–(3).

$$V_f = \frac{\rho_c}{\rho_f} W_f \tag{1}$$

$$V_v = \frac{\rho_{ct} - \rho_{ce}}{\rho_{ct}} \tag{2}$$

$$V_m = 1 - V_f - V_v \tag{3}$$

where V_f, V_v, V_m, ρ_c, ρ_f, ρ_{ct}, ρ_{ce}, and W_f are volume fraction of fiber, volume fraction of void, volume fraction of matrix, density of composite, density of fiber, theoretical density of composite, and experimental density of composites respectively. Procedure to conduct the burn test is reported by Behera[2] and Saha.[12] Five FRP components were made from both techniques to study the volume fraction of fiber and void content in the FRP component.

Coin test

This test gives an idea of delamination in the FRP products. The delamination is checked while tapping a coin on the FRP product. If the sound is like that of metal, that is, high frequency, it ensures good quality of product. Otherwise delamination or high void content may be present in the product.

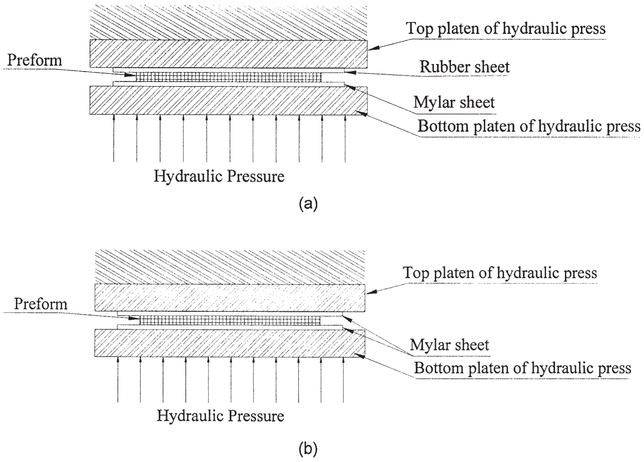


Figure 5 A schematic diagram for fabrication of FRP laminates by (a) RPM and (b) conventional techniques.

Scanning electron microscopy

Studies for wetting characteristics, delamination, and fiber matrix interaction were done by JSM-840 Scanning Electron Microscope, JEOL, Japan. The specimens of size 10 mm × 10 mm were cut from the product and the edges having fiber cross section were smoothened by a waterproof emery paper and 0.3 micron alpha alumina paper.

Interlaminar fracture toughness

The interlaminar fracture toughness test (mode I) was carried out on a double cantilever beam specimen using MTS-810 (100 kN capacity). It is calculated from eq. (4)

$$G_{1,c} = \frac{3}{2}\frac{A_1 A_2^2}{b} \quad (4)$$

where, $G_{1,c}$, A_1, A_2, and b are interlaminar fracture toughness, materials constant for a given specimen, another materials constant for a given specimen, and width of the specimen respectively. The testing conditions are reported by Behera,[2] Kelly and Zweben,[8] and Saha.[12] Five FRP laminates were made from each technique (RPM and conventional processes) for the characterization of interlaminar fracture toughness. The setup for fabrication of FRP laminates used for interlaminar fracture toughness is shown in Figure 5.

Interlaminar shear strength

The interlaminar shear strength was determined by short beam test method using MTS-810 (100kN capacity). It is calculated by eq. (5)

$$\tau = \frac{3F}{4bd} \quad (5)$$

where, τ, F, b, and d are interlaminar shear strength, applied load, width of the specimen, and depth of the specimen respectively. The test setup and testing conditions were followed as per specifications given by Behera,[2] Dornier,[3] Kelly and Zweben,[8] and Saha.[12] Five FRP laminates were also made from each technique for the characterization of interlaminar shear strength. The same setup as shown in Figure 5 for fabrication of FRP laminates for testing of interlaminar shear strength was used here.

