

A Review on Centrifugal and Electro-Centrifugal Spinning as New Methods of Nanofibers Fabrication

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Abstract- In the recent years, by developing the methods of nanofibers formation, electro-centrifugal spinning has been introduced as a new method of nanofibers fabrication. This nanofabrication technique is a combination of known technique called electrospinning and a new coming method called centrifugal spinning. This paper is a comparative study among conventional electrospinning, centrifugal spinning and the basis of electro-centrifugal spinning methods. For instance, although, the distance of nozzle to collector is a critical operating parameter to determining nanofibers diameter in electrospinning method, it only shows an effect on the morphology and has no significant effect on the fiber diameter in centrifugal spinning. Surface tension and viscosity of the solutions are the spinning-ability determinatives in these three methods which are affected by the type of polymers and solvents and also the concentration of the solution, and need to be overcome through electrical or centrifugal forces or both. The effective parameters on the process and the fiber morphology are investigated for each of the three methods.

Keywords: surface tension, viscosity, rotating velocity, morphology of nanofibers, orifice diameter, applied voltage

I. INTRODUCTION

Nowadays, nanostructure materials have attracted enormous research interests in versatile fields of studies. Among different types of nanostructures, one-dimensional nanostructured materials such as nanofibers, nanowires, nanotubes and nano-rods have attracted extensive attention due to their unique physical and chemical

characteristics [1]. Because of nanofibers flexibility and continuous form, they have been considered as a promising nanostructure in different applications. Although, the fibers with diameter below 100 nm were called nanofibers in their initial definition, in the recent years, all fibers with less than 1 μm in diameter are considered as nanofibers [2].

In nanofibers fabrications, due to the high surface-to-volume ratio, great mechanical and thermal properties and nanofibers special assemblies, there are some important advantages that lead to a 3-D and porous structure with excellent pore interconnectivity [3-7]. Combining nanofibers unique properties with the special materials functionalities would result in novel properties and applications. Polymers have attracted much interest among different types of materials due to their availability, easy production and flexible properties [8]. Therefore, polymeric nanofibers are considered as optimal candidates for many applications in different industrial features such as nano-filtration, (bio)chemical sensors, cosmetic skin masks, nano-medicine, tissue engineering, wound dressing, drug delivery carriers, electronic devices, etc. [9-12].

There are many literatures in the case of polymer nanofibers which are classified in three branches. First one is related to the nanofibers made from biopolymers such as chitin, alginate, chitosan, etc. [13-15]; the second branch is nanofibers produced from synthetic polymers such as different degradable polymers like poly(α -hydroxy esters), including poly(caprolactone) (PCL) [16], poly(glycolic acid) (PGA) [17], poly(lactic acid) (PLA) [18], and their co-polymer (PLGA and PLLA) [19,20]; and the third one is related to blend or hybrid polymer nanofibers in forms of synthetic-synthetic (such as PCL-PEG [21]), natural-natural (like chitosan-gelatin [22]) and natural-synthetic such as chitosan-PVA [23,24], chitosan-PCL [25-27], etc.

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Due to the high potential of nanofibers in the mentioned applications, it is desirable to create different methods for making nanofibers to approach the technique in which nanofibers can be produced in the fastest time, with the highest quality and lowest price [2]. In this way, various techniques such as splitting of bicomponent fibers [28], melt-blowing [29], physical drawing [30], flash-spinning [31], phase separation [32], self-assembling [33], solvent dispersion [34], centrifugal spinning [35], hydrothermal [36] and electrospinning [3] have been developed. These techniques are divided into two classes called top-to-down and bottom-up approaches based on the precursors scale comparing product scale (Fig. 1). For instance, self-assembly is a bottom-up technique, because of fabricating nanofibers as products from individual molecules as precursors. While electrospinning is a top-to-down technique due to creating nanofibers from polymeric solutions. All the mentioned techniques have weaknesses and also strengths in their processes. Among all approaches, electrospinning has been known as the most flexible method in designing nanofibers morphology [3,37].

During the recent decades, many efforts have been developed to improve the orientation and alignment of nanofibers for applying in special fields [38]. Rotating jet method (RJM) as a technique of electrospinning was developed by Dabirian *et al.* [39]. They used a stationary hollow metallic cylinder as a collector to align nanofibers. Khamforoush *et al.* [40,41] modified RJM by replacing the cylindrical collector with two metallic concentric hollow cylinders. The author reported that this modification successfully doubled the degree of alignment and increased the fabrication rate of nanofibers up to 41% [41]. Wu and Qin [42] used two metal needles in an opposite way for obtaining aligned nanofiber. Centrifugal jet spinning is another technique which is introduced for obtaining oriented fibers. In an effective effort, Ren *et al.* [35] found that the centrifugal technique is extremely practical

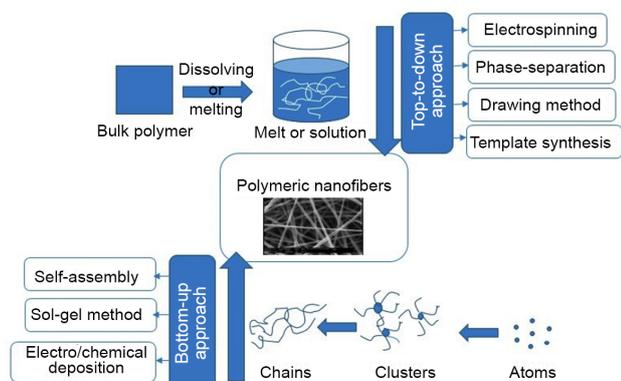


Fig. 1. Top-to-down and bottom-up approaches in fabrication of nanofibers.

to fabricate micron to nano-scaled fibers with an upper production rate limit of $0.5 \text{ g} \cdot \text{min}^{-1}$. Luo *et al.* [43] reported further information about conventional and modern electrospinning methods with the aim of comparing them in a comprehensive review. In 2010, Dabirian *et al.* [44,45] developed a unique technique including both electrostatic and centrifugal forces called electro-centrifuge spinning (ECS). This method has a prominent feature to produce aligned nanofibers [46]. Khamforoush *et al.* [38] modified the ECS method using two nozzles to increase the production rate of uniaxial nanofibers.

