A Study on Chemical Composition, Physical, Tensile, Morphological, and Thermal Properties of Roselle Fibre: Effect of Fibre Maturity

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Roselle fibre is a type of natural fibre that can be utilized as apotential reinforcement filler in polymer composites for different applications. This work investigates the chemical, physical, mechanical, morphological, and thermal characteristics of roselle fibre at different levels of maturity (3, 6, and 9 months). The diameter of roselle fibre increases as the plant matures. However in contrast to this, the moisture content and water absorption of roselle fibre decrease as the plant matures. Chemical content of roselle fibres from plants of different ages indicate that as the plant matures, the cellulose content decreases. Tensile strength of roselle fibre decreases from 3 months old to 9 months old. The cross section of roselle fibre shows a typical morphology of bast fibre, where there is a lumen in the central of fibre.Thermal analysis results show that the effect of thermal decomposition of roselle fiber is almost the same for all plant ages. It is concludedthat roselle fibres can be used as reinforced material for manufacturing of polymer composites. Based on its excellent properties, roselle fibres are suitable for different applications such as automotive and building components at lower cost.

Keywords: Roselle Fibre; Plant age; Physical properties; Mechanical properties; Chemical compositions; Thermal properties

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INTRODUCTION

Natural fibres such as hemp, kenaf, jute, sisal, banana, flax,and oil palm have been in considerable demand in recent years due to their eco-friendly and renewable nature (Khalil *et al.* 2012). Recently, in line with raising environmental concerns, scientists and researchers are now replacing synthetic fibres with natural fibres as the main component in composites (Favaro 2010; Nirmal *et al.* 2011; Reem *et al.* 2012; Begum and Islam 2013; Sathishkumar *et al.* 2013a; Cholachagudda and Ramalingaiah 2013). The advantages of natural fibres include low cost, good mechanical properties, abundant availability, material renewability, biodegradability,non-abrasive nature, and ease of recycling as compared to synthetic fibres (Jawaid and Abdul Khalil 2011; Ishak *et* *al.* 2013). These reasons have attracted material engineers to use natural fibres as reinforcing filler in polymer composites to reduce uses of timber or forest resources and explore under-utilized natural fibres. Natural fibres are widely used inautomotive (AL-Oqlaand Sapuan 2014) and construction engineering (Dittenber and GangaRao 2012). Natural fibres can be found in southeast Asian countries such as Malaysia, Indonesia, and Thailand (Ishak *et al.* 2013).

Natural fibres such as roselle fibres (*Hibiscus sabdariffa*) are found in abundance in nature and cultivated in Borneo, Guyana, Malaysia, Sri Lanka, Togo, Indonesia, and Tanzania. The scientific name for roselle is *Hibiscus sabdariffa* L., and it is from the Malvaceae family (Morton 1987). Roselle is found abundantly in tropical areas. They are commonly used as an infusion and to produce bast fibre. There are various uses of roselle. The fruit is commonly used in medical applications(Tori Hudson 2011;Mungole and Chaturvedi 2011) and in the food industry (Wilson 2009), while the fibre is used as a textile (Managooli 2009). Roselle fibre can be extracted by water retting (Thiruchitrambalam *et al.* 2010). Roselle stem is red in color, as illustrated in Fig. 1(a). In Malaysia, after a year, theroselle plant will be cut, and it will become a waste. This is because the quality of roselle fruit is not good after a year. In order to use this plant efficiently, the fibre can be used as reinforcement material for polymer composites. It is important to understand the physical, thermal, mechanical, and chemical properties of roselle fibre before it is used in industrial application such as in automotive and construction engineering.

Roselle fibre is one of the natural fibresthat have attracted researchers to explore their capability as a reinforcement material in composites. Researchers have reported work on modification of the roselle fibre to improve the fibre/matrix interfacial bonding for fabrication of polymer composites for different applications (Kaith and Chauhan 2008; Chauhan and Kaith 2011, 2012a,b; Ramu and Sakthivel 2013). Also, a few researchers have review paperscovering the chemical and mechanical properties of roselle fibre in polymer composites (Thiruchitrambalam *et al*. 2010; Chauhan and Kaith 2012b).

Some recent studies on chemical composition, mechanical, thermal, andmorphological characteristicshave been carried out on natural fibres (Rowell *et al.* 2000; Munawar *et al*. 2006; Rosa *et al.* 2009; Ishak *et al.* 2011; Sathishkumar *et al*. 2013; Mwasiagi *et al*. 2014; Yusriah *et al.* 2014). Ishak *et al.* (2011) have conducted a characterization on the thermal and tensile properties of sugar palm fibres. In their studies they found that the green fibre (matured fibres) from the plant give the greatest tensile results because of the optimum chemical composition, which consists of high cellulose, in addition tohemicelluloses and lignin content (Ishak *et al.* 2011). Yusriah *et al.* (2014) have discussed the effect of maturity (raw, ripe, matured) of betel nut husk (BNH) on the physical, mechanical, thermal, and morphology properties. They found that the ripe type of fibre shows the highest tensile results (Yusriah *et al.* 2014). Shahzad (2013) has conducted a study on mechanical and physical properties of hemp fibres. The characterization was focused on the moisture content of the fibre by using TGA and tensile properties, and they found that hemp fibres have potential as reinforcement material (Shahzad 2013). However, to this date, very limited studies have been done on the application of roselle fibres and their composites (Ramuand Sakthivel 2013).

The present work considers the properties of roselle fibre with respect to plant ages (3 month, 6 month, and 9 months old). Chemical composition, physical properties (density, water absorption, moisture content), tensile strength, morphology (Scanning electron microscopy), and thermal analysis of roselle fibre at three different plant ages were evaluatedrelative to suitability of the fibre as reinforcement in polymer composites. From the literature review, there has been no such study reported by any researchers until now. The aim of this paper is to provide extensive information on different fundamental properties of roselle fibres on the basis of plant age to enhance its utilization in different applications.