Tensile test

The tension test was conducted on MTS-810 (100 kN capacity) machine. The geometry of specimen used for this test and testing conditions are reported by Behera,[2] Kelly and Zweben,[8] and Saha.[12] Five FRP laminates were also made using both techniques for the characterization purpose under tensile mode. The same setup (Fig. 5) for fabrication of FRP laminates needed for tensile testing was used again.

RESULTS AND DISCUSSION

Burn test, coin test, and microstructure studies were performed to check the qualities of products like volume fraction of fiber/matrix, void content, presence of delamination, interaction between fiber and matrix, etc. It is clear from the burn test that the void content in all three parts of FRP component made by the RPM technique is within the range of 3.0%. The differences (i.e., standard deviation) of volume fraction of fiber/matrix, and void content within the component are also very small. Uniformity is maintained in all parts as the volume fractions of fiber are in agreement with all parts of the product. The volume fraction of fiber and void content in the FRP component made by the RPM technique is summarized in the Table II. To evaluate its performance, this complicated FRP components having three geometry elements were also made by the conventional method, where both parts of the mold are made of mild steel. A comparison is made between the products made by the RPM and conventional methods. The fiber, matrix, and void fractions of the FRP components were measured and included in the same Table II. It shows that fiber volume fraction in all these three parts varies from 38 to 43 (∼5% deviation, cylindrical to flat) and void fraction within 2.5 to 4.5, whereas in the RPM technique the deviation from cylindrical to flat portion is within 0.7%. The higher volume fraction of fiber in the FRP components made by RPM technique is due to the hydrostatic pressure developed by the rubber

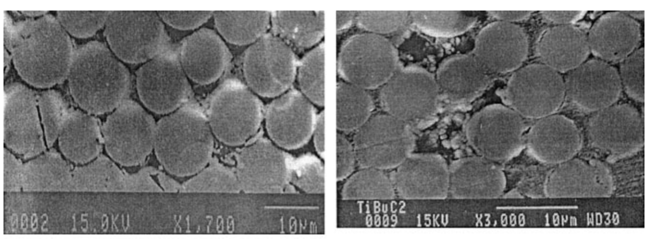
TABLE II
Burn Test Data of FRP Components made of Glass Fiber and Epoxy Resin

Process name	Part name	Volume fraction (%)		Standard deviation of volume fraction	
		Fiber	Void	Fiber	void
RPM using Butyl rubber	Cylindrical	46.3	2.8	0.1	0.1
	Conical	44.9	2.5	0.1	0.1
	Flat	45.3	2.6	0.1	0.1
Conventional	Cylindrical	38.3	3.9	0.4	0.3
	Conical	42.9	4.5	0.3	0.1
	Flat	43.1	2.7	0.1	0.3

mold during fabrication of products. The small deviation of both fiber volume fraction and void content in the RPM technique compared to the conventional process is attributed to the uniform pressure distribution on FRP component during fabrication.

Coin test for FRP components made by the both techniques gives sounds like that of metal, ensuring good quality of product without delaminations. Finally, electron microscopic studies for all portions of FRP product show good interaction between the fiber and matrix in the RPM techniques [Fig. 6(a)] compared to conventional technique [Fig. 6(b)]. It also demonstrates that the void of FRP component is more in the case of conventional process. This information is also observed in the burn test (Table II) discussed in the earlier section. Thus, the FRP products prepared by the RPM technique using butyl rubber punch give better uniformity through out the surface.

The aerospace, automobile, and other industries used FRP components since last four decades. But the biggest drawback, which has been noticed, is its low resistance to delamination. The delamination in products not only leads to complete fracture but also decreases its stiffness, which is a very important design parameter for the designers. In the present scenario, it is a challenge for the researchers to reduce this delaminating behavior of composites to increase its life and load bearing capacity. The parameter "Energy release rate, $G_{1,c}$" suits for studying the crack due to the delamination in composites. Because the crack plane is well defined and the material remains elastic in the vicinity of the crack tip except in the very thin layer of the interface. In the load-displacement curve, a hysteresis loop is found in each cycle and shown in Figure 7. The first cycle is excluded for the calculation of $G_{1,c}$. The first loading cycle in all experiments is observed to be non-linear because of some disturbances like polymer films placed (to create crack) in the mid plane of the specimen sticks to both cantilevers or during the cutting of specimen to the specified size some filler materials stick to the precrack surface. When the machine is switched off after the crack propagation, one can observe the drop of load with time, which indicates that the crack still grows after stopping the machine till self arrest. When the specimen is unloaded to zero load, a small permanent deflection is observed. However, the permanent deflection at zero load is much smaller than the displacement in the loaded condition and its effect is neglected in this analysis. The results of interlaminar fracture toughness are tabulated in Table III. The average value of