In this study, centrifugal spinning and electro-centrifugal spinning are considered as two novel techniques for fabricating nanofibers with aligned morphology. The aim of this study is to investigate these two methods and make a comparison between them. Effects of different parameters on produced nanofibers morphologies based on these two methods are also reviewed.

II. ELECTROSPINNING

Electrospinning is a simple, non-mechanical and also versatile technique in fabricating nanofibers by applying electrical forces. As it is known, this process has a long history among nanofibers fabrication techniques. There were some millstones in progressing electrospinning technique during the time. These progresses were done in 1902 by Cooley and Morton, 1914 by John Zeleny, 1930-1940 by Formhals, 1964-1969 by Sir Geoffrey Ingram Taylor and 1990 by Reneker [47,48].

Fig. 2 shows a conventional single-nozzle electrospinning system called conventional electrospinning. The set-up contains a syringe, a syringe pump, a metal nozzle, a power supply, and a collector. First the syringe of polymer solution would adjust on the syringe pump with appropriate extrusion rate. During electrospinning, a high voltage is applied between the nozzle and the collector. When the voltage reaches a critical value, the electrically-charged fluid generates a conical droplet (i.e., Taylor cone), from which a liquid jet is formed and elongated. The electrically

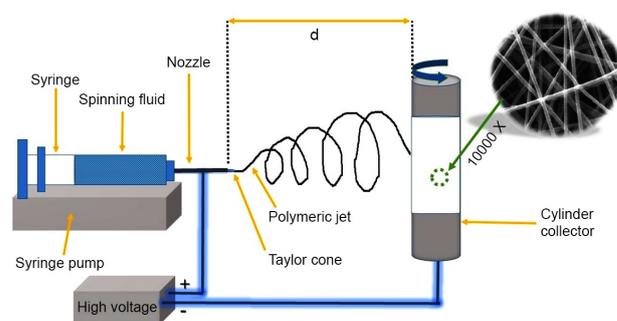


Fig. 2. Schematic representation of conventional electrospinning system.

charged jet then undergoes a stretching-and-whipping process, during which the jet diameter decreases from hundreds of micrometers to as small as tens of nanometers. The as-spun nanofibers are accumulated on the surface of the grounded collector [49].

Due to its high potential in different applications, it has attracted many research interests in variety fields from electrospinning of different kinds of materials for various applications to modifying electrospinning technique to obtain new procedures in electrospinning. These modified techniques are desired as they are more controllable in produced morphologies and also very effective in facilitating process conditions. Some of these modified techniques are improved to increase fiber alignment [50] such as rotating drum or disc collection, parallel pair electrodes complex, step collector, frame collector, auxiliary electric or magnetic electrospinning, centrifugal electrospinning, and electro conductive template setup [51]. The other ones are the techniques developed to overcome drawbacks of conventional electrospinning such as bubble-electrospinning [52], gas coating-electrospinning [53], nozzle-less electrospinning [54], rotating jet method (RJM) [39], magnetic electrospinning [55], centrifugal electrospinning [44], etc.

Among these methods, rotating jet method (RJM), centrifugal electrospinning and electro-centrifugal spinning are new methods to fabricate aligned nanofibers. In recent years, electro-centrifugal spinning method is introduced as a technique which beneficially merges the advantages of centrifugal spinning [56-58] and conventional electrospinning. This technique provides higher percentage aligned fibers follows by higher rate of fabrication at a lower applied voltage or slower rotating speed.

III. CENTRIFUGAL SPINNING

The centrifugal spinning is a well-known method especially in fiberglass industry for producing glass fibers which also called "glass wool" with average diameter of greater than 1 μm . Although this technique has a wide application in fiberglass industry, it is newly used in producing polymeric fibers, especially polymeric nanofibers [57]. Centrifugal spinning for nanofiber fabrication known as rotary jet spinning (RJS) has been introduced by Badrosamay *et al.* [58] since 2010 as an alternative nanofiber formation technique inspiring the cotton-candy production principle. The initial patent related to this technique is filed by Lozano *et al.* [59] in 2012. Its first commercial machine was introduced to market under the brand name of ForcespinningTM by FibRio[®] Technology Co. [60]. The technique can overcome some of the disadvantages of electrospinning, such as its low production rate and the

essential high voltage for industrial applications [58,61]. Due to the mentioned assets, centrifugal spinning has been considered as one of the most promising nanofibers formation techniques. Basically, in this technique, nanofibers are produced by centrifugal forces. Generally, a rotating chamber including a number of orifices would be filled with a polymer solution or melt [56]. Rotation causes centrifugal force that is responsible to guide the solution/melt towards the orifices by pushing it to the inner surfaces of the chamber. The polymer jet exits from the orifices as soon as the centrifugal force overcomes the viscosity and the surface tension of the solution or melt. Then during the jet flight towards the collector, it is stretched enough and the solvent is evaporated or the melt is cooled to form nano-scaled and dried fibers [58,62,63]. A schematic procedure of a centrifugal spinning device is shown in Fig. 3.

