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P. Jack Bourne

*University of Arkansas, Fayetteville*

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## EVALUATION OF RICE HULLS AS A LIGNOCELLULOSIC SUBSTITUTE IN WOOD PLASTIC COMPOSITES

P. Jack Bourne  
Department of Biological & Agricultural  
Engineering

Faculty Mentor: Dr. Sreekala Bajwa  
Department of Biological & Agricultural  
Engineering

### Abstract:

*Rising raw material costs and shortage of woody materials necessitate alternative sources for lignocellulosic material in wood plastic composites (WPC). This study was conducted to evaluate rice hull (RH), an agricultural residue, as a cellulosic substitute in WPC. Samples were fabricated with approximately 4% zinc stearate, 48% high-density polyethylene obtained from recycled plastics and 48% lignocellulosic material by mass. The composition of the lignocellulosic material was changed from 0 to 100% RH at 20% increments while the remainder was wood flour. The extruded samples were tested for mechanical properties such as specific gravity, water absorption, linear coefficients of thermal expansion, and strengths under compression, shear and bending. The results showed that increasing the proportions of RH to wood flour in the new composite increased the specific gravity but decreased the water absorption. The rice hull rates did not change any of the strength properties. Overall, physical and mechanical properties of the new composite was comparable to that of two of the commercial WPCs. Therefore, rice hull is a viable and renewable alternative for lignocellulosic material in WPC intended for non-structural applications such as decking, fencing, flooring and OEM.*

### Introduction:

Wood plastic composites (WPC) have the advantages of wood like appearance combined with low maintenance and durability for nonstructural applications. Wood flour is used as a lignocellulosic filler in a polymer matrix to form WPC. With diminishing forest resources, pressure from environmental groups and outsourcing of furniture manufacturing, the availability of wood flour has been diminishing. Accordingly, the price of wood flour has been steadily increasing, by as much as 300-500% in the past 5 years. The current reported price of wood flour is approximately 22-27 cents per kg. Demand of WPC has been steadily increasing in the last decade, with a market demand of over 1 billion kg in 2005 (Freedonia Group Inc. 2004). Considering

a typical composition of 50% wood in WPC, the current demand of 0.5 billion kg of wood flour is expected to grow to approximately 0.8 billion kg in 2009 based on the estimated demand of WPC at 1.7 billion kg (Morton et al. 2003). The price of filler (plastic) has been soaring in the past couple of years due to the steadily escalating oil prices. The growth in WPC demand and Although the mounting raw material prices have lowered the profitability margin significantly, the market demand for WPC has been steadily increasing. Therefore, it is imperative to find alternate sources of raw materials including the lignocellulosic filler and polymer in WPC. Agriculture waste is an untapped resource that can provide an inexpensive alternative for lignocellulose in WPC.

Rice hulls (RH) are a renewable agricultural waste that contains natural fibers. Global hull production has been estimated at 100 million metric tons annually, and the material is readily and cheaply available (Oliver, 2004). Rice hulls have been used as a fuel source for residential heating and as an energy source for industrial processes such as rice parboiling operations or electricity generation (Primenergy, 2006; The Agrilectric Companies, 2005). Rice hull is commonly included in animal bedding and used as a fiber source in feed products. Additionally, rice hulls have also been used in sustainable architecture as a loose-fill insulation having low cost and good flammability and insulative characteristics (Oliver 2004). Rice hull ash is suitable for commercial applications within the steel and cement industries. The ash has also been used in more limited quantities for applications as a soil amendment, pressing aid, insulating material, or oil absorbent (Bronzeoak, 2003).

Rice hulls have previously been used for the commercial production of structural plastic composites. Nexwood, a commercially available WPC used approximately 9.1 million kilograms of rice hulls for their WPC production in 2002 alone (Winandy 2004). However, this is not a prominent trend in the WPC industry. There is very little information available in literature regarding the incorporation of rice hulls in plastic

composites, and their impact on the physical and mechanical properties of the resulting composites.

Rice hulls form approximately 20% of the dried paddy on stalk. It is a good source of lignocellulose, with lignocellulose constituting approximately 60-78% of the weight (Luh, 1991). Bulk density of rice hull varies from 0.10 to 0.16 g/ml, and true density ranges from 0.67 to 0.74 g/cm<sup>3</sup>.

