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Twisted of Polyacrylonitrile /Lignin Nanoyarns for Enhanced Alignment and Rapid Stabilization

Intan Khazaimah Kushairi[1,2], Tamrin Nuge[2], Puteri Sri Melor Megat Yusoff[1], Mohamed Shuaib Mohamed Saheed[1,2,*]

[1] Department of Mechanical Engineering, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak, Malaysia
[2] Center of Innovative Nanostructures & Nanodevices (COINN), Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak, Malaysia

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ABSTRACT

Polyacrylonitrile **(**PAN) and PAN/lignin precursor fibers were fabricated using both conventional and modified electrospinning setups. The modified setup was designed to produce uniaxially aligned nanoyarns, applied in-situ tension and twisting to improve fiber alignment and reduce diameter. Twisted PAN fibers exhibited the best alignment and smallest diameter (~50 µm), while PAN mats showed broader alignment distribution with ~75 µm diameter. PAN/lignin nanoyarns had a larger average diameter (~150 µm) but retained good orientation. All twisted fibers were subjected to stabilization by varying temperatures and heating rates to evaluate their structural and chemical changes. Characterization using Fourier-transform infrared spectroscopy (FTIR) and thermogravimetric analysis (TGA) were analyzed to study the effects of different stabilization parameters. FTIR analysis revealed that lignin enhanced cyclization and oxidation at higher temperatures, indicated by a reduction in C≡N and C–H, and increase in C=N groups. At high temperature of 260˚C and at 10 ˚C/min temperature rate has reduced the stabilization duration tremendously by 60% without jeopardizing structures formation during stabilization process. These results highlight the effectiveness of the modified electrospinning setup and the potential of PAN/lignin nanoyarns for the subsequent carbonization stages as a greener and sustainable carbon fiber precursors, by improving the orientation and reducing the stabilization duration.

1. Introduction

The growing demand in automative and renewable energy for lightweight as well as high-strength materials in industries drives significant interest in carbon fibers. However, traditional carbon fibers are predominantly derived from crude-based polyacrylonitrile (PAN), which holds for 90% of the entire carbon fiber in the market [1]. As industries shift towards more sustainable alternatives, lignin, an abundant byproduct especially by the paper and biofuel industries, emerges as a promising renewable precursor for carbon fiber production due to its high carbon content and often compared to PAN for the cost-effectiveness.

* *Corresponding author.*
E-mail address: shuaib.saheed@utp.edu.my

Blending lignin with PAN has demonstrated improvements in fiber formation, precursor uniformity and cost-effective solution and reducing toxic emissions from the petroleum-based products [2]. However, studies on lignin-polymer blends highlight both the potential and the challenges associated with using lignin as a reinforcing component. One research [3] had previously discussed various polymer matrices combined with lignin and the associated drawbacks where lignin demonstrates poor compatibility due to its apolar nature, leading to phase separation and weak dispersion interactions.

Electrospinning has emerged as a preferred technique for producing nanofiber-based materials due to its exceptional ability to generate highly aligned, ultrafine fibers with tunable properties. Electrospinning provides greater control over fiber alignment, which is crucial for achieving superior mechanical [4] properties compared to other conventional spinning techniques [5] and cost-effective approach for manufacturing nanofiber yarns at an industrial scale [6]. Moreover, electrospun fibers exhibit enhanced compatibility with nanomaterials [7] , facilitates the fabrication of fibers with customizable morphologies [8]. This flexibility enables the development of tailored nanofiber architectures, whereby some study has proved by increasing the twisting level, the elongation at break also improved [9] and remains the leading method for fabricating fibers from lignin-based precursors.

Stabilization is a crucial process in carbon fiber production. Through this process, the linear polymer is transformed into a thermally stable and rigid ladder-like structure [1]. During stabilization, carbon content increased as oxygen and hydrogen levels gradually declined [10]. Therefore, the stabilization process is essential to convert fusible spun fibers into non-fusible fibers, where oxygen promotes lignin crosslinking and increases Tg during the process [11].