Wang *et al.* [64] demonstrated the centrifugal spinning as a mass production amenable technology for polymer fibers fabrication. They used centrifugal melt spinning (CMS) technique to fabricate a 3D fibrous web of PLGA and solvent-assisted centrifugal spinning technique to produce a 3D fibrous web of polystyrene, using two commercial cotton candy machines. In a valuable review paper, Stojanovska *et al.* [61] have investigated different aspects of various parameters such as angular speed (ω) of the chamber, the polymer mass (m) inside it and the chamber radius (R) which affect the centrifugal force (F_c). Literatures divided the general mechanism of centrifugal spinning to three stages of jet initiation, jet expansion and fiber formation along with solvent evaporation [61,65]. They concluded that the rotating speed must be beyond a certain critical angular velocity for initial jet exit. After that the jet is affected by elongation force which is a combination of the drag force (F_d), the Coriolis force (F_{cor}), the centrifugal force (F_c), the viscous force (F_v), and the resulting force from the Coriolis (F_r). In the third stage, the

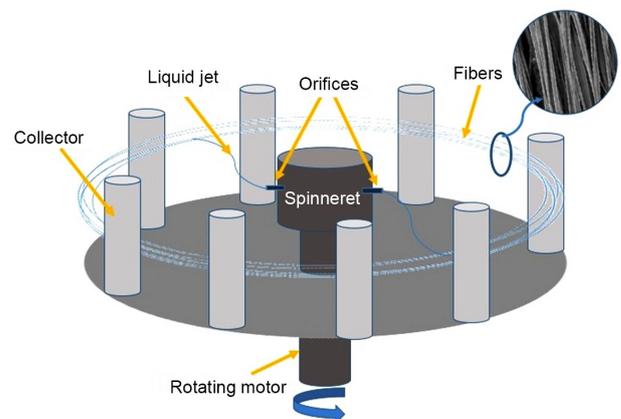


Fig. 3. Schematic representation of conventional centrifugal spinning system.

distance between the orifice and the collector becomes more important to provide enough time for solvent evaporation [61].

In order to apply a high rate of tension to a polymer solution or melt, methods are required, which can apply uniform distribution of stress [65,66]. During applying the centrifugal force on a solution, it provides uniform distribution of stresses. Consequently, it can be applied to provide high rates of tension on a polymer solution [67]. In this technique, a polymer solution with a proper viscosity would be stretched as a thread and turned to a polymeric fiber during drying process [67,68]. Weitz *et al.* [69] designed a nozzle-free centrifugal spinning force to produce polymer nanofiber. As it is known, the viscosity of the polymer solution increases due to the frictional forces between the polymer chains. So, the speed of the applied exterior forces can change the frictional forces. When the rate of external forces increases, the frictional forces between the polymer chains increase against it and cause a higher amount of viscosity. Consequently, with an increase in the applied external forces, the solutions with lower concentrations can be spun. Air movement is an environmental parameter which can affect fibers morphology and orientation by impressing rate of solvent evaporation and fibers alignment. Mary *et al.* [70] reported that the obtained PCL/PVP nanofibers produced through centrifugal spinning showed high alignment due to the air flow controller which was placed next to the orifice. When the polymer jets are ejected from the narrow orifice under high centrifugal force, the air controller pushes the fibers down to collect at the bottom with higher degree of alignment.

There are different parameters influence the morphology of nanofibers spun by centrifugal spinning. These parameters are divided into three different groups. The first group includes the solution properties such as viscosity, surface tension, concentration, vapor pressure of solvent, solution density, etc. [65]. The second one involves the operating parameters such as rotating speed, the distance between orifice to collector, temperature, etc. And the third group of parameters is related to the mechanical objects of the devices such as the number and diameter of the orifice(s) and collector(s) [67,68]. Although, there are various influential parameters on the quality of nanofibers [58,66], the most effective ones are considered in this study. Therefore, the effect of surface tension, viscosity, speed of spinneret, distance to collector, evaporation rate and diameter of the orifice on the nanofibers morphology is studied. The other parameters show moderate effects, so these parameters can be neglected. Table I presents a brief overview on different polymers which were spun under

various centrifugal spinning parameters.

A. Surface Tension

The surface tension of a polymer solution is a key parameter in fabrication of nanofibers in all spinning techniques. The fiber morphology as well as the rheological behavior is dependent on the surface tension [71]. In centrifugal spinning, the centrifugal force and the surface tension are two most powerful parameters which affect the morphology of nanofibers [56,58,59]. Surface tension is affected by the type of polymer(s) and solvent(s), the solution concentration and viscosity, temperature, etc. [66-68]. Generally, the surface tension of a polymer solution inhibits the spinning process because it results in jet instability [72]. To decrease the surface energy, a high surface tension tends to form a droplet of solution. On the other hands, the centrifugal force can assemble the polymeric droplets together to form a uniform jet of polymer solution [71]. So, the centrifugal force needs to exceed the surface tension to make a proper condition for nanofibers formation [58,66,71]. Thus, at the main stage of the procedure, the centrifugal force must overcome the surface tension to form the Taylor cone at the tip of orifice and make the polymer jet eject towards the collector. When other parameters are optimized, reducing the surface tension of the material will help to form the nanofibers [68].

Weitz *et al.* [69] reported that under all investigated parameters, polymer beads were formed as a derivative of the spinning process according to the amount of surface tension and the Rayleigh-Taylor instability. They concluded that surface tension acts to decrease the surface area by changing the jets into spheres. This report was in line with the Lu *et al.* [71] findings, who reported that no fibers were formed when the PAN solution concentration was lower than 6 wt% and by increasing the PAN concentration to 8% fibers were formed followed by beads due to high surface tension. They demonstrated that the surface tension of PAN solutions increases when the polymer concentration increases. However, as compared with the rapidly-changing viscosity, the increase in surface tension is relatively small when the solution concentration increases. Furthermore, the results show that with a further increase in the concentration of PAN solution in the range of 10 wt% to 15 wt%, the morphology changed to higher amount of fibers followed by few beads with an increase in fibers diameters from 406 nm to about 1077 nm. They concluded that further increase in concentration (above 10 wt%) showed significant effects on solution viscosity and slight effects on increasing surface tension. So, chain entanglement had an effective role on jet stabilization to form bead-less continuous fibers [71].