The high silica content of the hulls contributes to an effective hardness of 5.5 to 6.5 on the Moh's scale and yields abrasive properties (Luh 1991). As a heat source, rice hulls yield approximately 13.9 MJ/kg which is roughly one-third of that delivered by fuel oil (Cramer et. al. 1991). Chemical constituents of the rice hull are listed in table 1.

**Table 1: Chemical Composition of Rice hulls (Juliano, 1985; Miller and Eisenhauer, 1982; National Academy of Sciences, 1971 as cited by Luh, 1991)**

Component	Content
Moisture (%)	7.6-10.2
Crude Protein (%)	1.9-3.7
Crude fat (%)	0.3-0.8
Crude Fiber (%)	35.0-46.0
Available Carbohydrate (%)	26.5-29.8
Ash (%)	13.2-21.0
Silica (%)	18.8-22.3
Calcium (mg/g)	0.6-1.3
Phosphorous (mg/g)	0.3-0.7
Neutral Detergent Fiber (%)	66-74
Acid Detergent Fiber (%)	58-62
Lignin (%)	9-20
Cellulose (%)	28-36
Pentosans (%)	21-22
Hemicelluloses (%)	12
Total Digestible Nutrients (%)	9.4

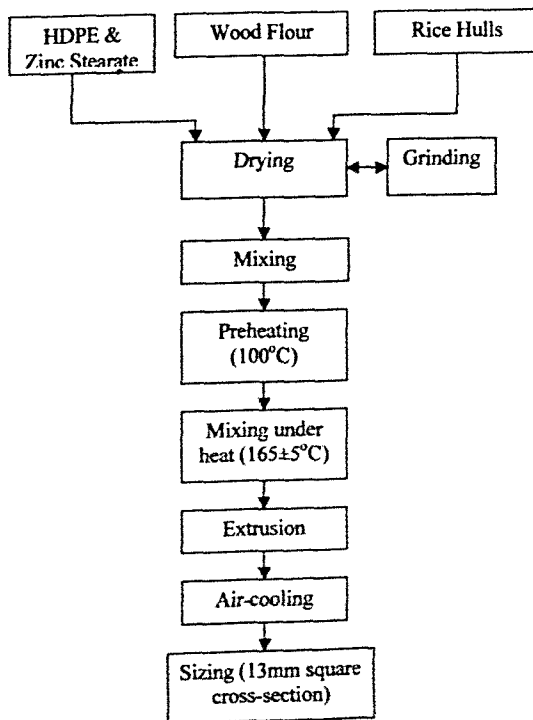
Arkansas is a major rice producing state, which produced approximately 50% of the total US production of 9.4 metric tons of rice each year. Rice hull is a byproduct produced during rice milling, which is available in plenty in Arkansas. Two of the major WPC manufacturers are also located in Arkansas. Therefore, alternate uses of rice hull will have value to the rice producers and processors in Arkansas. WPC manufacturers in Arkansas will benefit from using rice hull as a raw material in WPC production. Therefore, this study was undertaken with the goal of testing rice hulls as a substitute for wood flour in WPC. The specific objectives were to:

- (1) Develop a process for preparing lignocellulosic-plastic composite from rice hulls.
- (2) Evaluate the mechanical properties of this new wood-hull-plastic composite, and compare it with commercially available products.

**Materials and Methods:**

*Sample Preparation*

WPC samples were extruded from a mixture of high-density polyethylene (HDPE), raw or parboiled rice hulls (RH), wood flour and zinc stearate. Recycled plastic pellets were used as source of HDPE. Wood flour used in the study was made from shavings of southern yellow pine ground to a 40-mesh size. Zinc stearate was used as a lubricant. Rice hulls were obtained from Ricelands Inc. in Stuttgart. The experimental design consisted of 6 combinations of lignocellulose, 4% ZnSt and 48% HDPE. The six ratios of wood flour and RH were approximately 0:100, 20:80, 40:60, 60:40, 80:20 and 100:0 by weight. Due to resource constraints, varying numbers of samples were produced for each composition. Additionally, some samples were produced with only raw rice hulls while others contained only parboiled rice hulls. Statistical analysis indicated no significant difference in mechanical or physical properties of lignocellulosic composite samples made with raw and parboiled rice hulls (results not shown). Therefore, samples made with raw rice hulls and parboiled rice hulls were not separated in this study. The number and composition of samples under each treatment is shown in Table 2. For each composition, an adequate number of production runs were made such that the number of samples from separate batches used for each test corresponded to entries in the "Samples per Test" column of Table 2.