Despite the potential of lignin precursors as carbon fibers, several challenges persist, where it has relatively low mechanical strength due to irregular fiber alignment and uneven distribution during the conventional electrospinning process. Unaligned fibers–increase friction between individual fibers, thereby weakening fiber strength. Most published research focuses on PAN precursors [12,13], employing melt-spinning or wet-spinning methods for fiber formation, while some studies are conducted in batch processes, limiting their applicability to continuous carbon fiber production. This study aims to address these challenges through a modified electrospinning technique and systematic control of processing parameters during stabilization to increase fiber alignment and optimize stabilization duration hence increase time and resources management.

2. Experimental

The Polyacrylonitrile (PAN) was obtained from Sigma-Aldrich (product number 181315, average molecular weight (M_w) = 150,000). The alkaline lignin by Sigma-Aldrich. The N,N-Dimethylformamide (DMF) 99.99% was received from Sigma-Aldrich by Supelco. The materials were used directly, without modification.

Electrospinning dopes for PAN/lignin blends were prepared at a total concentration of 10 wt.% in DMF solvent. A 1.8 g amount of PAN powder was mixed into 20 mL of DMF in a 40 mL clear glass bottle, magnetic stirred with temperature at 60°C by speed of 220 rpm for 15 min or until the PAN was completely dissolved. Once the solution became homogeneous, A total of 0.2 g of alkaline lignin was added to the transparent PAN solution and mixed continuously for 45 min or until complete dissolution was achieved. A pristine PAN solution also prepared (PAN/lignin 100:0) as a benchmark.

Precursor fibres (PF) were prepared by two types of electrospinning setup to compare the fibre outcome. The first setup was using conventional setup, using single syringe pointed directly towards

a fibre collector (drum shaped) to get a precursor fibre mat. A 5 mL volume of the prepared solution was filled in a 10 mL syringe fitted with a 20-gauge needle. An automatic pump machine was used with a speed of 0.5 mL/min, while voltage was set (+10kV, -12kV). The second setup was using a modified electrospinning setup, where two syringes pointed towards the fiber collector (conical shaped) or known as a Neutral Metal Disk (NMD) to produce a Uniaxial Nanofiber Yarn (UANY). Two metal needle syringes with different velocity charge poles were positioned oppositely between the winder collector and a neutral metal disc. A 5 mL volume of the prepared solution was filled in a 10 mL syringe fitted with a flat-tip 18-gauge needle. The solution was automated pumped as mentioned above. The opposing voltages were applied between two needles to create oppositely charged polymer jets, where these jets were directed towards the NMD. Once the fiber formed on the NMD, next the NMD was rotated to form a stable spinning triangular cone between needles and the NDM, which resulted in the collection and twisting of aligned nanofibers into UANY. The UANY was then guided to the Winder Collector 1 (WC1) introducing twisting and tension at the same time. This setup was designed to enhance the alignment and mechanical qualities of the precursor fiber for the next stage of carbon fiber production. The difference between both setups is summarized in Table 1 below.

Table 1
Difference between conventional electrospinning versus modified electrospinning

	Conventional electrospinning	Modified electrospinning
Precursor syringe	Single source	Double sources
Voltage	+ve, ground	+ve, -ve, ground
Fiber collector	Drum shaped	Cone shaped
Yarn collector	No	Yes
Tension applicable	No	Yes
Twisting applicable	No	Yes
Fiber form	Mat	Twisted and non-twisted yarn

From the electrospinning process from both setups, samples were collected from precursor fiber from PAN-based in mat and yarn form, and PAN/lignin-based in yarn form and sent for FESEM analysis. Next, the morphology image was analyzed using ImageJ to collect fiber orientation and alignment data. The most aligned and oriented fibers subsequently proceeded to the next process which they were exposed to different stabilization parameters (temperature different and temperature rate) to explore the effect of stabilization techniques.