B. Viscosity of Solution

Viscosity of the polymer solution or melt is another parameter that must be taken under control to form a proper morphology of nanofibers. This parameter is affected by the temperature and the solution properties and directly influences the surface tension. So, it needs to be overcome by centrifugal force to form a proper polymeric jet. If the viscosity would be high, the centrifugal forces may not be strong enough to create a jet. This fact is due to the large interaction forces between the molecules due to the large viscosity that will make the molecules coiled

together, so, the external force cannot enforce the solution out of the orifice [73]. On the other hand, if the viscosity is too small, by applying centrifugal forces, the droplets will form instead of jets [65]. Moreover, the viscosity of solution will affect the solution velocity as well as the morphology of produced nanofibers. By decreasing the viscosity, the velocity of the solution increases and the diameter of nanofibers decreases [65]. This parameter is also affected by polymer molecular weight. In a centrifugal spinning effort, it was reported that a driven jet of a low molecular weight liquid forms spherical droplets. On

TABLE I
A BRIEF OVERVIEW ON DIFFERENT CENTRIFUGAL SPUN FIBERS

Polymer compound	Concentration wt%		Spinneret speed (rpm)	Distance to collector (cm)	Orifice diameter (mm)	Morphology	Average diameter of fibers (nm)	Ref. [no]			
PAN	<6%		4000	10	0.4	No fiber	---	71			
	8%		4000	10	0.4	More beads		71			
	10%		4000	10	0.4	Low beads	406±108	71			
	13%		2000	10	0.4	Fibers	663±232	57			
	13%		4000	10	0.4	Fibers	440±118	57			
	14%		4000	10	0.4	Fibers	665±114	57			
	14%		4000	10	1	Fibers	895±256	57			
	15%		4000	10	0.4	Fibers	1077±302	71			
PAN:PMMA	16%	90:10	4000	10	0.4	Fibers	≈5500	74			
		70:30	4000	10	0.4	Fibers	≈2500				
		50:50	4000	10	0.4	Fibers	≈1000				
PA6	<15%		4000	10	0.16	No fiber	---	77			
	17.5%		4000	10	0.16	Fibers	803				
	22.5%		4000	10	0.16	Fibers	1113				
PCL:PVP	12.5%	100:0	2000	A bottom collector very close to spinneret hole	Not reported	Highly aligned fibers	311	70			
		70:30					352				
		50:50					619				
		30:70					726				
		0:100					823				
PCL:PVP + 2 w/v% tetracycline	12.5%	50:50	2000	A bottom collector very close to spinneret hole	Not reported	Highly aligned fibers	927	70			
Amylose corn starch	15%	3000		6	0.26	Fibers-beads	1400±200	79			
							Amylopectin corn starch		14%	Fibers-beads	1300±400
							Amylopectin potato starch		11%	Fibers-beads	1500±600
							Waxy maize starch		8-16%	Only beads	---
PVP-BaTiO ₃	10%	7000		15	0.4	Fibers	1497	66			
		9000		15	0.4	Fibers	788				

the other hand, viscoelastic forces in polymer solutions resist rapid changes in shape, leading to beaded fibers due to transformation of the coiled macromolecules of the dissolved polymer by the elongational flow of the jet into oriented and entangled networks. It was also mentioned that the bead diameter increases as the viscosity increases, as is expected for the break-up of a liquid jet due to the Rayleigh-Taylor instability [68]. Lu *et al.* [74] prepared poly(acrylonitrile)/poly(methyl methacrylate) (PAN/PMMA) fibers through centrifugal spinning as precursor of porous carbon nanofibers (PCNFs). They reported that the morphology of fibers from different ratio of PAN/PMMA (9/1, 7/3, and 5/5) showed bead-less smooth fibers and the average diameter of fibers decreased by increasing PMMA content. And this significant reduction in fiber diameter is due to the lower molecular weight of PMMA component in the blend which results in lower viscosity and less polymer chain entanglement [74].

C. The Rotating Velocity of the Spinneret

As it was mentioned, the solution is forced by the surface tension and centrifugal force, and the speed of the spinneret directly affects the amount of centrifugal force. So, the speed of the spinneret or rotating velocity will determine the production rate of nanofibers as well as the morphology of produced nanofibers. Wang *et al.* [64] concluded that if the spinning speed of centrifugal melt spinning is too low, the “pulling” force is not sufficient to produce homogenous fibers, resulting in many beads of diameter about 200 μm in the PLGA fiber scaffolds. In another word, at a very low speed, the centrifugal force cannot overcome the surface tension, and consequently the polymer jet will not eject through the orifice. When the rotating speed is properly adjusted, the quality and morphology of the nanofibers are considerable [75-78]. If the spinning speed is too high, fibers can be easily bound to form fiber clusters, therefore, the average fiber diameter decreases continually with the increase of the spinning speed [64]. Generally, in a determined range of speed, with an increase in the speed, the diameter of the nanofibers will decrease due to larger effective forces applied on polymer jet. Weitz *et al.* [69] reported that PMMA nanofibers with average diameter of 25 nm were obtained from centrifugal spinning of 5% solution at a rotating speed of 3000 rpm.

Zhang *et al.* [65] exhibited that in some experiments the rotational speed about 14000 rpm is one of the best speeds to fabricate PAN nanofibers through centrifugal spinning. When the speed of the spinneret is increased and the ejected jet is expanded, finer fibers with more uniformity in size are produced. Based on the study reported by Zhang *et al.* [57] at constant parameters of 13% PAN, 10 cm

nozzle-collector distance and 0.4 mm inner diameter of nozzle and by increasing the rotational speed from 2000 rpm to 4000 rpm, the average diameter of fibers decreased from 663 ± 232 nm to 440 ± 118 nm.

