Influence of Moisture and Low Temperature on Notched Izod Impact Toughness in a Pultruded Reinforced Composite

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ABSTRACT

A preliminary assessment was made of the influence of low temperature on the impact-generated fracture of a commercial glass-reinforced polymer composite produced by the pultrusion process. Impact tests were performed using an Izod testing machine on the composite specimens with a V-notch resulting in a fracture surface parallel to the fiber direction. Tests were conducted at 25º, −5º, −25º, and −50ºC on both dry (as received) and wet (submerged for 12 hours) specimens. This paper reviews the existing data in the literature on low temperature fracture behavior of composites and compares the data obtained from these tests. Special emphasis was given to the influence of subzero temperatures on fracture behavior.

KEY WORDS: composites, moisture, low temperature, notched impact, Izod, fracture

INTRODUCTION

Composite materials are becoming very prominent and popular in the commercial sector. Manufacturers of fiber reinforced plastics (FRPs) are producing all kinds of bars, plates, and even structural shapes that are intended to compete with more traditional structural materials such as wood and steel. These new composites have several advantages over the traditional materials: composites are very light, have high strength-to-weight ratios, and are extremely stiff. As more of these FRPs are put into service, they will be required to perform in extremely adverse environmental conditions. Yearly temperature variations of 80°C or more, very dry to very humid climates, mechanical cycling, and physical abuse are contributing factors to the performance degradation over time. The question that arises then is how will these materials behave/perform under these extreme conditions?

It is well understood that internal residual stresses exist in a fiber reinforced composite due to mismatch in the coefficients of thermal expansion between the fiber and the polymer matrix. These residual stresses exist at room temperature, and further cooling only increases these stresses, thus increasing the potential for microcrack development. Lower temperatures also increase the strength and stiffness of the matrix (Dutta et al, 1993, 1994a, 1994b). The absorption of moisture at room temperature causes the matrix to swell, thus reducing the internal residual stresses. Upon cooling, this absorbed moisture freezes, expands, and thus increases internal strains. Any existing or newly formed cracks or notches only serve to complicate the problem.

The objective of this study has been to determine the effects of moisture and reduced temperature on the parallel-to-the-fiber Izod notch toughness of a commercially available fiber reinforced plastic (FRP). This information provides an assessment of the fracture toughness of the material along the fiber direction.

BACKGROUND

Notched Impact (Charpy/Izod)

Although a wealth of data exists regarding the impact properties and energy absorbing abilities of polymer composites during fracture, only a few investigations have been undertaken to explore the effects of reduced temperatures on this behavior. One of the more comprehensive studies on low temperature impact properties of composites was performed by Voelker (1991), who conducted notched Izod impact tests on four thermoplastic composites reinforced with long glass fibers, at temperatures ranging from 22º to −31.7ºC. These values were then compared to those in the literature for similar short glass fiber composites. The results, shown in Tables 1 and 2, indicate that the effect of low temperature on impact behavior depends on the material system. In all cases, an increase in content and length of glass fibers resulted in an increase in impact strength at all temperatures. However, the type of resin used had a significant effect on the influence of low temperatures. In general, the long fiber reinforced nylon composites showed a decrease in impact strength with decreasing temperatures, with reductions of 7 to 16% at −31.7ºC relative to room temperature (22ºC). However, the other long fiber reinforced polymer resins [polypropylene (PP), polyethylene terephthalate (PET), and styrene maleic anhydride (SMA)] showed increases in impact strength ranging from 7 to 28% at −31.7ºC relative to room temperature. As is evident in Table 2, with the exception of polypropylene, short fiber reinforced composites exhibited a decrease in notched Izod impact strength from 20º to −40ºC.