2.1 Stabilization Process

Initially, the fiber was exposed to different temperature conditions which were low temperature (LT) at 200˚C, medium temperature (MT) at 230˚C, and high temperature (HT) at 260˚C. The process was done with a fixed temperature rate (0.2 ˚C/min) in a furnace. The stabilized fiber (SF) was then sent for characterization to understand the changes to the fibers. Subsequently, different temperature rates (0.2, 0.5, 1.0, 2.3, 5.0, 10 ˚C/min) were studied to grasp more understanding of the effect of the temperature rate to the completion of different stabilization settings and stabilization duration.

2.2 Characterization

Thermogravimetric analysis (TGA) measurements were performed using a STA 6000 Perkin Elmer. The sample ranges from 25˚C to 900˚C with heating rate fixed at 10 ˚C/min exposed to nitrogen flow. For the morphology images, the equipment used for field emission scanning electron microscope (FESEM) was Clara, Tescan. After receiving the surface image of the data, the images were analysed in ImageJ for fibre diameter and orientation. The Fourier transform infrared (FTIR) characterization was using FT-IR spectrometer Frontier by Perkin Elmer. After the measurement, further analysis, the graphs were plotted using Origin software.

3. Results and Discussion

After spinning process was done, the precursor fiber mat and yarn were collected for PAN-based by conventional setup and yarn for PAN/lignin based by modified setup. The precursor fibers were characterized by FESEM to see the fiber morphology and subsequently process in ImageJ for orientation and diameter identification. The most oriented and lower diameter was then proceeded with various stabilization temperature and rate to understand the changes to the fibers during the pyrolysis.

Fig. 1 shows the fiber orientation angle of fiber mat (non-twisted fibers) from the conventional electrospinning process and yarn (twisted fibers) as well as less twisted fiber from modified electrospinning. The twisted yarn fiber shows the most oriented fiber where most of the fibers were accumulated at the 0˚ angle, while the less twisted and fiber mat has the scattered fiber where mostly at -40˚and 80˚ angle.

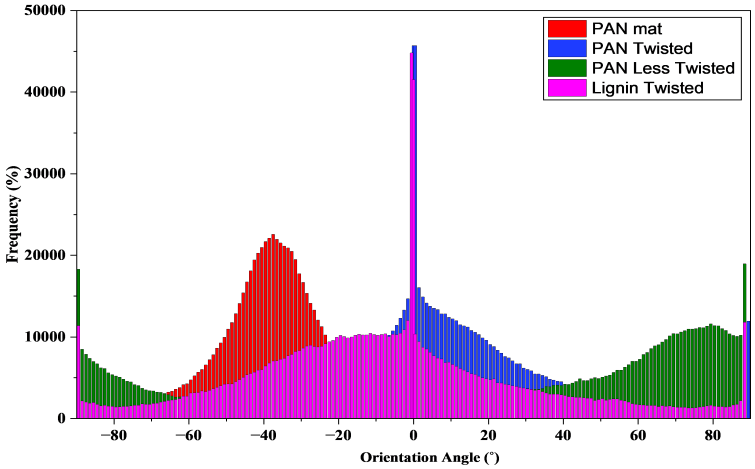

Fig. 1 Fiber orientation angle for fiber in the mat and twisted form from PAN and lignin-based precursor fiber

This works for both PAN and PAN/lignin-based precursors. From the ImageJ analysis, the fiber mat has ~25-75 µm diameter, twisted fibers range from 50-150 µm while the less twisted fiber shows the largest diameter distribution which is around 150-275 µm. Even though the fiber mat has the lowest diameter range, the twisted yarn fiber shows a notable orientation compared to less or non-twisted fiber. This was due to molecular alignment with high orientation introduced by the stretching process [14].