**Figure 1: Process Flow Chart**

Table 2: Recipe (compositions) of the lignocellulosic plastic composite.

Treatment	Samples per Test	Ingredients of the lignocellulosic plastic composite (g)			
		ZnSt	HDPE	Wood Flour	Rice Hull
Wood	3	13.3	160	160	0
RH10	5	13.3	160	128	32
RH20	2	13.3	160	96	64
RH30	2	13.3	160	64	96
RH40	2	13.3	160	32	128
RH50	4	13.3	160	0	160

A process was developed to fabricate lignocellulosic plastic composite samples from rice hulls, wood flour and plastic (Figure 1). Rice hulls and 40-mesh wood flour were dried separately in a convection dryer at 60°C until all evaporable water was lost. Rice hulls were then ground until passing through a 2 mm screen, at which time they were returned to the dryer. Lignocellulosic materials were stored in the convection dryer until use. For sample preparation, all ingredients were mixed in the correct proportion and preheated to approximately 100°C. The pre-heated mix was then manually mixed under heat until the material temperature reached 160±5°C. The samples were then extruded into blanks with a square cross-section of 19 mm using an arbor press. The extruded samples were air-cooled. To ensure equivalent surface qualities between batches, air cooled samples were planed on all sides to a final cross section of 13 mm square. Samples were then cut to the desired lengths for testing according to the recommendation of the ASTM standard followed.

The mixing, heating and extrusion chambers were fabricated in-house. The extruder was built from a hollow steel cylinder with an internal diameter of 62.4 mm and length of 103.5 mm. An end cap with the die was welded to one end of the cylinder. The die was fabricated from a seam welded square tube with a length of 23.7 mm and hole size of 19 mm. The inside surface of the die was polished to a fine finish to obtain a clean extrusion surface. Several trial runs were made before extruding the actual samples to ensure smooth operation of the die. The die was cleaned and polished after each batch of samples was extruded.

#### Sample Testing

The extruded samples were tested for specific gravity, water absorption, linear coefficient of thermal expansion (LCTE), bending properties, shear strength, and parallel compressive strength. Specific gravity of the samples was tested according to the ASTM D 6111-97 standard test method for bulk density and specific gravity of plastic lumber and shapes by displacement. Specific gravity tests were conducted as specified in the standard with the exception that measurements were made at 19°C. Water absorption by the samples was tested according to the standard

test method for water absorption of plastics (ASTM D 570-98). Water absorption was measured using a 24 hr immersion test conducted at 19°C. Sample dimensions for both tests were 13 mm x 13 mm x 305 mm.

LCTE was determined using the standard test method for determination of the LCTE of plastic lumber and plastic lumber shapes between -30°F and 140°F (-34.4°C and 60°C) (ASTM D6341-98). Samples were 13 mm x 13 mm x 305 mm and the LCTE was measured in the axial or extruded direction. The actual low and high temperatures used for this test were 26°F and 168°F, respectively.

Flexural properties of the composite samples were determined following the standard test methods for flexural properties of unreinforced and reinforced plastic lumber (ASTM D 6109-97). With the sample depth-to-width ratio less than two, a typical four-point bending test configuration was used. A test jig was constructed in accordance with the above ASTM standard. The load span was 68 mm with a support span of 204 mm. Tested samples had a 13 mm square cross-section with a length of 254 mm. The deflection rate was 6 mm/min. The modulus of rupture (MOR) was calculated as the ultimate strength under flexion, and modulus of elasticity (MOE) was calculated as the slope of the stress-strain curve under flexion, as specified by the ASTM standard.

Shear properties were tested using the standard test method for shear properties of plastic lumber and plastic lumber shapes (ASTM D 6435-99). Samples were tested in double shear in the plane perpendicular to the axis of extrusion. The specified deflection rate of 0.6 mm/min was not possible with available equipment. A deflection rate of 0.75 mm/min was used, as this was the closest realizable strain rate. Samples were tested using a custom-built jig following the specifications in ASTM D 6435-99 standard. The maximum shear strength (MSS) was calculated as the ultimate strength of the samples under shear test.