EXPERIMENTAL

Materials

Roselle plants with different ages (3, 6, and9 months old) had been collected from a roselle plant field at Mersing, Johor, Malaysia. The roselle fibre was extracted by using a water retting process with different duration of time. Table 1 shows the water retting process for different ages of roselle fibre. The older ages of roselle plant required more time for extraction by water retting due to the skin of the roselle plant being thicker than is the case forthe younger plant. The retted stem of the roselle plant was washed in running water, and fibres were removed manually. Next, fibres were cleaned, and then dried in the sunlight. The roselle fibres were then prepared for several tests to study its potential as reinforcement material in polymer composite. Figure 1a) shows the roselle plant and b) water retting.

Table 1. Duration of Water Retting forDifferent Plant Age

Ages of roselle plant	Duration of water retting
3 months	5 days
6 months	7 days
9 months	14 days

Fig. 1a) Roselle plant **b)** water retting process

Chemical Composition

The chemical composition of roselle fibre was analyzed by using Neutral Detergent Fibre (NDF) and Acid Detergent Fibre (ADF). This is a common way to evaluate the mainfibre constituents, cellulose, hemicelluloses, and lignin. The percentage of cellulose and hemicelluloses can be determined by using Eqs. 1 and 2, respectively.

$$\text{Cellulose} = \text{ADF} - \text{lignin} \tag{1}$$

$$\text{Hemicelluloses} = \text{NDF} - \text{ADF} \tag{2}$$

Physical Characterization

Diameter

The diameters of roselle fibres were measured by using an optical microscope, Zeiss model. Fifteen samples of single fibres were measured, and the average diameter was obtained.

Density

The density of roselle fibre was calculated by dividing mass over volume as shown in Eq.3based on the ASTM D792. The fibre was in the powdered form. First, thevolume and mass of the container were measured. The volume of the container was determined by Eq. 4. An analytical balance with the capability of reading up to 0.0001 g was used to measure the mass of the fibre and the container. Initial mass of the container was recorded as M_0. The fibre was heated in the oven for 24 h at 104°C before the mass was measured to eliminate the effect of moisture and water absorption. The powdered fibres were then filled in the container. The container with powdered filled was weighed as M_1.

$$\text{Density (g/cm}^3\text{)} = \frac{mass\ (M_1 - M_0)}{volume} \tag{3}$$

In Eq. 3, $M_1$is the Mass of the container plus the dried fibre, whereas M_0 is the mass of the container.

$$\text{Volume (cm}^3\text{)} = heigth\ x\ width\ x\ depth \tag{4}$$

Water absorption

The percentage of water absorption of roselle fibre was determined by using Eq.5. Five samples were prepared, and the average of percentage water absorption was calculated. The samples were weighed as M_0 first before being immersed in fresh water for 24 h at room temperature. After 24 h of immersion, the samples were then weighed again as M_1.

$$\text{Water absorption (\%)} = \frac{M_1 - M_0}{M_0}\ x\ 100 \tag{5}$$

Moisture content

Five samples were prepared for the moisture content evaluation. Percentages of moisture content of roselle fibre were determined by using Eq. 6. The samples were heated in an oven for 24 h at 105 ^{o}C (Baley *et al.* 2012). Before heating the samples, the weight of fibre was measured as M_0. After 24 h in the oven, the fibre was weighed again as M_1.

$$\text{Moisture content (\%)} = \frac{M_1 - M_0}{M_0} x\ 100 \qquad (6)$$

Tensile Properties

The tensile test is a simple method in order to determine the mechanical strength of natural fibre. Several significant mechanical properties can be obtained from thetensile test such as Young's modulus, tensile stress, maximum elongation, tensile strain, and yield stress. The tensile properties of rosellefibre were determined using aUniversal Testing Machine; model Instron 5556, as shown in Fig. 2. ASTM D3379 standard was used for the single fibre tensile test. Gauge length of the roselle fibre samples was 20 mm, and the cross-head speed was 1 mm/min with a 5 kN load cell. The fibre was properly selected under optical microscope before being tested to ensure that the specimen yields accurate result. The fibre was glued on the sample holder as shown in Fig. 3. Before testing was commenced, the sample holder was cut at the middle. Fifteen samples of roselle fibre were prepared to perform thetensile test.

Fig. 2. Tensile test for roselle fibre

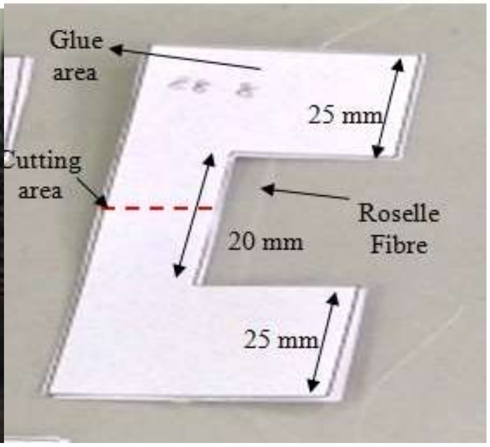


Fig. 3. Sample preparation for tensile test

Thermogravimetric Analysis (TGA)

Thermal characterization of roselle fibre was carried out using aQ series thermal analysis machine from TA Instrument company. TGA measures weight changes in a material as a function of temperature (or time) under a controlled atmosphere. It is important to determine the degradation of natural fibre at high temperature before it is used in polymer composites. This is because the degradation temperature of natural fibres needs to be obtained first before the manufacturing process with polymer in order to know the compatibility of the fibres with polymer during manufacturing process at high temperature. To avoid an off-quality composite, the fibres must not degrade during the manufacturing process in order. 4.8 mg of roselle fibre was placed in the chamber. Analysis was done in air with temperature range of 50 to 600 ^{o}C, and the heating rate was 10 ^{o}C.

Morphology Analysis (SEM)

The morphology and the cross-section of roselle fibre was observed under scanning electron microscope (SEM), model Hitachi S-3400N. Roselle fibre is very fine, such that it is difficult to obtain the cross-sectional morphology of the fibre. In order to overcome this problem, roselle fibres were immersed in liquid nitrogen to harden them. The fibres were gold coated in order to obtain a good quality of results. The working distance used to examine the samples was 71 mm, and the acceleration voltage was 15 kV.