3.1 Temperature Difference during Stabilization

The PAN/lignin-based twisted precursor fiber was selected due to the best orientation with acceptable fiber diameter and sustainable option for the next process of stabilization. Different heating profiles were exposed to fiber from low temperature to high temperature at fixed heating rate (0.2 ˚C/min). Fig. 2 shows a comparison of the precursor fiber (PF) with stabilized fiber (SF) temperature ranging from 200, 230 and 260˚C. In the initial state of PF, the C≡N (2240 cm^{-1}) and C-H (1454 cm^{-1}) functional groups were noticeable. C=N (1600 cm^{-1}) structure has the least peak among C≡N and C-H structures. However, when the yarn (SF) treated at the highest temperature (HT), the C≡N structures show the most significant reduction of the peak intensity and in contrast, the C-H structure appeared to be reduced. The formation of C=N structures with the highest peak intensity compared to the other temperature (LT and MT) conditions show that there is a formation of "ladder structure" for the stabilized fiber [15]. Additionally, by adding lignin, the structure of C-O structure (1142 cm^{-1}) was observed to be stretched after stabilization, thereby indicating a successful blending of lignin into PAN.

At least 19 h of duration was required to complete one batch of stabilization process at HT at 0.2 ˚C/min rate. Hence, an attempt to reduce the duration time by increasing the temperature rate was made. The temperature rate increased until 10 ˚C/min at high temperature.

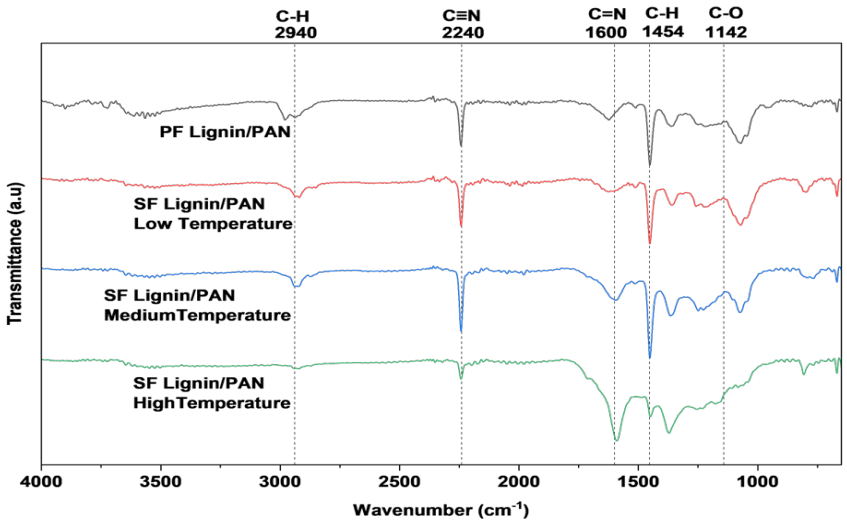

Fig. 2. FT-IR Spectra of fibers at different stabilization temperatures from low temperature to high temperature

Table 2 shows the stabilization duration at different temperature rates. When the temperature rate increased by two times, the stabilization time was reduced by more than 60%, which resulted in a total of 8 h of total duration for the whole stabilization process. Increasing the temperature rate until 10 ˚C/min, the stabilization duration was completed by less than half an hour.

FT-IR result in Fig. 3(A) shows the temperature rate at 2.3 ˚C/min has deeper intensity of C=N band around 1600 cm^{-1} and a weakened intensity at C≡N band around 2240 cm^{-1} and, C-H band around 1454 cm^{-1} compared to other temperature rates. The second temperature rate that shares the same trend with slightly weaker intensity and stretching is at temperature rate of 10 ˚C/min.

The initial degradation and weight loss of the material was analyzed with TGA analysis. Fig. 3(B) shows the degradation of weight (%) of different temperature rates. During the temperature of 260˚C, the temperature rate at 0.5 ˚C/min degrades slightly below the benchmark rate (0.2 ˚C/min), while 10 ˚C/min degrades slightly lower from the 0.5 ˚C/min. The sample decomposed more weight

at higher temperature rate. At 5 ˚C/min, the weight reduced around twice the weight of 2.3 ˚C/min. The lignin degradation of weight is known at 260˚C where lignin begins to depolymerize and releasing small volatile compounds and removal of moisture and cellulose from lignin [16].