(a) (b)

Figure 6 SEM micrographs of FRP component made by (a) RPM and (b) conventional techniques.

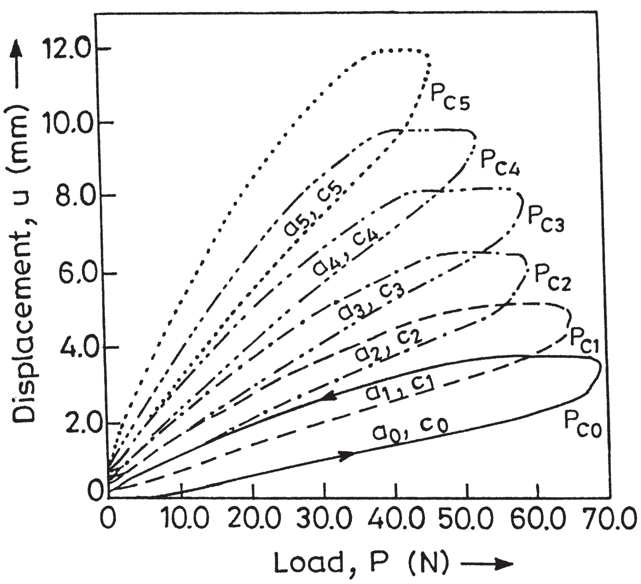


Figure 7 A typical hysteresis loop of FRP components for calculation of interlaminar fracture toughness.

TABLE III
Mechanical Properties of FRP Components

Method of preparation	Inter laminar fracture toughness, $G_{1,c}$ (J/m^2)	Inter laminar shear strength (MPa)	Tensile strength (MPa)	Modulus of elasticity (GPa)	Elongation (%)
RPM using butyl rubber	218 ± 7	60 ± 2.5	353 ± 10	21.0 ± 0.6	2.2 ± 0.1
Conventional method	210 ± 19	57 ± 8.0	312 ± 35	19.0 ± 1.0	2.0 ± 0.1

$G_{1,c}$ for the specimen made by the RPM technique using butyl rubber is 218 ± 7 J/m^2. The fiber volume fraction of the specimen made by the RPM technique using butyl rubber punch is 53%. Whereas the average value of $G_{1,c}$ for the specimen made by the conventional method with volume fraction of fiber 50% is 210 ± 19 J/m^2. The specimens prepared by the RPM technique using butyl rubbers have equal interlaminar fracture toughness compared to the specimen prepared by the conventional method and shown in Figure 8. It is worth to mention that the standard deviation of $G_{1,c}$ for FRP component made by RPM technique is very small within ± 7% (numerical values for five different laminates are 218, 220, 225, 211, 216), whereas for conventional technique it is ± 19% (numerical values for five different laminates are 234, 190, 200, 200, 227). This suggests that RPM technique is the best technique to apply a uniform pressure on the curved surface of FRP component, which is not possible in the conventional technique.