RESULTS AND DISCUSSION

Chemical Composition

Chemical composition is one of the important elements that influences the physical, mechanical, and thermal properties of a natural fibre. The common chemical content of natural fibre is cellulose, hemicelluloses, lignin, and ash. The different proportion of these contents depends on the source of the fibre, theextraction process, and theage of the fibre (Mukherjee and Radhakrishnan 1975). Table 2 shows the chemical content of roselle fibres with respect to their ages. As shown in Table 2, roselle fibres have high cellulose content, which ison average more than 60%. At the age of 3 months, roselle fibre has thehighestcellulose content, which is 64.50% compared to the 6 and 9 months with 60.51% and 58.63%, respectively. Cellulose is the main structural component, which provides the stability of the stem plant wall and strength to the fibre (Reddy and Yang 2005). It also influences the properties, the cost of fibre production, and the usage of the fibre for various applications. Figure 4 shows the chemical content of roselle fibre with respect to plant age. The proportion of chemical content is different depending on the plant's age. Cellulose content gradually decreases as the plant matures. This process occurs naturally as the tree dies. For all dead trees, the chemical compositions are broken apart eventually and the residuals are returned naturally to the environment. These remaining constituent atoms will be used as building blocks for other existing organisms for growth (Ishak *et al.* 2011). Higher cellulose content contributes to the higher strength of fibre, which makes it preferable for textile, paper, and other fibrous applications (Favaro 2010). Hemicelluloses and lignin content also change as the plant matures. However, the changes are dependent with each other; a decrease of hemicelluloses content is accompanied by an increase of lignin and *vice versa*.

Table 2. Chemical Content of Roselle Fibre

Months/ Chemical content	Cellulose	Hemicellulose	Lignin	Ash	References
3	64.50	20.23	6.21	1.25	Current study
6	60.51	16.27	10.26	1.03	Current study
9	58.63	20.82	7.87	2.08	Current study
Kenaf	31-63.5	17.6-23	12.7-19	2-5	Li *et al.* 2007; Jonoobi *et al.* 2009
Jute	45-71.5	13.6-21	13-26	0.5-2	Li *et al.* 2007; Wang *et al.* 2008
Hemp	57-77	14-22.4	3.7-13	0.8	Li *et al*. 2007
Flax	64 - 71.9	16.7 – 20.6	2 – 2.2		Sathishkumar *et al.* 2013

Hemicelluloses constitute a class of polysaccharide polymers in which the degree of polymerization and orientation are less than cellulose (Sathishkumar *et al.* 2013). They normally occupy the space in between cellulose and lignin. The composition of hemicelluloses is mainly sugar, glucose, xylose, galactose, arabinose, and mannose (Reddy and Yang 2005). In terms of mechanical properties, hemicelluloses providea small contribution to the stiffness and strength of the fibre (Reddy and Yang 2005). Hemicelluloses hydrolyze more easily into sugar compared to cellulose. A high content of hemicelluloses is preferable for producing ethanol and other fermentation products. Although hemicelluloses do not directly contribute the strength to the fibre, theyact as a binder of microfibrils and provide structural reinforcement to microfibrils.

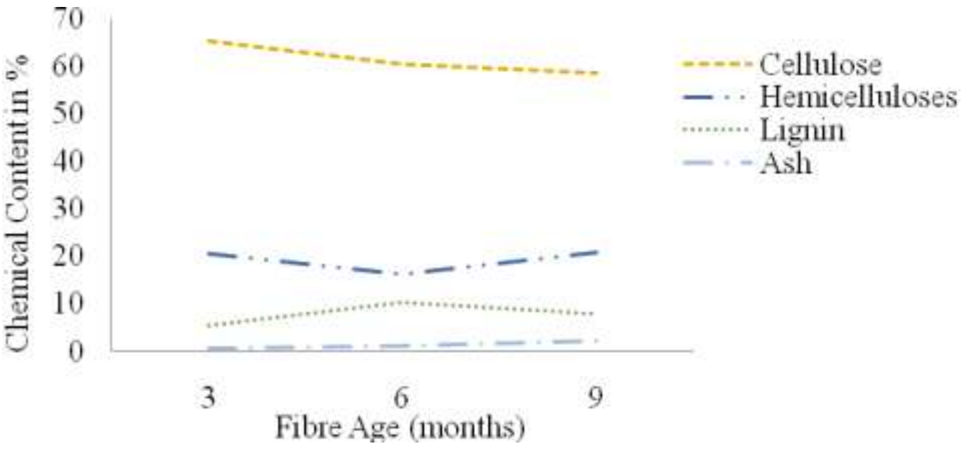
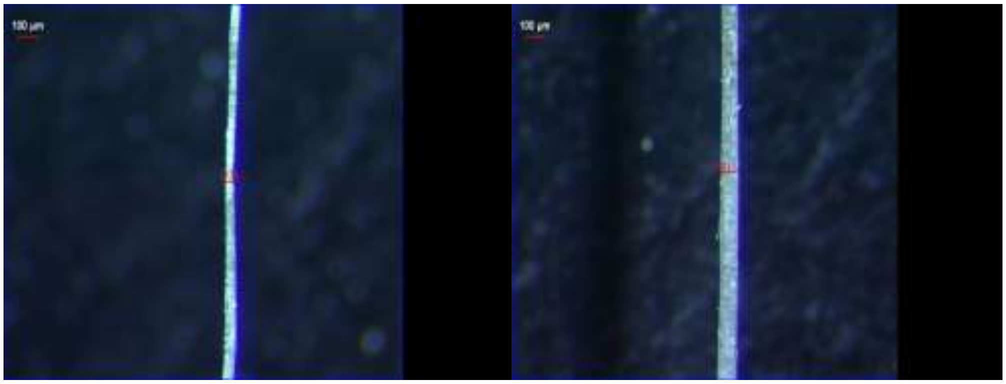


