Recent progress concerning the production of controlled highly oriented electrospun nanofibrous arrays

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Abstract. Among the foreground domains of all the research-development programs at national and international level, a special place is occupied by that concerning the nanosciences, nanotechnologies, new materials and technologies. Electrospinning found a well-deserved place in this space, offering the preparation of nanomaterials with distinctive properties and applications in medicine, environment, photonic sensors, filters, etc. These multiple applications are generated by the fact that the electrospinning technology makes available the production of nanofibers with controllable characteristics (length, porosity, density, and mechanical characteristics), complexity and architecture. The apparition of 3D printing technology favors the production of complex nanofibrous structures, controlled assembly, self-assembly of electrospun nanofibers for the production of scaffolds used in various medical applications. The architecture of fibrous deposits has a special influence on the subsequent development of the cells of the reconstructed organism. The present work proposes to study of recent progress concerning the production of controlled highly oriented electrospun nanofibrous arrays and progress in research on the production of complex 2D and 3D structures.

1. Introduction

The 21st century has begun with the promise of nanotechnology, which is expected to harness novel properties of materials and unique features of phenomena at the nanometer scale. Over the last years, research and development in the field of nanomaterials, which are materials with structural units on a nanometer scale in at least one direction, has enjoyed unprecedented support, nanomaterials becoming the fastest growing area in materials science and engineering [1-6]. Large surface, porosity and controlled deposition of electrospun nanofibers create favorable premises for electrospinning technology utilization [7-12]. Nanofibers deposition on the collector surface and their architecture can differ from 1D to 3D [13-16]. The manner of their disposal can be random geometric disposal or 3D orientation, uniaxial or biaxial, multistrat disposal cross deposits, in double or triple layers resulted from the utilization of beam bars or by depositing layer by layer [17-23]; and differential deposits. Specialist’s preoccupation to obtain uniaxial aligned nanofibers is related with the fact that this distribution is absolutely necessary for certain applications. For example, the electrospun nanofibers are ideal mediums to realize neural fabrics. This example proves the importance of the distribution and architecture of electrospun deposits for their implementation in various applications [24-26].

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2. The present stage of the development of knowledge concerning the production of aligned electrospun fibers

There are several technological and constructive factors that act on nanofibers manner of deposition in electrospinning process:

- **Concentration of polymeric solution**- adequate solvent selection is a problem of first importance in workability and solubility of various polymeric solutions; a higher polymer concentration favors the generation of uniaxially oriented deposits [27-32];

- **Collector speed** (in the case of drum-type rotating collector mechanisms) [33-37] significantly influences the orientation of electrospun nanofibers. Yet, there is an optimum value of the rotating drum speed up to which one can obtain fibrous deposits arrays with dimensional characteristics which allows their utilization;

- **Electric field voltage** significantly influences the manner in which the electrospun nanofibers are disposed; high values of electric voltage (kV) determine the increase of orientation degree [38-42];

- **Distance between electrodes** is a constructive parameter with direct influence on the manner in which the electrospun nanofibers are distributed; a small distance between electrodes ensures an increased control on depositing manner [43-48];

- **Constructive type of collecting mechanism** and modular attachment of devices to facilitate a certain arrangement [49-51] seems to be modalities mainly approached in specialized literature what concerns the realization of ordered arrays. It is well known that one can not obtain perfectly ordered arrays due to the chaotic motion of polymeric jet. In order to obtain ordered fibrous arrays, additional elements, such as additional electrodes of various constructive types, wires, metallic rings or bars have been developed, all these facilitating electric field guiding and aligned distribution of electrospun fibers.

1. **Collecting mechanism type rotary drum** provides, as compared to the stationary plane or stationary rotary collectors, the increase of the evaporation time and implicitly the increase of ordering degree;

2. **Collecting mechanism type rotary drum with bars/bar rotary drum** developed by Xu figure 2, in spite of permitting to obtain ordered arrays and easy servicing [52], has several shortcomings, such as: limitation of nanofibrous deposit thickness; impossibility to align the fibers at any moment of electrospinning. A particular case of this constructive type is that of rotary collecting mechanism with copper wires wound on the drum (wire rotary drum), promoted by Bhattarai, (2005). This mechanism offers the advantage of modifying the depositing area of aligned fibers by modifying the thickness of the copper wires disposed on the surface of rotary cylinder. A major disadvantage of this constructive type is that the electrospun fibers resulted from polymeric jet are mainly gathering on copper wire direction, instead of being deposited on the entire surface of the collector drum figure 2.

3. **Collector mechanism with rotary drum and lamellar electrode**
Brown and Stevens, (2007) as well as Teo, Ramakrishna, (2006) [53] developed another principle to obtain perfectly ordered nanofibrous deposits, a principle according to which under the collector mechanism itself made of a rotary drum of a very small diameter, there is a knife blade-type electrode loaded at a negative potential figure 3.

The advantage of this constructive type would be the possibility to obtain aligned fibers on the entire surface of the collecting cylinder. The constructive disadvantages of this type of collector consist in limitations imposed by drum diameter, complexity of programming the constructive and technological parameters, as well as the technological impossibility to obtain thin layers;
Figure 1. Electrospinning equipment with collector type drum with bars: (1. syringe; 2. polymeric solution; Taylor cone; 4. polymeric jet; 5. drum with bars; 6. copper bars; 7. fibrous deposit) [54].

Figure 2. Rotary collector mechanism with wound copper wires (Source: Bhattarai, 2005).

Figure 3. Collector mechanism with rotary drum and lamellar electrode (1. syringe; 2. needle; 3. Taylor cone; 5. delivered jet; 6. rotary drum; 6. sharp blades [Source: Brown and Stevens, 2007].