Dutta et al (1994a) conducted a similar study to determine the influence of low temperatures on the impact strength of a composite made of recycled plastics reinforced with wood fiber. Charpy impact tests were conducted on 82.5-mm-long specimens with 25.4 by 25.4-mm cross sections containing a 45º notch of 5-mm depth, at temperatures ranging from 50º to –60ºC. The results, shown in Table 3, reveal a dramatic drop in the absorbed impact energy between 50º and 24ºC, followed by a gradual decline in absorbed energy from 24º to –60ºC. In contrast to these findings, Nishijima and Okada (1982) reported mixed results in their study of the influence of low temperatures on the Charpy impact strength of cloth reinforced epoxide resin. Charpy impact tests were performed on unreinforced samples of epoxide resin, as well as samples reinforced with woven E-glass fiber cloth and samples reinforced with woven carbon fiber cloth. Tests were conducted at room temperature (RT) and liquid nitrogen temperature (LNT), and the effect of fiber orientation was evaluated by cutting the specimens at various angles (0º, 15º, 30º, 45º) with respect to the weave orientation. Although the impact strength of the unreinforced epoxy decreased significantly from RT to LNT (approximately 40 to 65% reduction, depending on impact velocity), temperature had a varying effect on the reinforced specimens, depending on the fiber orientation. Approximate values of impact strength from this study are shown in Table 4. As is evident, the LNT values were higher than RT values in some cases, but lower in other cases. Nishijima and Okada attributed these variations to differing failure mechanisms that are dominant as the fiber orientation changes, and which are affected differently by changes in temperature. In yet another study of Charpy impact strength at cryogenic temperatures, Wigley (1971) reported almost no change in impact energy from 22º to –196ºC for a high modulus, carbon fiber/epoxy composite.

Unnotched Impact and Fracture Energy

A number of additional studies have been undertaken to investigate the influence of low temperature and moisture content on the unnotched impact strength and energy absorbing abilities during fracture of composites. Karbhari and Pope (1994) tested resin transfer molding specimens made of epoxy resin reinforced with layers of chopped and continuous strand glass mat. Specimens were impacted with a Dynatup drop-weight impactor after being subjected to various environmental conditions, such as water immersion, freezing in water, and low temperature conditioning. The samples were then tested in flexure to determine the post-impact flexure strength. Competing effects were found in their study. For example, the addition of moisture appeared to strengthen the composite after impact, relative to the dry condition; however, subsequent freezing of the water caused further strength degradation due to expansion. When conditioned at –17.8ºC, the composite exhibited improved impact and flexural strength. Fiber architecture also had a significant effect.

Dutta et al (1994b) performed dynamic compressive tests to induce diametral tensile splitting in a pultruded glass fiber composite. The high strain rate tests were performed using a split Hopkinson bar machine, and the energy absorbed during fracture was calculated. Tests were performed on dry, wet, and thermally cycled (100 cycles at ±60ºC) specimens at temperatures of 24º, –5º, and –40ºC. The results, shown in Table 5, indicate a general increase in the energy absorbed during fracture with decreasing temperature, due in part to an increase in strength at reduced temperatures. The influences of moisture content and thermal cycling on the energy absorbed during fracture at a given temperature were mixed: beneficial in some cases, detrimental in others.

In a separate study, Dutta et al (1993) performed short-duration impact tests on graphite/epoxy (AS4/3502) laminated plates using a modified Hopkinson pressure bar. By measuring incident, reflected, and transmitted stress waves, the force-displacment relationship and absorbed energy were determined. Tests were conducted at 21º and –31ºC for impactor velocities ranging from approximately 2.5 to 6.6 m/s. The existence of a "transition velocity" for energy absorption was reported. Below the transition velocity, less energy was absorbed during impact at –31º than at 21ºC. Above the transition velocity, the opposite occurred.

Finally, in a study of mixed-mode delamination fracture in graphite/epoxy composites, Russell and Street (1985) measured the fracture energy during delamination for unidirectional specimens at –50º, 20º, and 100ºC, using a variety of specimen designs. Although the unreinforced matrix exhibited a significant decrease in fracture energy with decreasing temperature, the graphite fiber reinforced material exhibited increases ranging from roughly 25 to 35% over the temperature range from 100º to –50ºC, depending on the specimen type. The authors argued that the results suggest residual stresses and constraint effects play a dominant role in the influence of temperature on delamination behavior.

Based on the available results, it is evident that there are no definable trends regarding the influence of low temperature on the impact strength or fracture energy of polymer composites. Reduced temperatures may result in an increase, decrease, or minimal change in the impact or fracture energy. Other variables, such as fiber length, fiber volume fraction, fiber orientation, moisture content, etc., may contribute to the varying temperature effect by influencing the failure mechanisms and residual stresses in the composite.

EXPERIMENTAL PROCEDURES AND RESULTS

As already noted, the purpose of this study is to determine the effects of moisture and temperature on the parallel-to-the-fiber notch toughness of the chosen FRP. The material selected for this study is a pultruded glass-reinforced polymer composite that utilizes a fire retardant isophthalic polyester resin with a UV inhibitor. The material has a fiber volume fraction of 0.59 and an average density of 2048 kg/m^3 (0.074 lb/in^3). All samples used in the study were cut from 3.81-cm (1.5-in.) square bar stock.