Interlaminar shear strength (ILSS) is also another important material property for the design of laminated composite structures subjected to the transverse loads. The delamination in FRP products can be caused because of the shear stress, as the laminated composites are made of several plies and bonded by the polymeric materials. To find out the suitability of this RPM technique, the ILST is carried out on the specimen with 0° fiber orientation (warp direction) made by (i) conventional and (ii) RPM techniques using butyl rubber punch. The ILST is calculated using eq. (5) using the load and displacement curve as shown in Figure 9. It is included in the same Table III. From this Table it is clear that the interlaminar shear strength of the specimen made by the conventional method (fiber volume 54%) is 57 ± 8.0 MPa and in the RPM technique using butyl rubber (fiber volume 52%) is 60 ± 2.0 MPa. It is clear from earlier values that the specimens prepared by both the techniques have same interlaminar shear strength and is marginally higher (5%) for the RPM technique. It is also shown in the same Figure 8. The standard deviation of ILST in the RPM technique is less (2%) compared to the conventional technique (8%). The numerical values of ILST in conventional process vary from 46 to 67 MPa whereas in the RPM

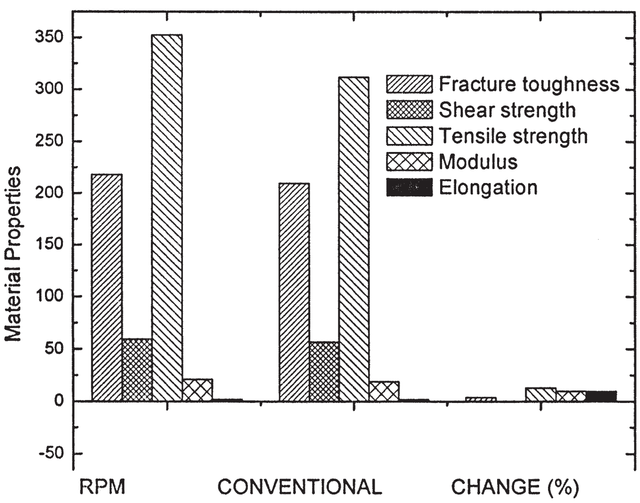


Figure 8 Interlaminar fracture toughness, interlaminar shear strength, tensile strength, modulus, elongation of FRP component made by RPM and conventional process.

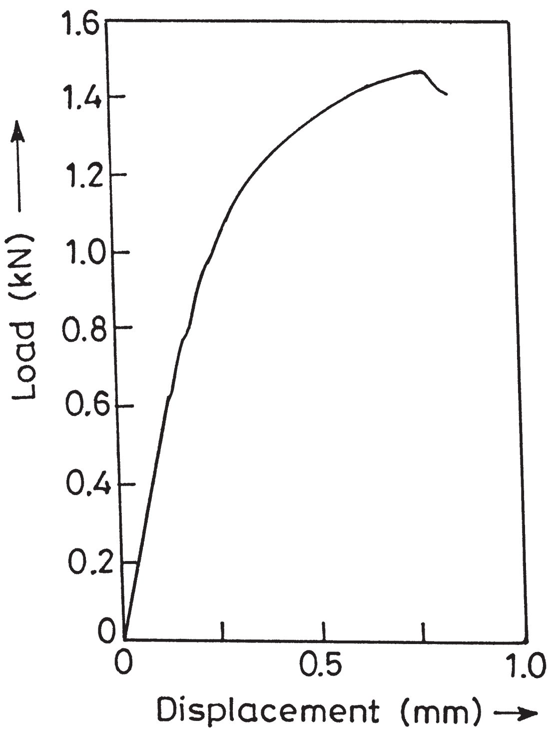


Figure 9 A typical load versus displacement curve of FRP component for calculation on interlaminar shear strength.

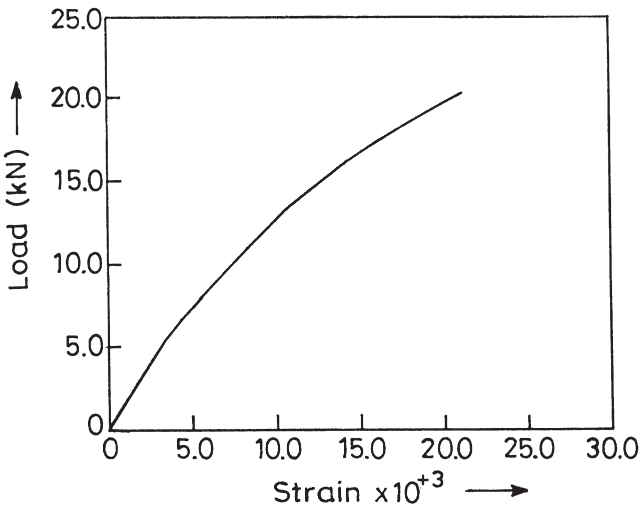


Figure 10 A typical load versus strain curve of FRP component for calculation on tensile strength, tensile modulus, and elongation.