Fig. 4. Chemical content in % versus plant age of roselle fibre

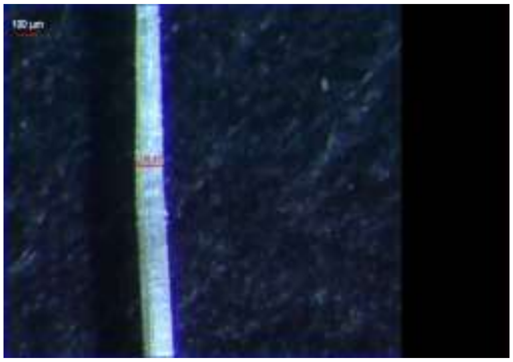
It can be seen that the lignin content of roselle fibre is comparable with other established bast fibre. The specimens corresponding to6 months of plant fibre age showed the highest lignin content, which was 10.26%. Lignin is amorphous and has aromatic rings with various possible branches. It acts as glue between individuals' cells and between the fibrils forming the cell wall. Lignin is first formed between neighboring cells in a middle lamella, bonding them tightly into a tissue, and then it spreads into the cell wall, penetrating the hemicelluloses and bonding the cellulose fibrils. Lignin provides plant tissue and individual fibres with compressive strength and stiffness to the cell wall to protect the carbohydrate from chemical and physical damage. The lignin content influences the structure, properties, morphology, flexibility, and rate of hydrolysis (Reddy and Yang 2005).

Physical Properties

Table 3 shows the physical properties of roselle fibre with respect to different ages of roselle plant. Based on the diameter measurement, the 3-month-old fibre exhibited the smallest range of fibre diameter, which was 40 to 100 µm. Diameters for the other two groups of different plant age of 6 and 9 months were 80 to 120 and 90 to 150 µm, respectively. Viewed under amicroscope, the 3-month-old fibre appeared brighter than the 9-month-old fibre. However, the diameter of roselle fibre varies because of the bundle of single fibres. It is hard to get a single fibre measurement with naked eyes. It can be seen that there was a difference in diameter between the three different age classes of fibre, and the diameter of roselle fibre increased from 3 months to 9 months. From the result obtained, it can be concluded that fibre diameter increases with age because the cell wall gets thicker as it matures. The factors affecting the different physical properties of roselle fibre are the cell wall thickness, diameter, and length of tracheid in the fibres (Rowell *et al.* 2000). The results shown were taken from 15 samples. Roselle fibre is a fine bast fibre. Figure 5 shows the diameter measurement of roselle fibre by using an optical microscope (Leica) with 4(a) showing the diameter at 3 months old, while 5 (b) and 5 (c) at 6 and 9 months old, respectively. Physical properties of natural fibres depend on several factors (Rowell *et al.* 2000). Naturally, it is hard to get the consistence properties of natural fibre (Chandramohan and Marimuthu 2011), and the measurement of the properties depend on the maturity of the plant, source of the fibre, extraction process, and condition of the plant (Reddy and Yang 2005).



(a) 3-month-old roselle fibre (b) 6-month-old roselle fibre



(c) 9-month-old roselle fibre

Fig. 5. Diameter measurement of roselle fibre

Roselle fibre density for 3, 6, and 9 months was found to be 1.332, 1.419, 1.421g/cm^3, respectively. From the results obtained, it is clear that density increases as the plant matures. The density of roselle fibre is relatively low. This feature is mainly contributed by the presence of a lumen in the fibre structure (Aziz and Ansell 2004; Vilay *et al.* 2008). Lumen structure is hollow with thin walls, as depicted in Fig.8 (a), (b), and (c). This characteristic contributes to natural fibre lightness. This is one of the most desirable factors of natural fibre as reinforcement material for composite products.

Although natural fibres have many advantages relative to their use as a reinforcement material such as being environmental friendly and having relatively similar properties as synthetic fibre, there are still flaws in natural fibre. The hydrophilic behavior of natural fibre makes it difficult to have a good adhesion between fibre/matrix and contribute to high water absorption of natural fibre; and this will weaken the composites product in application (Nguong *et al.* 2013). However, this problem can be overcome with surface treatment of natural fibre (Aziz and Ansell 2004; Xie *et al*. 2010). From the results obtained, percentages of water absorption of roselle fibre 3, 6, and 9 months were high, *i.e.* 320%, 306%, and 289%, respectively. The lumen structure has a great affinity toward water. As more lumen is present, more water is absorbed by roselle fibres. These phenomena also exist due to the cellulose content in natural fibres in general and roselle fibre in particular. A higher percentage of cellulose content increases free hydroxyl groups (Athijayamani *et al*. 2009; Yusriah *et al*. 2014). In this research, it was found that cellulose content was the highest for plant age 3 months, and this fact shows that the water absorption results were in good agreement with other published literature. Water absorption of natural fibre must be reduced to produce a high quality composite. In addition, fibre and matrix adhesion can be further improved by strengthening the composite.

Moisture content of natural fibre is an important criteria that needs to be considered in choosing natural fibre as reinforcement material. This is because moisture content affects dimensional stability, electrical resistivity, tensile strength, porosity, and swelling behavior of natural fibre in a composite material. From other published literature, it was found that low moisture content of the natural fibre is the most desirable criteria for polymer composites in order to overcome the problems mentioned above (Jawaidand Abdul Khalil 2011).Composites combined with less moisture content fibre are less likely to decay in contrast to composites combined with high moisture content. This is probably because of the ability of fibre to retain water within the composites, which may promote degradation of the composites (Rowell *et al.* 2000). The result of moisture content of roselle fibre indicated that, plant age of 9 months old roselle fibreswas the lowest, as shown in Table 3. Therefore, 9-month-old roselle fibre is most suitable in fabricating composites product in order to have a high dimensional stability and quality of the product. Overall, fibre withinthese 3 different plant ages is acceptable to beused as a natural fibre in composites application in terms of moisture content and water absorption because the results are similar to other established natural fibre such as kenaf and jute, where their range is 3 to 5% and ~200%, respectively (Saheb and Jog 1999).