The test matrix is a 2x4 matrix with two levels of moisture and four levels of temperature. The two moisture levels are as received, or what is referred to as DRY in this study, and WET which represents a moisture content of roughly 0.18 percent by weight. To achieve this moisture content, the machined specimens were first stored under vacuum for 24 hours, and then immediately submerged in water and soaked for 18 hours. The four temperature levels are +25º, –5º, –25º, and –50ºC. At each temperature and moisture combination 15 samples were tested for a total of 120 test specimens.

Test specimens were cut from square bar stock as shown in Figure 1(a). A diamond-tipped bandsaw blade was used to cut the specimens from the bar stock and a 90-degree end mill was used to produce the notch. The critical dimension on the notch is the distance from the base of the notch to the opposite side of the specimen; this dimension was kept to 1.118 ± 0.005 cm (0.440 ± 0.002 in.), see Figure 1(b). The length of the notch varied and was recorded for each specimen, so that the results could be presented on an energy per unit area basis.

All specimens were conditioned at their respective test temperatures for a minimum of 24 hours. To minimize the effects of heat absorption during the actual testing, the reduced temperature experiments were conducted in one of two coldrooms. Each room was maintained at a set temperature, one at –5º, the other at –25ºC. Since a –50ºC room was not available, these specimens were conditioned in a cooling chamber, then sealed in a thermal canister, and transported to the –25ºC room for immediate testing.

The results of the experiment are presented in Figure 2. Several observations can be made regarding these results. At room temperature the mean notch toughness of the wet specimens was slightly (~15%) higher than the mean value of the dry specimens. This is possibly attributable to the reduction in the internal residual stresses in the matrix due to the matrix swelling when moisture is absorbed.

At –5º and –25ºC the mean energy is essentially the same for both the dry and wet specimens, this value being the dry room temperature value. This observation indicates perhaps that the beneficial effects of moisture, when introduced at room temperature, are negated when the absorbed moisture freezes. However, a noticeable increase in Izod energy occurs for both test groups at −50ºC. Compared to the mean dry room temperature value, Izod energies at −50ºC increase by roughly 20 percent. This is consistent with the findings of Voelker (1991).

ANALYSIS AND DISCUSSION

Test Results

As already noted, the swelling of the matrix caused by absorbed moisture at room temperature tends to reduce the internal residual stresses, thereby increasing the notch toughness. In addition, there appears to be a slight reduction in the mean notch toughness of the wet specimens at −5ºC, relative to 25ºC, while the dry specimens show no such trend. It is believed that this is due to the expansion of the absorbed moisture that occurs near the freezing point. The expansion of absorbed moisture upon freezing results in an increase in the internal pressure around the water-filled cracks and voids, thus reducing the energy required for fracture to occur.

As the temperature is reduced further to −50ºC, both sets of specimens show an increase in notch toughness values. Two phenomena contribute to this behavior. The first addresses the wet specimens and deals with the physical properties of ice; that is, at temperatures below approximately −6ºC the density of ice increases slightly with decreasing temperatures. This decrease in ice volume with decreasing temperature is very small and does not independently explain the roughly 20 percent increase in notch toughness values as observed relative to room temperature values. The second phenomenon pertains to both sets of specimens and concentrates on the well-documented increase in strength and stiffness exhibited by polymers at reduced temperatures. It is believed that the increased strength and stiffness of the matrix at lower temperatures (below −20ºC) becomes much more influential on impact energy values than does the presence of moisture within the specimen. The increased strength and stiffness of the matrix at temperatures below about −20ºC becomes dominant, decreasing the importance of absorbed moisture on notched impact strength, and resulting in higher overall notched impact energies.

Possibly more important than the observations made regarding mean value tendencies are observations regarding the variance or scatter between the dry and wet groups at various temperatures. It is clearly evident that the introduction of moisture into this material increases the scatter significantly. The observed scatter is consistent, essentially paralleling the mean curves for each moisture group.

One possible explanation for this type of scatter is material variability. Figure 3 is an image of a cross section of the composite showing the nonhomogeneous nature of this material; i.e., the fiber spacing is not always consistent, resulting in regions that are rich in resin and others that are rich in fibers. Depending upon where the notch is, either adjacent to a resin-rich area or a fiber-rich area, the notch toughness values will change. Further, if the moisture influences the resin-rich areas to a different extent than it does the fiber-rich areas, it would be expected that the presence of moisture in the composite would result in an increase in scatter in notch toughness values.