Parallel compressive properties of extruded samples were determined using the standard test method for compressive properties of plastic lumber and shapes (ASTM D 6108-97). Blanks tested had 13 mm square cross sections and were cut to

25 mm in the axial/extruded direction. Properties were tested along the axis of extrusion on an Instron model 1011 universal testing machine with a loading rate of .03 (mm/mm/min). Platens produced by Instron were used to support the sample. Compressive strength of samples was calculated as the ultimate strength of samples under compression.

The effect of relative proportion of rice hull on physical and mechanical properties of the composite was tested with a general linear model (GLM) procedure in SAS (SAS, 2003). The means of various mechanical properties under different levels of rice hull substitution were compared with Fisher's least significant difference (LSD) analysis. All tests were considered significant at an alpha value of 0.05.

*Sample Comparison*

Mechanical properties of the new composite samples were compared to commercial decking composites, based on the availability of their property data. The two commercial WPCs used were Trex (Trex Company, Inc.) and Rhinodeck (Master Mark Plastics). Properties reported for a 2x6 rice hull-polyethylene composite board produced by Nexwood are also used in comparison (Pacific Lumber Resources). The mechanical properties and standards used for testing these products were obtained from data published on the company's or distributor's websites (Trex Company, Inc. 2005, Master Mark Plastics 2004, Pacific Lumber Resources, 2006). Trex produces a WPC lumber

composed of approximately 40%-50% HDPE and 50%-60% wood fiber (Trex Company, Inc. 2003). Master Mark's Rhino Deck product line contains 30%-50% HDPE, 50%-65% wood flour (Master Mark Plastics, 2004). Nexwood's composition is approximately 60% rice hull and 40% polyethylene (Pacific Lumber Resources, 2006).

**Results and Discussions:**

The lignocellulosic composite having wood flour substituted with rice hull at varying proportion exhibited light weight with specific gravity lower than unity (Table 3). Specific gravity of the new composite varied from 0.95 to 0.99, and as the relative proportion of rice hull in the composite increased, its specific gravity increased. A low specific gravity is preferred for building material applications. When the new composite was compared to commercially available composites, its specific gravity fell within the range of reported values for commercial products. Specific gravities of the new product were all greater than that of both Nexwood and Rhino Deck composites.

Water absorption is an important property for composite materials that are exposed to environmental extremes. A lower water absorption rate is preferred as water absorption can lead to decay and shortened life of cellulosic materials in the composite. Water absorption rate of the new sanded composite samples varied from 1.1 to 2.4 % (Table 3). Mean water absorption for

Table 3: Mechanical properties of the produced lignocellulosic composite material developed from rice hulls, in comparison to two commercial WPC materials.

Composite material	Sp. Gr. at 19/19°C	Water Abs. (%)	LCTE (µm/m <sup>2</sup> C)	MOE (MPa)	MOR (MPa)	MCS (MPa)	MSS (MPa)
Wood	0.960 <sup>ac</sup>	2.2 <sup>a</sup>	54.61 <sup>a</sup>	931 <sup>a</sup>	9.29 <sup>a</sup>	12.2 <sup>a</sup>	5.22 <sup>a</sup>
RH10	0.977 <sup>b</sup>	1.8 <sup>b</sup>	48.85 <sup>a</sup>	1057 <sup>a</sup>	10.76 <sup>a</sup>	11.6 <sup>a</sup>	5.05 <sup>a</sup>
RH20	0.973 <sup>bc</sup>	1.6 <sup>bc</sup>	62.59 <sup>a</sup>	836 <sup>a</sup>	9.86 <sup>a</sup>	12.1 <sup>a</sup>	4.94 <sup>a</sup>
RH30	0.975 <sup>bc</sup>	1.8 <sup>b</sup>	59.79 <sup>a</sup>	1099 <sup>a</sup>	12.39 <sup>a</sup>	15.9 <sup>a</sup>	6.94 <sup>a</sup>
RH40	0.979 <sup>bc</sup>	1.5 <sup>bc</sup>	64.58 <sup>a</sup>	1163 <sup>a</sup>	15.78 <sup>a</sup>	15.5 <sup>a</sup>	6.90 <sup>a</sup>
RH50	0.989 <sup>b</sup>	1.2 <sup>c</sup>	62.28 <sup>a</sup>	793 <sup>a</sup>	12.17 <sup>a</sup>	13.3 <sup>a</sup>	7.53 <sup>a</sup>
Trex	0.91 to 0.95	4.3-sanded 1.7-unsanded	28.98 to 34.56	1207	9.81	12.5	3.87
Rhino Deck	1.08	N/A	35.82	N/A	33.46-edge 21.79-flat	N/A	9.58
Nexwood	1.178	<.8%	48.7	2780	20.2	13.6	N/A
ASTM standard	D6111 <sup>1</sup> D2395 <sup>2,3</sup> D4442 <sup>3</sup> D792 <sup>4</sup>	D570 <sup>1</sup> D1037 <sup>2,4</sup>	D6341 <sup>1,2,3,4</sup>	D6109 <sup>1,4</sup> D4761 <sup>2</sup>	D6109 <sup>1,4</sup> D4761 <sup>2,3</sup>	D6108 <sup>1,4</sup> D198 <sup>2</sup>	D6435 <sup>1</sup> D143 <sup>2,3</sup>