Table 3. Physical Properties of Roselle Fibre ofDifferent Age

Months / Physical properties	3	6	9
Diameter (µm)	40-100	80-120	90-150
Density (g/cm^3)	1.332	1.419	1.421
Moisture content (%)	5.8	4.9	3.7
Water absorption (%)	320	306	289

Tensile Properties

Figure 6shows a typical stress strain curve for roselle fibre obtained from different plant age. It is apparent that roselle fibres fail in a brittle manner when sudden load is applied. It is difficult to study single fibre tensile test results of small brittle natural fibres due to high scatter that occurs. This scatter can be mainly related to three factors, namely test parameters/conditions, plant characteristics, and area measurements (Silva *et al.* 2008). The obtained results in Fig. 7 show that increase of plant age will decrease the tensile properties of roselle fibre. At 3 months of age, roselle fibre gave the highest average tensile strength, 453.477 MPa, while 6 and 9 months gave 247.28 and 228.57 MPa, respectively. This result is influenced by cellulose content, where higher cellulose structure in the fibre contributed to the high strength properties. This is due to the cellulose crystalline structure, which contributed to stability of the plant stem (Reddy and Yang 2005).

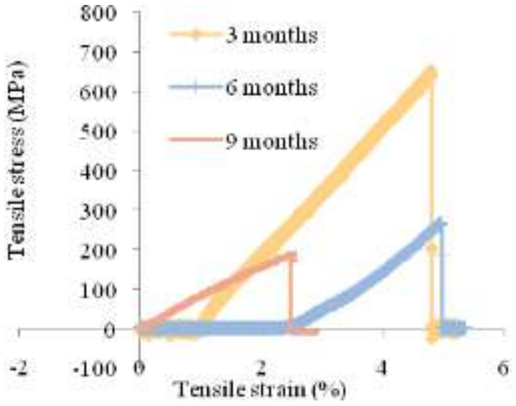


Fig. 6. Stress strain curve of roselle fibre

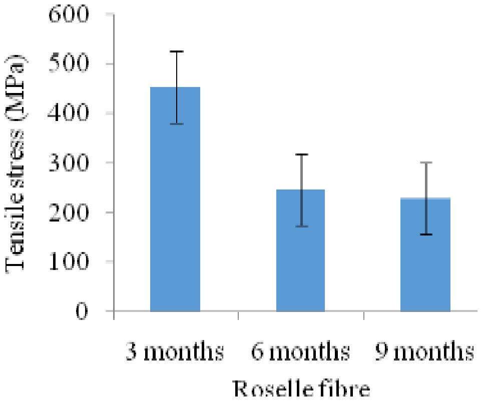


Fig. 7. Tensile stress versus plant age of roselle fibre

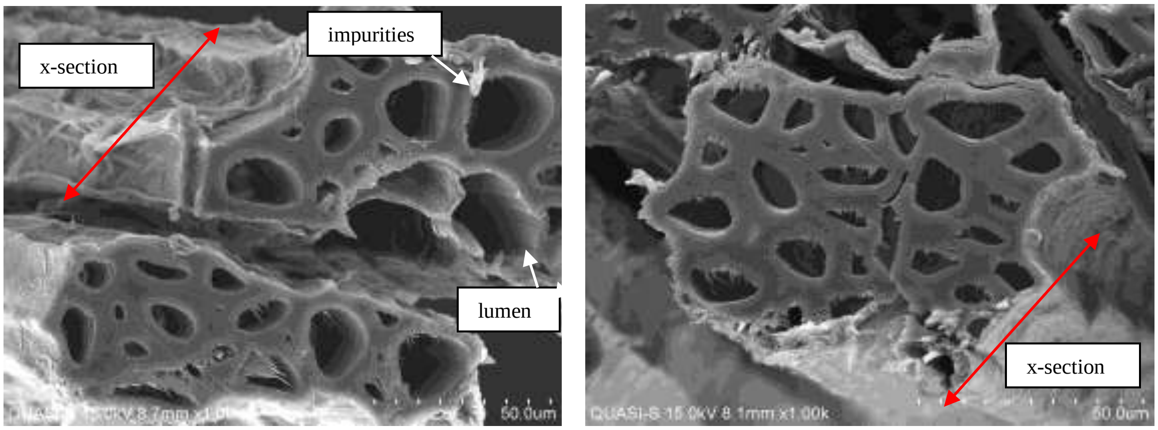
Table 4 shows the tensile strength of roselle fibre compared to other bast fibre. As shown, the capability of roselle fibre as reinforcement material for composites was in good agreement with other fibres. The study of tensile properties of fibres is important because the load applied to composites will transfer to the fibre first. The fibre helps to sustain the load applied, and once the fibre has failed, then the composite as a whole will have failed. The structure of bast fibre is almost similar for all type of fibres (Kalia *et al.* 2011). The differences between the plant fibres are their compositions, *i.e.*, the ratio between cellulose and lignin/hemicelluloses and in the orientation or spiral angle of the cellulose microfibril (Kalia *et al.* 2011). Generally, natural fibres consist of cellulose, lignin, and hemicelluloses. Usually, the tensile strength and Young's modulus of fibres increase as the cellulose content increases (Ishak *et al.* 2011). The ductility of the plant fibres depends on the orientation of microfibrils to the fibre axis. If it is spiral, thenit is ductile, while if it is parallel, it is rigid, inflexible, and has high tensile strength. Another factor that affects the properties is the fibres' defects. The fibre used as a reinforcement material must have a minimum of defects, where if the defects present in the structure, the failure will start at the weak point (defects). Thus, a detail inspection under microscope needs to be performed in order to determine the quality of a fibre.