Table 2
Stabilization duration at different temperature rate

Sample name	Temperature rate (˚C/min)	Stabilization time (min)	Stabilization time (h:min)
Lignin 0.2	0.2	1150	19.17
Lignin 0.5	0.5	460	7.67
Lignin 1.0	1.0	230	3.83
Lignin 2.3	2.3	100	1.67
Lignin 5.1	5.1	45.1	0.45
Lignin 10	10.0	23	0.23

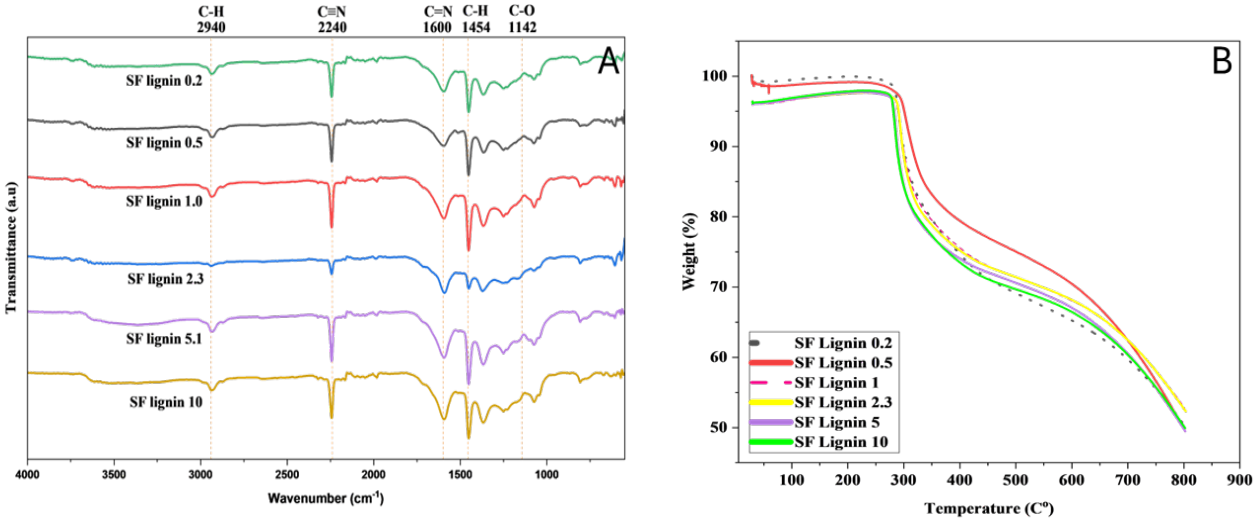

Fig. 3. (A) FT-IR spectra at different stabilization rates, (B) TGA curves at different stabilization rates

4. Conclusion

The process was started by selecting the best orientation and diameter reduction of fiber by comparing the conventional with modified setup. The precursor fiber produced from the conventional setup was in mat form while the yarn fiber was produced by the modified setup. Yarn precursor fiber has been selected for the next stabilization process since both diameter and orientation have been improved significantly. During the stabilization process, the temperature range of LT, MT, and HT were exposed to fiber. From the FTIR outcome, during stabilization, lignin has imposed a high intensity peak for C=N and a broad spectrum for C-O, showing the successful stabilization process in lignin-based precursor fiber at high temperature compared to low and medium temperature. All stabilization rates were fixed at 0.2 ˚C/min. Next, to reduce the stabilization time, temperature rates were varied to see the effect on the fibers. FTIR results show an almost identical spectra trend for all the rates except the at 2.3 and 10 ˚C/min. To confirm the result, TGA analysis was done and from the graph, even though 0.5 has the closest weight reduction compared to 0.2 C/min, however, the second closest result to the mass decomposed while undergoing stabilization was 10 ˚C/min. Hence, to reduce the stabilization time, which lead to lower energy and time consumption, the temperature rate of 10 ˚C/min at 260 ˚C temperature condition was preferred

without compromising the effect to crystallinity formation and structures formation in lignin-based stabilized fiber. This outcome could improve time management during stabilization tremendously while preparing for the next stage of carbon fiber which is carbonization process.

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