D. The Spinneret to Collector Distance

The spinneret to collector distance is another operating parameter which shows the effects on the morphology of nanofibers, but does not affect its diameter. Although the diameter of nanofibers does not show a significant change by changing the distance of the collector, there are some changes in the morphology of the nanofibers such as twisting and/ or breaking [71,76]. However, changing distance will not directly affect the diameter, but this parameter must be changed by different amounts of other parameters to produce uniform morphology. For instance, if the speed of the spinneret increases, the distance of the collector should also increase due to expanding the trajectory of the material. On the other hand, by increasing the viscosity of the solution, the speed of trajectory will decrease, so the distance of the collector should decrease [65]. It also should be noticed that when the distance to the collector is not long enough, the fiber will not be allowed to stretch enough and fine fibers will not be produced. The sufficient distance is determined based on the time required for solvent to evaporate.

Zhang *et al.* [57] also reported the centrifugal spinning of PAN under different nozzle-collector distances (10 cm to 30 cm) and showed that increasing the distance has a slight effect on decreasing fibers diameters.

E. Rate of Solvent Evaporation

The evaporation rate of the solvent from polymer solution is one of the important parameters that affects the nanofibrous morphology in all spinning techniques. Lu *et al.* [71] reported that centrifugal spinning leads to extensively stretching of the jets and accompanies solvent evaporation. During ejecting the jet from the spinneret and flying toward the collector, the solvent will rapidly evaporate from the jet [77]. If the rate of evaporation is too low, there is not enough time to get a solid fiber and the wet ones may form a thin film by joining together on the surface of the collector. When the rate of solvent evaporation is too high, then the jet gets dry very soon and stretching process is disturbed which causes producing thicker fibers [68]. Overall, the rate of solvent evaporation can be affected by a lot of factors such as the rate of applied heat, boiling point and surface tension of the solvent, vapor pressure, interaction between solution components, nozzle to collector distance and air velocity on the surface of jet [57,79,78]. Li *et al.* [79] reported that by applying a 50-100 $^{\circ}\text{C}$

hot air under an air stream velocity of 190–200 L/min, an increase in the rate of evaporation was observed. Zhang *et al.* [57] concluded that 10 cm is a sufficiently long distance for solvent evaporation and the distance greater than 10 cm has negligible influence on the fiber diameter.

F. Orifice Diameter

The diameter of an orifice is a critical parameter of machine devices which directly affects the solution flow and the fiber diameter. If the inner diameter of orifice is too fine, many of polymer solutions because of their higher viscosity cannot penetrate through the nozzle and this will limit the functionality of the device. On the other hand, if the diameter of orifice is too large, it will result in forming the droplets [35,66]. This fact may be due to the smaller friction force between the droplet formed in the tip of orifice which leads to removing higher amount of solution and no jet will form. If the orifice is sufficiently fine, nanofibers will be produced easily. Based on a number of reports, decreasing in the diameter of orifice decreases the final diameter of the produced fibers [35,57]. Li *et al.* [79] used needles with inner-diameter of 0.26 mm as nozzles for centrifugal spinning of amylose from different types of starch at different concentrations (11-15%) and obtained an average fiber diameter greater than one micrometer. Zhang *et al.* [57] investigated the effect of nozzle diameter on PAN fibers morphology using nozzles with different inner diameters of 0.4, 0.8, and 1 mm at 14% PAN concentration, rotational speed of 4000 rpm and nozzle-collector distance of 10 cm. They revealed that by increasing the diameter of nozzle from 0.4 mm to 1 mm the average diameter of fibers increased from 665 ± 114 nm to 895 ± 256 nm.

IV. ELECTRO-CENTRIFUGAL SPINNING

As it was mentioned earlier, although electrospinning is perhaps the most versatile process in producing nanofibers, increasing the production rate of nanofibers in this technique has always been one of the most important aims of researchers. Most efforts in this regard have been focused on increasing the number of nozzles [80,81]. In another approach, attempt has been made with the development of electro-mechanical system. Some researchers have combined electrospinning with centrifugal spinning to fabricate nano-scale fibers [44,82,83]. This technique is called electro-centrifugal spinning and includes high voltage supply with electrodes attached to the orifice fixed on a rotating solution chamber and the inner wall of a cylinder centered with the solution chamber. Technically, when a drop of the solution is transferred to the surface of the orifice, it can be electrically charged with positive charges immediately. The charged polymeric jet is

stretched along the rotating surface under the forces of the electrical field as well as the centrifugal field [84]. This unique combination would result in high alignment of the produced fibers as well as higher rate of production at a lower applied voltage or slower rotating speed [83].

Electro-centrifugal spinning, as a novel innovation, has been limited by a stream of rotating air surrounding the orifice. Exposing the ejected liquid jet to the high velocity airflow causes the jet to lose its solvent rapidly and as a consequence, the extension of the jet becomes more difficult, resulting in thicker nanofibers [51].

Basically, in this apparatus, the rotating polymer solution container with the orifice (nozzle) or a syringe containing spinning solution is attached to a rotating shaft and is centered at a hollow metallic cylinder as the collector. Both the collector and shaft are connected to a high voltage supplier. The spun fibers will pull towards the immobile cylinder. Due to centrifugal force and opposite charges between the nozzle and the collector (because of applying high voltage), the emerging jet, from the top of Taylor Cone formed at nozzle tip, rotates onto the interior surface of the cylinder. During the tension process, the jet passes a straight path towards the collector followed by a spiral path due to bending instability. Through this way, the jets are elongated by the repulsive force of charges on the surface [39,44]. Fibers with nano-scale diameter are fabricated during the drying of jets solidify and collecting on the cylindrical collector. Fig. 4 exhibits the schematic of electro-centrifugal spinning apparatus.

Although, the general set up for electro-centrifugal spinning is based on simultaneous use of electrostatic and centrifugal forces, different studies have prepared different set-ups by adjusting the configuration and size of different parts of device. Table II presents brief report different polymeric nanofibers fabricated through electro-centrifugal spinning with or without modification.