Table 4. Tensile Strength of Bast Fibre

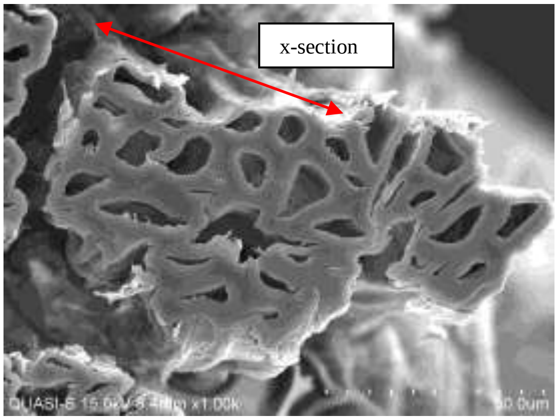
Fibre	Tensile Strength (MPa)	Reference
Roselle 3 months	414.72	Current study
Roselle 6 months	252.64	Current study
Roselle 9 months	228.5738	Current study
Kenaf	18–180	(Akil *et al.* 2011)
Hemp	300-800	(Clemons 2010)
Jute	340-384	(Xia *et al.* 2009)
Flax	500-900	(Clemons 2010)
Ramie	400-938	(Ku *et al.* 2011)

Morphological Properties

The morphology of the natural fibre is one of the factors that influences the physical and mechanical properties of the fibre as a reinforcement material for composites material (Munawar *et al*. 2006). It clear from Fig. 8 that the cross-section of roselle fibre has a clear lumen structure in the center, becauseas fibres become older,the lumen structure shrinks due to the presence of a thicker cell wall. From the observation, thelumen structure of 3 months old roselle fibre is bigger compared to the 6 and 9 months. The cell wall structure is thicker with increase of plant age (Ayre *et al.* 2009). According to Yusriah (2014), lumen sizes are related to the water uptake of the fibre (Yusriah *et al.* 2014). The bigger lumen size will contribute to the higher water uptake and moisture content of the fibre. The obtained result is in good agreement with their finding. This happens because the increase of lumen structure size has improved the capability of the fibre to absorb more water. The fibre of 3 months old shows the biggest lumen structure.This confirms that as the plant grows, more water is supplied for its consumption to grow. For most of plant fibers, the lumen structure is mainly filled with air, which makes the natural fibers as a potential material for acoustic absorbents and thermal insulators (Kymäläinen and Sjöberg 2008; Meredith *et al.* 2012; Liu *et al.* 2014).



(a) 3 months (b) 6 months

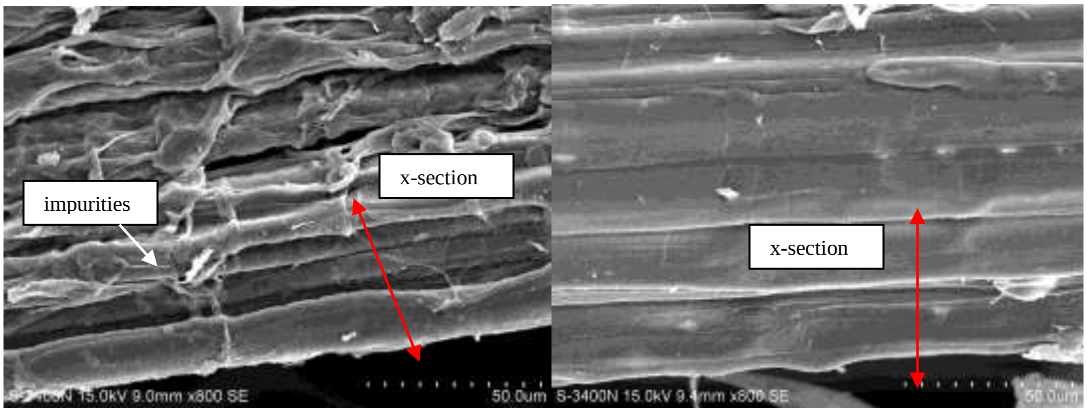


(c) 9 months

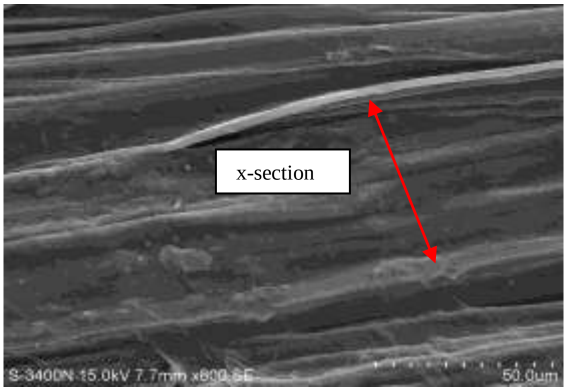
Fig. 8. Cross section of roselle fibre after (a) 3 months, (b) 6 months, (c) 9 months

Figure 8 shows the surface of roselle fibre of 3, 6, and 9 months, respectively. Naturally, bast fibre consists of a bundle of elementary fibre, and it is overlapped along the length of the fibres and bonded firmly together by pectin and other non-cellulosic compounds that give strength to the bundle as a whole (Rosa *et al*. 2009). The region at the interface of two cells is termed middle lamella. In common terminology the bundles of elementary fibres are referred to as technical fibres or single fibres (Mohanty *et al.* 2005). Figure 8 shows thatthe diameter of a bundle fibre is bigger with the increase of thicker cell wall. The bundles of roselle fibre were overlapped. A 3-month-old fibre appears thinner, and the surface is coarse compared to the older roselle fibre.

It can be concluded that the plant cell wall becomes thicker as it matures. For the interfacial bonding between the fibre and the polymer, the surface structure of plant age of 9 months old is more desirable in composites. This is because the structure is clean and rougher compared to 6 months. If the surface of natural fibre is coarse, it provides good interlocking between fibre and matrix. The fibre at the age of 9 months still absorbs a significant amount of water, but it can be treated chemically in order to overcome this deficiency.



(a) 3 months (b) 6 months



(c) 9 months

Fig. 9. Surface structure of roselle fibre after (a) 3 months, (b) 6 months, (c) 9 months

Thermal Stability / TGA

Thermogravimetric analysis was carried out to provide precise information on thermal stability. Thermal stability or thermal degradation of natural fibre is important, as the fibres are also affected during exposure to elevatedtemperature during the manufacturing process with polymer/resin. It is crucial to confirm that fibre used in composite has the capability to withstand the temperature applied during the manufacturing process or application of the product.

Table 5, Fig.10, and Fig. 11 show characteristic data of thermal behavior of roselle fibre at high temperatures. Approximately 5 mg of roselle fibre was used to evaluate the thermal behavior. Generally, there are 4 phases on the thermal degradation of natural fibre (Rosa *et al.* 2009; Shahzad 2013; Sathishkumar *et al.* 2013).