It should be emphasized that it is the lower bound information on tests like these that are critical to designers. Consistently focusing on mean values, whether it be tensile strength, yield strength, or notch toughness, can result in nonconservative designs.

Statistical Analysis

To validate the observations made concerning the graphical analysis, a statistical analysis was conducted to measure the importance and effects of moisture and temperature on Izod notch toughness. The experimental design chosen was a two-factor, completely randomized, full factorial design with multiple replications. Both factors, temperature and moisture, were treated as fixed factors. Temperature was set at the four different levels (−50º, −25º, −5º, and 25ºC) and moisture at two levels (dry and wet). To allow accurate estimation of experimental error, and enhance the accuracy of main and interaction effects, the experiment was conducted with 15 replications.

Initially, when a complete general linear model (GLM) was entertained (one that included two main effects for the temperature and moisture and a first order interaction term between the two factors), an analysis of variance (ANOVA) procedure, along with the respective statistical tests, revealed that there is no significant interaction between temperature and moisture at 10% Type-I error. The results obtained from the GLM procedure for the reduced model (a model without the interaction term) are shown in Table 6. Based on the evidence, it is clear that the temperature main effect is strongly significant (statistically) at 10% Type-I error and that the moisture main effect is marginally significant (statistically) at 10% Type-I error.

Based on the evidence, Tukey’s HSD tests, shown in Table 7, suggest that the material’s average notch toughness at −50ºC is statistically different from the average toughness observed at −25º, −5º, and 25ºC, at 5% Type-I error (Alpha).

The cursory graphical analysis of the experimental data revealed that moisture seems to significantly increase the material’s notch toughness variation. This led to additional F-tests to check the statistical significance of this increase in variation. Results from the four tests conducted at each of the four levels of the temperature factor are summarized in Table 8. It was decided to control the overall Type-I error for the four tests at 10%. This leads to setting the Type-I error for each of the tests approximately at 2.6%. Based on the results, the F-tests, conducted with 10% overall Type-I error, strongly suggest that moisture does increase the material's notch toughness variation at all temperatures other than the room temperature of 25ºC.

SUMMARY AND CONCLUSIONS

With the increasing use of polymer composite materials in structural applications, it is necessary to fully understand and characterize the mechanical behavior of such materials under a wide variety of potential environmental conditions. Although a significant amount of research currently is being conducted on high temperature applications, relatively little is known regarding the effects of reduced temperatures on the behavior of polymer composites. In this study, notched Izod impact tests were conducted on dry and wet specimens of a pultruded, glass fiber reinforced polymer composite at temperatures of 25º, −5º, −25º, and −50ºC. Fifteen specimens were tested at each condition, with all tests being conducted such that the impact direction was parallel to the fibers. A statistical analysis was then performed to determine the effects of low temperature and moisture on the notch toughness of the composite.

The results of this study appear to reveal a slight reduction in the mean impact strength of the wet specimens at −5º relative to 25ºC, likely due to the internal stresses created by expansion of the moisture during freezing. For both the wet and dry specimens, the impact energy increases by approximately 20% as the temperature is lowered to −50ºC. This trend is consistent with the results reported by Voelker (1991). Although the statistical analysis revealed no significant effect of moisture on the mean value of the impact energy at any temperature, the analysis did show an increase in the variation or scatter in the notch toughness values due to the presence of moisture, at all temperatures below freezing. This is likely due to competing effects between moisture and temperature in resin-rich and fiber-rich areas; i.e., the effect of freezing moisture on the internal stress state in an area with low fiber density would likely differ from that in an area of high fiber density. This scenario, coupled with the random placement of the notch in the material, would thus provide an explanation for the increase in the scatter in notch toughness values in the wet specimens. Whatever the mechanism, the increase in scatter in the wet specimens must be accounted for in the design of components that will be subjected to similar environmental conditions.