technique it varies from 58 to 63 MPa. This again proves that the pressure is uniformly distributed throughout the curved surface of FRP component and possible only in RPM technique.

Tension test is also conducted on the FRP laminates to evaluate the mechanical properties like tensile strength, elastic constants, percentages of elongation, etc. These properties are very useful for the design and analysis of structure made by composite materials. A typical load versus strain for composite materials made by the RPM technique is shown in Figure 10 (representative). The results of mechanical properties of composite materials made by both techniques with the help of Figure 10 are also included in the same Table III. The specimens are found to fail at the center portion of the specimen and the facture line makes 45° to the line of loading (figures not shown here). It is linear at the low load, but nonlinear at the higher load due to the breaking of fibers. The tensile strength of the FRP specimens with 54% fiber volume prepared by the RPM technique using butyl rubber is 353 ± 10 MPa, whereas the tensile strength of the FRP specimens with 52% fiber volume made by the conventional method is 312 ± 35 MPa. But the tensile elastic modulus of the specimen made by the conventional method is 19.0 ± 1.0 GPa, whereas the elastic modulus of the specimen prepared by the RPM using butyl rubber is 21.0 ± 0.6 GPa. Similarly, the percentage of elongation of the specimen made by the conventional method and RPM technique are (2.0 ± 0.1)% and (2.2 ± 0.1)% respectively. The tensile strength, tensile elastic modulus, and percentage of elongation/strain of FRP components made by both techniques are also shown in Figure 8. It is observed from the measurement of tensile test that the tensile strength, elastic modulus, and elongation of FRP specimens made by the RPM method using butyl rubber is slightly better (~13 to 10%) than that of the specimen prepared by the conventional method. The numerical values of tensile strength in conventional process vary from 277 to 366 MPa whereas in RPM technique it varies from 343 to 361 MPa. This again proves that the pressure is uniformly distributed throughout the curved surface of FRP component and possible only in RPM technique.

CONCLUSIONS

In this article, a process known as rubber pressure molding (RPM) to fabricate FRP components is developed. The technique uses a matching die set, where the die is made of hard metal like steel and punch from flexible rubber like material. Butyl rubber is used to prepare rubber punch. The following conclusions are made from this investigation.

1. Epoxy resin cures well with butyl rubber and all other rubber chemicals like zinc oxide, stearic acid, accelerator (TMTD), sulfur, and carbon black (N330).
2. Burn test, coin test, and microstructure studies through scanning electron microscopy indicate that the FRP products made by the RPM technique using butyl rubber punch has void content less than 3%, free of delamination and better bonding between fiber and resin compared with that of the conventional technique.
3. In interlaminar fracture toughness test, the FRP specimens prepared by RPM using butyl rubber have equal interlaminar fracture toughness compared to the FRP specimen prepared by the conventional method.
4. FRP specimens prepared by the RPM using butyl rubber have same or marginally higher (5%) interlaminar shear strength compared to the specimens prepared by the conventional method in the interlaminar shear test.
5. In tension test, the specimens made by the RPM using butyl rubber have slightly higher value (~10 to 13%) of tensile strength, modulus of elasticity and strain% compared to the specimen prepared by the conventional method.
6. The standard deviation of $G_{1,c}$, ILST, and tensile strength for FRP component made by RPM is very small within (6%) whereas in conventional process it is 35%. This suggests that the pressure is uniformly distributed throughout the curved surface.

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