Table 5. Thermal Degradation Analysis of Roselle Fibre

Age of Roselle Fibre (months)	Weight loss (%) at temp range of 30 to 110 °C	First Degradation phase			Second Degradation phase			Char Residue (wt.%)
		T_1 (°C)	Weight loss (%)	T_{peak} (°C)	T_2 (°C)	Weight loss (%)	T_{peak} (°C)	
3	10.28	220-350	-	346	350-400	76.36	364.05	10.31
6	8.25	200-315	14.16	298.01	315-390	62.27	363.24	14.21
9	4.1	210-320	15.21	298.58	320-390	63.69	366.08	12.24

T_1= First degradation temperature; T_2: second degradation temperature

Figure 10 indicates the curves for weight loss and differential weight loss for roselle fibres as the temperature rises.

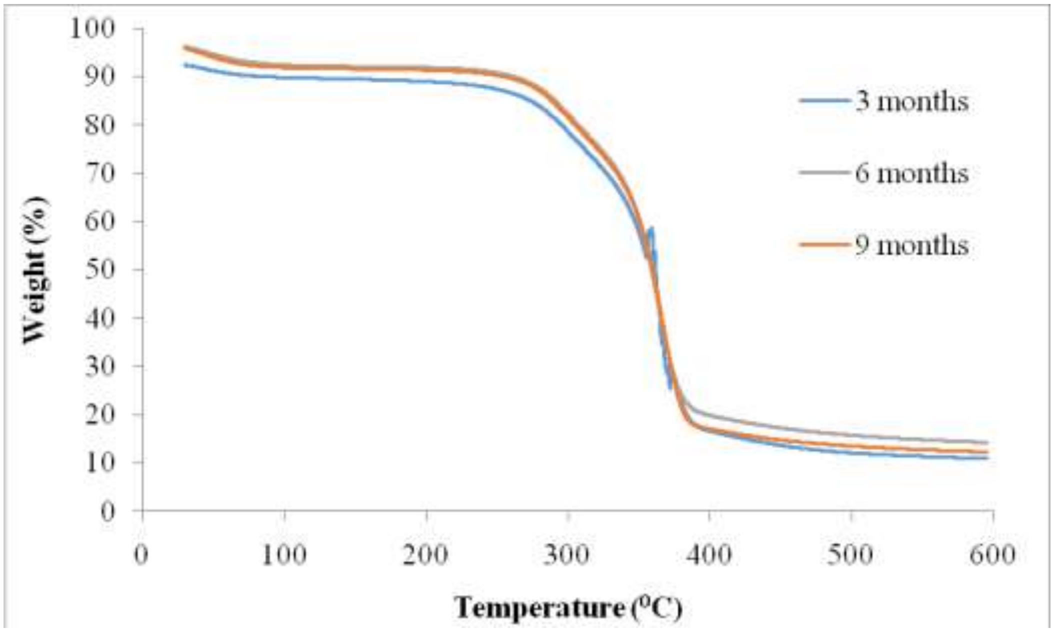


Fig. 10. Thermogravimetric analysis (TGA) of roselle fibre after (a) 3 months, (b) 6 months, (c) 9 months

Generally there were 4 stages of main thermal degradation of the roselle fibre. The first degradation is moisture evaporation, followed by the decomposition of hemicelluloses, cellulose, and lignin, leaving ash as the final residue (Ishak *et al.* 2011). The first degradation of natural fibres occurs between 30 ^{o}C to 110 ^{o}C (Rosa *et al.* 2009). This is due to the evaporation of moisture content in the fibre. In the case of roselle fibre, evaporation of moisture mainly ranged from 30 ^{o}C to 110 ^{o}C. As the temperature of fibre increases while it is heated, the fibres became lighter because of the evaporation of bound water and volatile extractives. Although less volatile extractives are still present, they tend to move toward the outer part of the fibre stem surface. This movement of volatile extractives occurs due to the water movement from the inner to the outer part of the fibre stem surface as the water available at the outer part evaporates. Eventually, the volatile extractives coalesce and migrate to the fibre surface (outer part of fibre stem). From the obtained results, it can be seen that 9-month-old roselle fibre exhibited the lowest percentage of mass loss which 4.1%, whilst the3 months and 6 months specimens showed 10.28% and 8.25%, respectively. The results of mass loss reflect the moisture content of the roselle fibre.

It can be seen that lignocellulose component decomposes in the range of 200 to 520 ^{o}C. The second phase thermal degradation of roselle fibre is due to the thermo-chemical change of hemicelluloses content in the fibre caused by the cellular breakdown as the temperature is increased. For 3-month-old roselle fibre, hemicelluloses start to decompose in the range of 220 to 350 ^{o}C. The 6- and 9-month-old roselle fibre starts to degrade at 200 to 315 ^{o}C and 210 to 320 ^{o}C, respectively. Hemicelluloses degrade earlier than other lignocellulosic components, cellulose, and lignin. Cellulose structure is more thermally stable compared to hemicelluloses. This is due to the fact that the hemicellulose structure contains heterogeneous polysaccharides such as galactose, glucose, mannose, and xylose. Such polysaccharides normally are very amorphous in nature, which allows them to easily migrate from themain stem. Eventually, the hemicellulosic saccharides become volatiles at relatively lower temperatures (Yang *et al.* 2007).