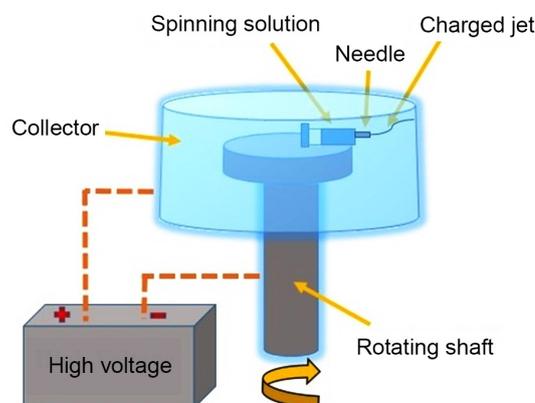


Fig. 4. Schematic representation of basically electro-centrifugal spinning system.

Dabirian *et al.* [44,45] have developed an electro-centrifugal spinning apparatus included a needle of 18 mm in length with interior and exterior diameter of 160 and 300 μm , respectively which was attached to a container with 4.6 mm interior diameter to form a nozzle. The nozzle was located on a circular plate which was connected to the positive electrode. The collector was a 26.6 cm diameter and 10 cm height metallic cylinder which was attached to the negative electrode of power supply. They assembled the set-up in a way which provided an 8 cm distance between the needle tip and the collector. To overcome the premature drying of polymer solution on the tip of nozzle due to

surrounding air flow, they used a cylinder surrounded the nozzle and container and only 2 mm of nozzle was left uncovered. Consequently, the solvent vaporization rate is reduced and the jet has enough time to be stretched before drying.

Liu *et al.* [51] used new configuration including a syringe with a right-angle needle which was fixed horizontally on a circular platform called "support disk". Fig. 5 shows a schematic of an apparatus similar to the one made by Liu and coworkers. The disc was attached through an axis to a speed-adjustable motor, and the distance of the needle tip to the shaft was about 16 cm. A static aluminum foil

TABLE II
FABRICATING NANOFIBERS THROUGH ELECTRO-CENTRIFUGAL SPINNING METHOD WITH OR WITHOUT ASSISTING TECHNIQUE

Assisting technique	Polymer compound	Concentration (%)	Spinneret speed (rpm)	Distance to collector (cm)	Applied voltage (kV)	Morphology (alignment)	Average diameter (nm)	Ref.
---	PAN (Mw=100000)	15	6360	8	15	Semi-aligned	≈ 430	45
		16	6360	8	15	Highly aligned	≈ 440	
---	PAN (Mw=100000)	13	1440	8	15	Random	285 \pm 87	83
			2160				255 \pm 38	
			2880				298 \pm 54	
			3600				290 \pm 38	
			4320				291 \pm 53	
			5040				251 \pm 32	
Air-sealed spinning head	PAN (Mw=100000)	13	1440	8	15	Random	267 \pm 62	83
			2160				230 \pm 47	
			2880				199 \pm 34	
			3600				136 \pm 19	
			4320				134 \pm 20	
			5040				142 \pm 20	
---	PAN (Mw=100000)	16.5	2650	8	20	Highly aligned	763 \pm 47	46
			2650		22		812 \pm 39	
Support disk	PMMA (Mw=250000)	13	420	2	4.5	Highly aligned	Not reported	51
		13			6	Semi aligned		
		13			7	Random		
		8			4.5	Beaded/disorder		
		10			4.5	Semi aligned		
		15			4.5	Aligned		
Support disk	PS	15	420	2	5	Fiber/bead	Not reported	51
		18	420	2.5	3	Highly aligned		
Support disk	PVP	8	360	3	6.2	Coiled fibers	Not-reported	51
			420		6.2	Highly aligned		
			540		6.2	Aligned		
			420		4	Semi-aligned		
			420		6	Highly aligned		
			420		8	Coiled fibers		
Air-sealed spinning head	PLLA	10	2000	8	15	Random	946 \pm 547	83
			3000				682 \pm 252	
			4000				900 \pm 414	

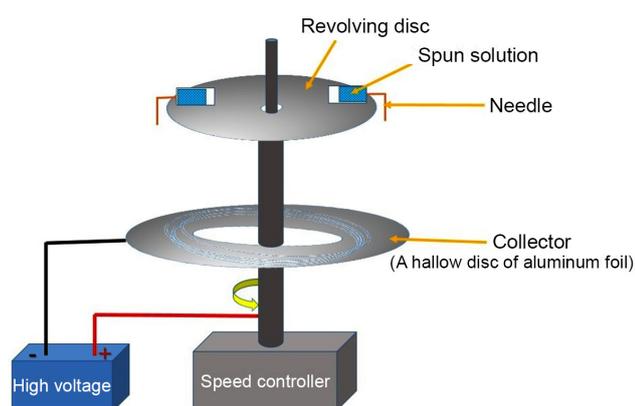


Fig. 5. Schematic illustration of electro-centrifugal spinning using a disc collector.

with 20 cm inner diameter and 44 cm outer diameter was selected as a collector which was located under the support disk. When high voltage was applied between the needle and collector, fibers with ultrafine diameter are collected on the static annular collector.

Valipouri *et al.* [83] prepared Air-sealed Centrifuge Electrospinning (ASCES) set-up to fabricate nanofibers. At their produced apparatus, the rotating cylindrical receptacle holds a syringe containing polymer solution and a 300 μm outer-diameter needle with 18 mm length was attached to the syringe. Positive electrode from high-voltage power supply was connected to the nozzle and the surrounding cylindrical collector was attached to the opposite polarity. The distances from the nozzle tip to the rotation center and to the collector were adjusted to 5.3 and 8 cm, respectively. A movable transparent door was used to prevent air from entering and exiting.