- <sup>1</sup>Rice Hull Samples with Postscript Denoting Hull Content as Percent by mass, <sup>2</sup>Trex, <sup>3</sup>Rhino Deck, <sup>4</sup>Nexwood.
- <sup>a,b,c</sup> Different letters in a column indicate significant difference (P <0.05) between the treatments.

each treatment dropped from 1.8 to 1.2% as the rice hull content was increased from 10 to 50%. This was significantly lower than the water absorption rate of samples with no rice hull, which was 2.2%. All the samples tested in this experiment had superior water absorption property compared to the Trex product. The reported water absorption rate for Trex is 1.7% for unsanded and 4.3% for sanded samples. The reported value for Nexwood was less than .8%. Our samples did not contain any water phobic additives that are commonly used in commercial samples to decrease water absorption. The incorporation of water phobic additives in WPC can further reduce water absorption substantially. Additionally, water absorption for sanded products is significantly higher than unsanded samples as the plastic coating protects the sample from water in unsanded samples.

Linear coefficient of thermal expansion expresses the temperature stability of the composite material. This is an important property if the material is intended for use in outdoor environments, as the expansion and compression under extreme weather events would lead to failure of joints. The mean LCTE value of the samples under different rice hull composition varied from 49-65%, which was much higher than reported values of Trex and Rhino Deck and was somewhat higher than that reported for Nexwood (Table 3). A comparison with the samples extruded under similar conditions with no rice hull indicated a high LCTE for these samples as well. A statistical analysis of means with Fisher's least significant difference (LSD) indicated no difference in LCTE between different rice hull treatments. For these reasons it is presumed that the differing production processes may have affected the LCTE.

The MOE, determined using the 4-point bending tests indicates the toughness of the composite material. MOE for the new composite varied from 793 to 1163 MPa. The MOE of the new composite is lower than both Trex (1207 MPa) and Nexwood (2780 MPa). However, it is believed that this property can be increased significantly by using higher extrusion pressure than were achievable with the arbor press used. This is also supported by the fact that the composite samples produced with the same

process with no rice hull also showed a low MOE of 931 MPa. Analysis of mean MOE for each treatment showed no significant difference in MOE between various rice hull treatments.

The ultimate strength properties (ultimate strengths in parallel compression, transverse shear, and pure bending) of the composite samples were comparable to reported values for commercial WPC products (Table 3). The strength properties of the new composite showed numerical increase with increasing proportions of rice hull. The average MOR of the new composite samples varied from 9.86 to 15.78 MPa for samples with 9.6-48% rice hull, which was significantly higher than the Trex MOR of 9.8 MPa. Mean parallel compressive strengths varied from 11.6 to 15.9 MPa. The compressive strength of both Trex and Nexwood were within the range for the new composite. Shear strength showed significant increases with the amount of RH average values varying from 5.05 to 7.53 MPa. The shear strength of the new composite was superior in its shear strength properties compared to Trex (3.5 MPa). It, however, must be noted that shear strength presented by both manufacturers are in the longitudinal plane whereas shear strength tests of RH samples were conducted in the transverse plane.