The second phase of degradation involvesthecellulose structure. Degradation of cellulose will only start after hemicelluloses decomposition is complete. The main reason behind this is the higher content of crystalline chain compared to amorphous. This will make cellulose more thermally stable (Ishak *et al.* 2011). Overall, for all different ages of roselle fibre, cellulose starts to decompose at a temperature of 315 ^{o}C and completely decomposes at 400 ^{o}C. From the published literature, cellulose will start to degrade at a high temperature of 315 ^{o}C (Yang *et al.* 2007). Once the required temperature is achieved, the decomposition starts and the mass loss rate is rapid. The percentage of weight loss for 3-, 6-, and 9-month-old roselle fibre is 76.36, 62.27, and 63.69%, respectively. Three-month-old specimens showed the highest weight loss in this temperature rangebecause of the highest cellulose content.It can be observed in Fig.11 that the portion of crystalline cellulose showed its highest peak during their decomposition process. Table 4 shows the degradation temperature range of cellulose structure. From the results, it can be seen that there were no significant changes between these three different ages of roselle fibre. It can be concluded that the degradation of lignocelluloses component of hemicelluloses and cellulose is in the same temperature range. This might be due to the different composition in chemical content between 3 to 9 months of roselle fibre. Chemical content of roselle fibre is related closely to their thermal behavior of the fibre.

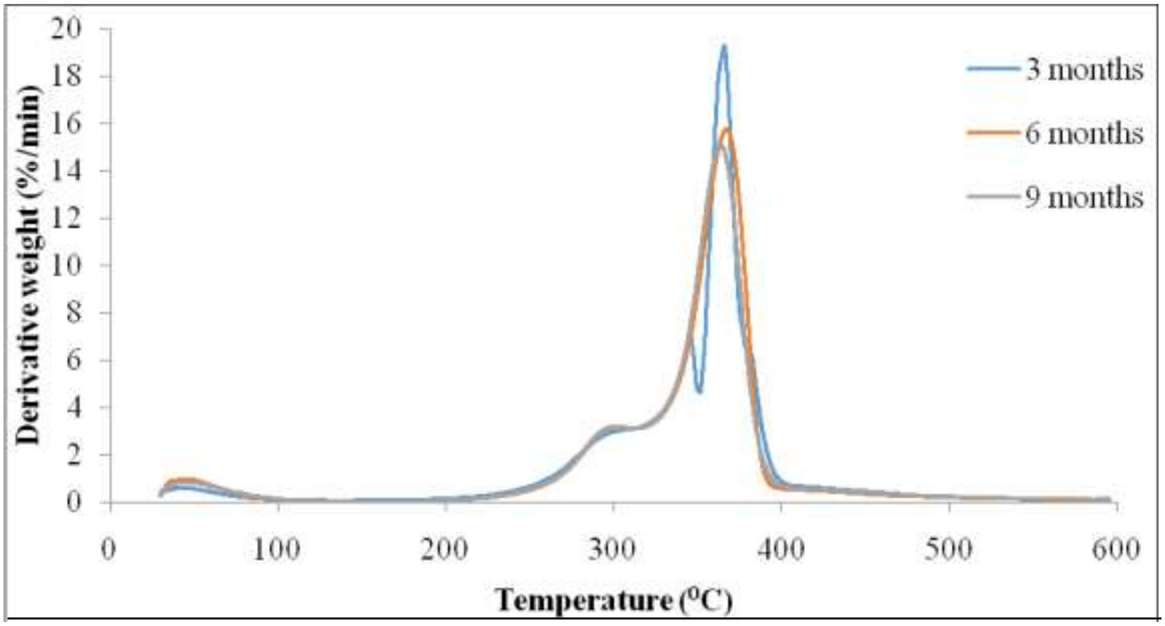


Fig. 11. DTG curves of roselle fibre

Lignin is the last component in the fiber to be decomposed because its structure is relatively complex. The complexity is further defined with the presence of aromatic rings with various possible branches (Vanholm *et al.* 2010). It is hard to decompose lignin structure, as can be clearly seen in Fig. 8, where it degrades slowly within the whole range oftemperature. Due to its complexity, lignin is thermally decomposed over a wide range and very low mass rate as compared to hemicellulose and cellulose, and because of that it is hard to see the peak of lignin (Ishak *et al.* 2011). Lignin is completely decomposed at a high temperature as 900 °C(Yang *et al.* 2007).Lignin is a very tough component and known as the compound that provides rigidity to the plant materials. It is responsible for providing stiffness to the cell wall and also bonds individual cells together in the middle lamella region (Vanholme *et al.* 2010).

The residual char at the conclusion of the TGA analysis represents the remaining materials after all volatiles materials have been eliminated throughout the pyrolysis process. Lignin was reported to be the main constituent of the residual char. Residual char of roselle fibre for 3, 6, and months old specimens was found to be 10.31, 14.21, and 12.24%, respectively. In this study, it was found that the char residue was closely related to the lignin content. This can be supported with the finding that lignin is the main component of the residual char. The highest lignin content was from 6-month-old roselle fiber. However, the percentages different of char residue did not show significant differences between them.

Table 6 shows thermal properties of roselle fibre in comparison with other bast fibres such as kenaf, hemp, ramie, and jute fibres. In conclusion, there is no significant effect on the plant ages of roselle fibre on the thermal behavior.From the comparison with other bast fibre, it can be seen that roselle fibres have good thermal stability compared with other established bast fibres such as kenaf and jute.

Table 6. Decomposition Temperatures for Selected Natural Fibre

Natural fibre	Temperature of initial decomposition (^oC)	Maximum decomposition temperature (^oC)	Reference
Flax	220	339.4	(Van de Velde & Baetens 2001)
Hemp	250	390	(Rosa *et al*. 2009)
Ramie	~240	380	(Gurumurthy & Radhalakshmi 2011)
Kenaf	219	284	(Rosa *et al.* 2009)
Jute	205	283	(Rosa *et al*. 2009)
Roselle 3 months	220	364.05	Current study
Roselle 6 months	200	363.24	Current study
Roselle 9 months	210	366.08	Current study

CONCLUSIONS

Roselle fibres are comparable with other established bast fibres in terms of their physical, chemical, tensile, and thermal properties. Three-month-old roselle fibre shows highest chemical content of cellulose, which contribute to the higher strength and thermal stability of the fibre. Nine-month-old roselle fibre has the highest average of diameter, as the diameter and density of roselle fibre increases as the plant matures. The moisture content and water absorption of roselle fibre decrease from 3 to 9 months old, and this is due to the reduction of cellulose content and lumen size. The result obtained from this study confirms the potential of roselle fibres to be used as reinforcement material in polymer composites in various applications.

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