Khamforoush *et al.* [38] modified the electro-centrifugal spinning technique by equipping it with more nozzles (two or four nozzles) as the spinneret for the aim of increasing the production rate of highly aligned nanofibers. This study concluded that although increasing the number of nozzles had no effect on the degree of alignment, the rate of nanofibers production increased considerably. They reported that the setup with two nozzles obviously doubled the production rate of nanofibers relative to the conventional Electro-centrifugal spinning.

A. Effect of Collecting Distance

The collecting distance in conventional electrospinning is generally about 10 cm to 20 cm. According to the Reneker's theory [85], the jets curve under the bending instability, and then when several turns are formed, a new electrical bending instability caused lots of new smaller bending coils along with the previous larger coils, until the jets dried into nanofibers. When the collecting distance is about 5 cm,

the bending occurs before the jet reaches to the surface of collector [71,72]. Dabirian *et al.* [45] observed that in an electro-centrifugal spinning device, two distances must be set at the optimum values, which are the distance between the center of rotation and the needle tip and between the center of the rotation and collector surface. The authors reported that for the amounts below the optimum points, wet fibers reach the collector surface whereas above the optimum point, fibers are dispersed in the cylinders space before reaching to the collector. The setup was assembled in which the distances between the tip of needle and the collector was about 8 cm. They prepared a 16% PAN in DMF for spinning through electro-centrifugal spinning at voltage of 15 kV, rotating speed of 6360 rpm and the distance of 8 cm.

Liu *et al.* [51] revealed that in an electro-centrifugal spinning device, centrifugal force supplies a horizontal velocity for the bending jet to be forced to move in horizontal direction. Therefore, the dried collected fibers become straight and aligned. To produce a uniformity in all fibers morphology, the distance between nozzle and collector should be controlled within the first bending distance. According to literature, the straight segment of the jets is about 1 cm [86], thus, they proposed that the collecting distance should be between 2 and 4 cm. From their study, shorter distances do not help to increase the rate of solvent evaporation and jet solidification; and larger distances need higher rotational speeds for stretching the jets to produce straight and aligned fibers. In an effort, Liu *et al.* worked on polystyrene (PS) fibers that were electro-centrifugal spun using 18 wt% concentration of PS in tetrahydrofuran (THF) and concluded that highly aligned straight fibers were fabricated (97.7% of fibers within $\theta=5^\circ$), at the collecting distance of 2.5 cm. They reported that the obtained amount of alignment is considerably higher than those produced by other centrifugal spinning [58,69,87] and electro-centrifugal spinning [40,44,45] methods. In another effort, Khamforoush *et al.* [46] reported electro-centrifugal spinning of 17% PAN at 10-22 kV applied voltage, 1000-2000 rpm rotating speed and 14 cm distance.

B. Effect of Rotational Velocity

Rotational speed is an effective parameter in both centrifugal and electro-centrifugal spinning methods, which affects centrifugal forces as well as solution flow rate. In electro-centrifugal spinning, due to applying the electronic force which helps clearly the centrifugal force to overcome the surface tension, lower rotational speed is required. Generally, at a constant flow rate, a decrease in the diameter of the produced fibers occurred upon increasing the rotational speed due to raising centrifugal

forces [39,40]. However, higher rotational speed could also increase the fiber diameter as a result of an increase in the impact intensity of the air surrounding the nozzle with the exiting jet, which leads quick drying of the jet. As the velocity of the air flow increases, the polymer jet gets dry before it can be elongated further by the centrifugal force. Therefore, it seems that at every flow rate, there is a rotational speed in which the minimum fiber diameter can be spun. On the other hands, increasing rotational speed leads to higher flow rate which induced thicker jet and in consequence, thicker produced fibers. Moreover, by increasing the frictional forces between the polymer chains, the viscosity of the solution rises. As it is known, the frictional force naturally is dependent on the rate of the external forces. Therefore, with an increase in speed of applied forces, the frictional forces between the polymer chains increase. Regards, the solutions with lower concentrations can be spun by increasing the rate of the applied external forces.

In an effort, 15 wt% PAN solutions were electro-centrifuged at a constant flow rate of 1.2140 mL/h [88]. The results showed that the fibers diameter reduces first by increasing the rotational speed and then increases again. The authors revealed that the observation was a result of the increase in the jet velocity and impact intensity of the air with the jet [88].

Liu *et al.* [51] reported that the velocity from 300 rpm to 600 rpm is large enough to get over the surface tension and the faster or slower speed have not any significant effect on the degree of fibers alignment. They produced PVP fibers under 6.2 kV applied voltage, 3 cm collecting distance and three rotational velocities of 360, 420, and 540 rpm. They reported that 420 rpm is most sufficient rotating speed for the applied voltage of 6.2 kV, and at these conditions the polymer fibers fabricated are relatively parallel. Slower rotational velocity (360 rpm) cannot provide enough horizontal velocity to pull the jet fully straight.

C. Effect of Applied Voltage

Applied voltage is a determinant of fiber morphology (e.g. fiber alignment), and with less spars it determines the diameter of fibers [46]. In general, by increasing the voltage, fiber diameter may increase, decrease, or even remain unchanged [46]. Actually, the influence of applied voltage on fiber diameter is not very clear. Hosseinian *et al.* [89] investigated the effect of applied voltage (0-14 kV) as well as rotational speed (197-4051 r/min) on morphology of PVP nanofibers. They reported that lower applied voltage and rotating speed resulted in producing micro-particles instead of fibers, while increase in the applied voltage resulted in a decrease in the minimum rotational

speed that is required to form continuous fluid jet.