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Table 1
Notched Izod Impact Strength (J/m) of Long
Glass Fiber Reinforced Composites (Voelker, 1991)

Material[1]	22ºC	–1.1ºC	–23.3ºC	–31.7ºC
Nylon 6 (30)	249.3	232.7	229.5	231.7
Nylon 6 (40)	438.2	403.5	376.8	367.2
Nylon 6 (50)	490.6	450.3	433.4	438.2
PP (30)[2]	436.6	433.4	421.6	465.4
PP (40)	502.8	528.4	549.8	576.5
PP (50)	531.6	573.2	610.1	677.9
PET (30)[3]	214.0	217.2	226.3	228.4
SMA (30)[4]	218.3	224.7	241.2	247.6

[1] Numbers in parentheses refer to weight percent fiber
[2] PP = Polypropylene
[3] PET = Polyethylene terephthalate
[4] SMA = Styrene maleic anhydride

Table 2
Notched Izod Impact Strength (J/m) of Short
Glass Fiber Reinforced Composites (Voelker, 1991)

Material[1]	20ºC	–40ºC
Nylon 6	119.7	114.3
PP Homopolymer	103.4	108.9
PP Copolymer	185.1	114.3
Nylon 66	250.4	163.3
PU Elastomer	517.1	103.4
Polyester Elastomer	288.5	147.0
PC	185.1	147.0
ABS	81.7	76.2
Acetal	98.0	87.0

[1] All composites contain 30 wt% fiber

Table 3
Charpy Impact Energy (J) for
Wood Fiber Composite (Dutta et al, 1994a)

Temp. (ºC)	Energy Absorbed during Impact		
	Upper Bound	Lower Bound	Average
50	54.0	36.5	47.3
24	7.2	4.7	5.9
–5	6.5	3.8	5.8
–20	8.1	3.4	5.5
–40	6.8	3.4	4.5
–60	6.5	3.4	4.5

Table 4
Charpy Impact Strength (kg-cm/cm^2) of Woven Cloth Reinforced Epoxide Resin (Nishijima and Okada, 1982)

		Specimen Orientation			
Material	Temp.[1]	0º	15º	30º	45º
Glass	RT	395	270	245	110
Cloth	LNT	365	375	285	95
Carbon	RT	75	120	135	150
Cloth	LNT	85	145	110	85

[1] RT = Room Temp., LNT = Liquid Nitrogen Temp.

Table 5
Energy Absorbed (J) during Dynamic Tensile Tests of Glass Fiber Reinforced Composite (Dutta et al, 1994b)

Specimen Condition	Temperature		
	24ºC	–5ºC	–40ºC
Dry, uncycled	10.1	10.7	10.7
Dry, cycled[1]	9.6	10.1	11.2
Wet[2], uncycled	9.7	10.3	11.1
Wet[2], cycled[1]	10.2	11.1	10.4

[1] 100 thermal cycles at ±60ºC

[2] 0.35% moisture content

Table 6
General Linear Models Procedure Results
Dependent Variable: NT (Notch Toughness)

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	0.145674	0.036418	4.50	0.0021
Moisture	1	0.021277	0.021277	2.63	0.1078
Temperature	3	0.124396	0.041465	5.12	0.0023
Error	115	0.931593	0.008101		
Corrected Total	119	1.077267			

R-Square	C.V.	Root MSE	NT Mean
0.135225	23.9246	0.090005	0.376201

Table 7
Tukey's Studentized Range (HSD) Test Results
Dependent Variable: NT (Notch Toughness)

Type-I Error = 5% df = 115 MSE = 0.008101
Critical Value of Studentized Range = 3.687
Minimum Significant Difference = 0.0606

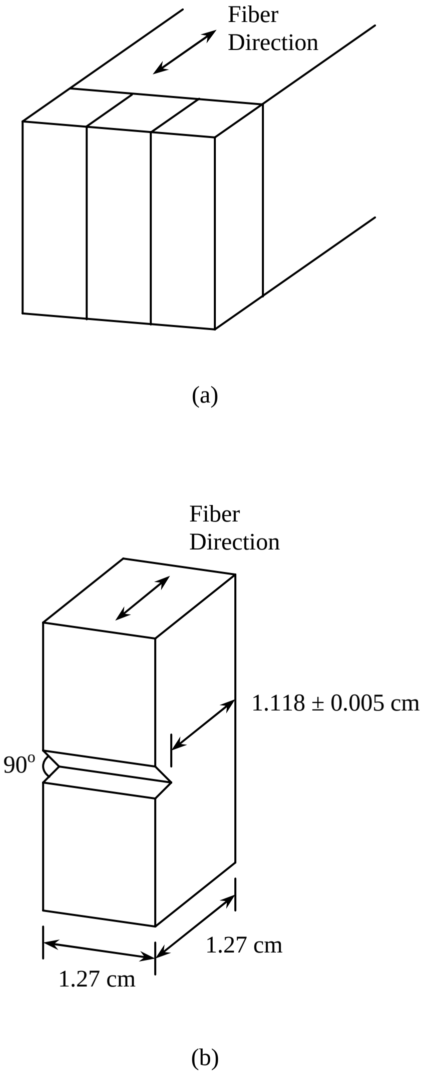
Tukey Grouping	Mean Notch Toughness	Number of Data Points	Temp.
A	0.43118	30	–50ºC
B	0.36617	30	25ºC
B	0.35628	30	–25ºC
B	0.35117	30	–5ºC

Note 1: Means with the same letter are not significantly different.