Analysis of variance (ANOVA) on measured properties of the new composite samples with respect to the relative proportion of rice hull showed that percentage rice hull significantly influenced both specific gravity and water absorption (Table 4). Approximately 55% of variability in specific gravity and 78% of variability in water absorption was explained by the relative proportion of rice hull in the new composite.

It is clear from the comparison between the physical and mechanical properties of the new composite with various proportions of rice hull and the commercial products that lignocellulosic composites made from rice hull can be reasonably be offered as an alternative for the currently available WPCs in the market. Most of the market available WPCs are used for non-structural applications where high strength properties are not required. The new composite we have developed can be used for

*Table 4. Effect of rice hull substitution on various physical and mechanical properties of a lignocellulosic composite with 48% plastic. The rows with bold-italic numbers indicate that substitution with RH significantly influenced the specific property.*

Property	Model DF	Mean	RMSE	p-value of treatment	R <sup>2</sup> value
Specific gravity	5	<b><i>0.98</i></b>	<b><i>0.0099</i></b>	<b><i>0.0584</i></b>	<b><i>0.55</i></b>
Water absorption	5	<b><i>1.68</i></b>	<b><i>0.22</i></b>	<b><i>0.0013</i></b>	<b><i>0.78</i></b>
LCTE	5	31.21	8.19	0.4773	0.28
MOE	5	140574.1	54533.36	0.8126	0.15
MOR	5	1663.33	738.51	0.7816	0.17
Shear Strength	5	874.97	269.75	0.3545	0.33
Compressive Strength	5	1888.64	595.62	0.7607	0.18

non-structural applications such as decking, fencing, flooring and OEM applications. It has to be noted that we have employed an extruder developed inhouse to manufacture the samples. The pressure and temperature controls of this extruder were not as accurate as that of a commercially available extruder. Similarly, the extrusion pressure was much lower than that of a commercial extruder. Therefore, it is possible that samples generated with similar proportion of rice hull in a commercial extruder will have significantly high strength properties due to the high proportion of silica in the hull. Similarly, it may also exhibit higher specific gravity. The major drawback of using rice hulls in lignocellulosic composite materials is its high abrasiveness, which increases the wear of the extruder.

### Conclusions:

A study was conducted to test whether raw or parboiled rice hulls could be used as a lignocellulosic substitute in wood-plastic composites. A typical WPC composition of 48% wood, 48% plastic and 4% Zinc stearate was used for the study. Wood flour was substituted by RH in increments of 20% by weight, varying from 0% (no RH) to 100% RH. The composites exhibited low specific gravity and water absorption, two critical properties for WPCs. The rice hull in the composite tended to increase the specific gravity and decrease water absorption. The new composite was superior to commercial products in its water absorption, mainly due to the hydrophobic nature of rice hull. Properties such as specific gravity, LCTE, MOR, compressive strength and shear strength of the new composite was comparable to that of commercial WPC products. With better temperature and pressure control in the extrusion process, the properties of the new composite could improve significantly. In conclusion, rice hull is a renewable and viable alternative source of lignocellulosic material, and could be successfully used in lignocellulosic-plastic composites. While handling and processing of the rice hull will be similar to wood flour, its use could prove desirable to WPC manufacturers in rice producing regions.

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### Mentor comments:

Dr. Skreekala Bajwa recommended her student's work highly. In her letter in support of publication of Mr. Bourne's work, she said,

I was excited to nominate the article 'Evaluation of rice hulls as a lignocellulosic substitute in wood plastic composites' for publication in *Inquiry*. This article provides a solution to an environmental problem faced by the crop processing industry in Arkansas. Although it is known that rice hulls could substitute for wood in wood plastic composites, there is no

published data on the properties of such composites. This research provides valuable information to researchers and the wood plastics composite industry while providing a value-added product to the rice processing industry for their waste stream. The wood plastic composite industry is a fast growing industry with significant presence in Arkansas.

I would attribute the success of this project primarily to Jack's motivation, and to a well-planned research program. Jack was a quick learner and a pleasure to work with. He took courses on mechanics of materials, and mechanical design at the same time he was working on the project. He was excited to see practical applications of the theory he was learning in those courses. I was impressed by his quick intellect, sincerity towards the project, and the extra measures he took to ensure reliability of data generated. He performed Master's level research, and it is the first time in my five-year career that I have come across a student of such high caliber.