By increasing the voltage, electrostatic force on the polymer jet increases and it may reduce the fiber diameter. Otherwise, due to increasing flow rate as the result of increasing the applied voltage, thicker fibers will produce. In addition to the influence of voltage on the fiber diameter, it can lead to the production of a mixture of beads and fibers [90,91]. Dabirian *et al.* [88] investigated the average fiber diameter at different applied voltages. They fabricated PAN nanofibers using electro-centrifugal spinning of 15% PAN at five voltages of 5, 10, 15, 20, and 25 kV. The results did not show any specific trend of voltage on diameter. In another study, the authors concluded that the average fiber diameter of 16.5% electro-centrifugal-spun PAN was 763 ± 47 and 812 ± 39 nm, respectively, at 20 and 22 kV applied voltages (Table II). It indicated that increasing the applied voltage increased the fiber diameters [46].

On the other hand, Liu *et al.* [51] studied the effect of applied voltage on the alignment degree of PVP fibers (at 4, 6, and 8 kV) and PMMA (at 4.5, 6, and 7 kV). The experiments were done at a constant rotating speed of 420 rpm, leading to producing highly aligned fibers. The results showed that at 6 kV applied voltage, the most aligned PVP fibers can be achieve compared with 4 and 8 kV. They revealed that the alignment degree of PVP and PMMA fibers raised at first and then reduced by arising the applied voltage, at the same polymer concentration. When the applied voltage is low (4 kV), PVP fibers show columnar structure because of small electrical force that cannot provide enough elongation for the diameter of the coiled jets. Conversely, jets are over-elongated when the applied voltage is raised. In consequence, finer fibers through a new stability are formed. The authors also studied the relationship between applied voltage and rotating velocity. Both parameters affect the morphology of as-spun fibers [92]. More precisely, the rotational velocity influences the process where jets solidify into fibers [87]. At the beginning stage of the process, both electrical and centrifugal forces control the straight part of the jet. The moving of the jet in this segment through vertical direction can be defined as a relationship between the applied voltage and the speed of the jet within the electrostatic field [51]. Regards, Hashemi *et al.* [93] reported the numerical investigation of an initial stable jet through the air sealed electro-centrifugal spinning. They numerically found that by increasing the rotating speed, the centrifugal force dominates on electric force. In consequence, changing the applied voltage does not significantly affect stretching or ejecting the forming jet. With an increase in the rotational speed, centrifugal force (rotational velocity) increases directly and the effect of electric force suppresses in overcoming the surface

tension. This fact leads to the decrease in required applied voltage. However, in condition of centrifugal spinning, it is difficult to obtain a uniform morphology of highly alignment microfibers [87]. The authors concluded that when both vertical and horizontal velocities are very close, more aligned spun fibers will be produced [51].

D. Effect of Polymer Concentration

In all fiber spinning techniques, the concentration of polymer plays an important role in determining fiber morphology and diameter due to its effect on solution viscosity. For instance, in electrospinning technique, at lower concentrations, the surface tension is an influential parameter on the morphology of fibers. It is also known that below a certain concentration, drops or beads will form instead of fibers. While at high concentrations, electrospun-ability of the solution will decrease due to difficulty of passing the solution through the nozzle [94]. In electro-centrifugal spinning, an increase in polymer concentration leads to reduce the bending instability of the jet. So, the jet travels in a straight path toward the collector and highly aligned nanofibers are deposited on the cylinder [45]. When using electro-centrifuge spinning, the nanofiber diameters change compared to centrifuge spinning. In an effort, different electro-centrifugal-spun fibers were produced from PAN solution with the concentration of 10 wt% to 16 wt% at voltage of 15 kV and rotational speed of 6360 rpm [88]. Their results indicated the linear increase of nanofibers diameter by concentration addition [95].

Electro-centrifugal spinning of PMMA fibers from various concentrations of 8 to 15 wt% were also studied by Liu *et al.* [51]. With increasing in polymer concentration to 13 wt%, PMMA fibers with sufficient degree of alignment as well as uniform morphology were obtained. They revealed that by increasing the polymer concentration and fighting the jet through an approximate straight path toward the cylinder, the degree of fiber alignment significantly increased [45]. This process continued until the polymer concentration increased to 15 wt%. The authors also worked on PVP and PS nanofibers from different solution concentrations and similar results were obtained. Briefly, both as spun fibers produced from 8 wt% PMMA/THF and 8 wt% PVP/ethanol solutions showed beaded morphology [51]. By increasing the concentration to 15%, the alignment and uniformity increased, but still micro-beads were observed on the fibers. When the concentration of PS solution increased to 18 wt%, micro-beads disappeared and only uniform and smooth fibers were formed [51].

V. CONCLUSION

In this study, three nanofiber fabrication procedures

-electrospinning, centrifugal spinning and electro-centrifugal spinning- and the effect of effective parameters on the nanofibers morphology were reviewed. While the electrical force is the basic agent in electrospinning, centrifugal force created due to rotation of spinneret is the main agent of spinning in centrifugal spinning. Both electrical and centrifugal forces are applied in electro-centrifugal spinning. There are some identical determinants of fiber morphology in all three methods. In both centrifugal and electro-centrifugal spinning, rotational speed directly affects the relative velocity between jet and surrounding air. Therefore, by increasing the rotational speed, produced jets tend to dry faster. This fact suppresses the grow of wave instability through electro-centrifugal spinning in comparison with electrospinning and leads jets to travel straighter to form more aligned nanofibers on the collector. Although, the distance of nozzle to collector is a critical operating parameter to determining nanofibers diameter in electrospinning method, it only shows an effect on fiber morphology and has no significant effect on the fiber diameter in centrifugal spinning. Surface tension and viscosity of the solutions are the spinning-ability determinatives in these three methods which are affected by the type of polymers and solvents and also the concentration of the solution, and need to be overcome through electrical or centrifugal forces or both. While in many cases, the applied voltage shows impressive effects on resulted fibers diameter in electrospinning, the influence of applied voltage on fiber diameter is not very clear in electro-centrifugal spinning. Although both centrifugal and electro-centrifugal spinning are very promising methods for fabrication of aligned fibers, electro-centrifugal spinning is a more flexible procedure to produce wide range of nanofibers based on the type and morphology.

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