Note 2: This test controls the Type-I experimentwise error rate in contrast to the least significant difference tests that control the Type-I comparisonwise error rate.

Table 8
F-Test Results
Dependent Variable: NT (Notch Toughness)

Temp.		Notch Toughness	
		Dry	Wet
25ºC	Mean	0.340836	0.391509
	Variance	0.006906	0.010235
	Observations	15	15
	df	14	14
	F	0.674762	
	P(F<=f) one-tail	0.764450	
–5ºC	Mean	0.351486	0.350865
	Variance	0.003041	0.010172
	Observations	15	15
	df	14	14
	F	0.298965	
	P(F<=f) one-tail	0.984525	
–25ºC	Mean	0.345133	0.367428
	Variance	0.002854	0.014704
	Observations	15	15
	df	14	14
	F	0.194061	
	P(F<=f) one-tail	0.997939	
–50ºC	Mean	0.414072	0.448282
	Variance	0.003920	0.013966
	Observations	15	15
	df	14	14
	F	0.280720	
	P(F<=f) one-tail	0.988210	



(a)

(b)

Fig. 1 (a) Specimen extraction from solid bar stock and (b) specimen geometry.

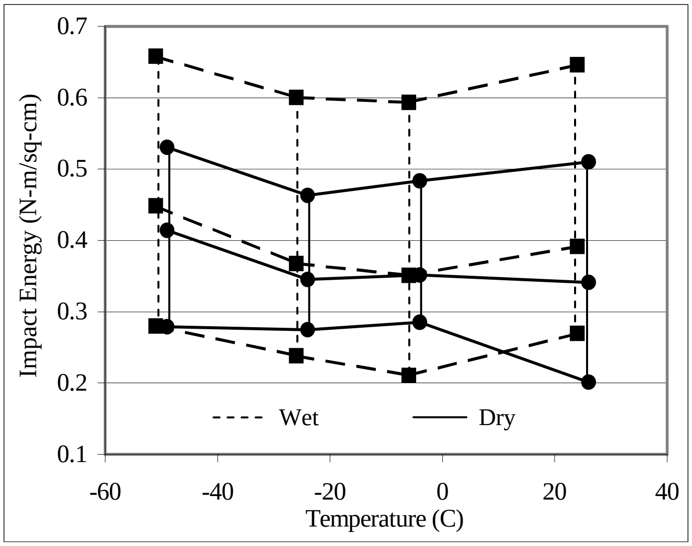


Fig. 2 Results of Izod notch toughness experiments (high-avg-low).

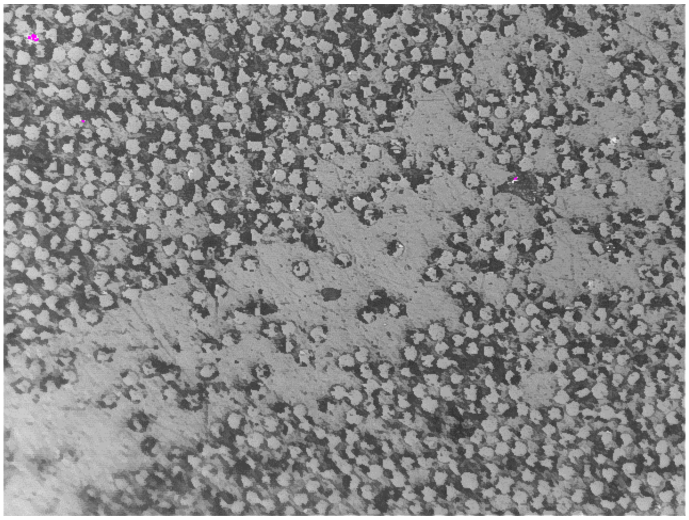


Fig. 3 Magnified view of a cross section of the composite material.