4. Disk-type collecting mechanism was developed by Subramanian, (2005), Czaplewskia, (2003), Theron, (2001) [53-55]. The principle of obtaining ordered fibrous arrays by using collecting mechanism with rotary disk is based on the modification of the profile of electrostatic field between electrodes, correlated with the disk rotation motion, figure 4. The main advantages would be: easy construction, installation and servicing; getting highly aligned fibrous arrays, easy detachment of fibrous deposits from the collector. The disadvantages of these collecting mechanisms with disk are marked by the following aspects: fiber breakages occurred at high drum rotation speed; impossibility to maintain fibers aligned in the case when obtaining large diameter fibers; small area of fibers array.

5. Lamellar collecting mechanism. The principle of obtaining ordered arrays by using lamellar collecting mechanisms was developed by Deitzel, (2001) [56], figure 5 and Teo, (2005), [53] figure 6; this consists in collecting the fibers within interlamellar space. The main shortcoming of this set-up is that it does not permit to obtain long fibers.
6. The needle type collecting mechanism was studied by Sundaray [9];

7. Collecting mechanism type collecting frame Huang and Dersch (2003) have developed frame-type collecting mechanisms (wood or aluminum) to obtain highly oriented nanofibers [9] figure 7;

8. Collecting mechanisms with parallel electrodes were developed by Li (2003), [57, 58] and Yang [2008] [59]; in this case, figure 8, the principle used to obtain ordered fibrous arrays is based on the idea that the manner the jet is dissipated is influenced by the profile of the formed electrostatic field, such that the fibers will be arranged transversal along the space between electrodes. The collecting mechanisms with parallel electrodes have the advantage that provides an easy fibers transfer on another substrates. The main disadvantages of this constructive type of collecting mechanism which ensures the production of aligned fibrous arrays would be: limitations of fibrous layer thickness; limitations of fibers length, this being given by the distance between electrodes.

9. The static collecting mechanism with auxiliary electrodes have been developed by Bormat (2005), who added an auxiliary electrode from parallel steel strips [9], meant to concentrate the electric field along them, such that the polymeric jet has the tendency to align along the strips figure 9.

![Figure 7. Principle of obtaining oriented fibrous arrays by using frame-type collecting mechanisms (1. syringe; 2. polymeric solution; 3. wooden frame driven in rotation motion; 4. aligned fibers; 5. source; 6. polymeric jet) [27].](image1)

![Figure 8. Mechanism with parallel electrodes (1. syringe; 2. polymeric solution; 3. needle; 4. polymeric jet; 5. parallel electrodes; 6. source) [58].](image2)

![Figure 9. Static collecting mechanism with auxiliary electrodes (1. syringe; 2. polymeric solution; 3. needle; 4. polymeric jet; 5. mandrel with rotation motion; 6. auxiliary electrodes) [27].](image3)

Based on the same principle, Teo (2005) [27] used instead of steel bars, parallel bars with sharp edges. Gibson and Schreuder- Gibson (2004), [9, 27] executed electrospinning directly on a metallic mesh, the fibers being arranged parallel with the metallic mesh axis figure 10.

10. Introduction of auxiliary metallic rings between the needle and the collector also permitted to obtain ordered arrays by generating a more uniform electric field, with influences on improving the jet stability. In this connection, Jaeger (1998), Detzel (2001) figure 11, Stakus (2004) figure 12, Dalton (2005), Buttafoco (2006) [9, 10] developed various constructive types of rings which permit to obtain aligned fibrous arrays [60-65]. These constructive elements added to obtain ordered distributions, permit to obtain fibrous arrays with controlled distribution areas, depending on ring diameter. The main disadvantage of these systems consists in the limitation given by the fact that the ring/rings must be positively charged. There are also collecting mechanisms with rings arranged in parallel, with a
view to obtain ordered distributions (Dalton, 2005); these mechanisms permit to obtain twisted fibers. The main disadvantages of this constructive type are: getting fibers with limited lengths; necessity that one of constitutive rings twists the fibers subsequently deposited in the yarn structure figure 13.

Figure 10. Collecting mechanism type metallic mesh (Source: Gibson and Schreuder-Gibson, 2004).

Figure 11. Collecting mechanism with three copper rings (polymeric solution; 2. syringe; 3. needle; 4. Taylor cone; 5. copper rings; 6. plane collector) [56].

Figure 12. Collecting mechanism with one copper ring (1. syringe 2. needle positively charged 3. source 4. ring positively charged 5. plane collector) (Source: Stakus, 2004).

Figure 13. Collecting mechanism with parallely placed rings [60].

3. The present stage of the development of knowledge concerning the production of organized assemblies of 2D and 3D highly oriented electrospun nanofibers.

The present researches are focused on the production of complex structures 2D, 3D, controlled assemblies, self-assemblies of electrospun nanofibers to obtain the scaffolds used in bone tissue engineering, drug delivery systems, neural tissue engineering, reconstruction construction of heart, skin, tendons, and cartilages tissues. Each of these complex structures implies a certain structure, density, porosity, architecture etc. The manner of nanofibres self-assembly and deposition are the result of controlled technological processes, resulting in structures with pre-established characteristics and shapes, such that to permit the future development of the cells of the organism to be reconstructed. Ordered fibrous structures, rightfully aligned, favor the controlled development of the cells in the reconstructed organism, with the idea to obtain anisotropic structures similar to the human ones.

4. Conclusions

Electrospinning is a nanotechnology leader that ensures the production of controlled highly oriented electrospun nanofibrous arrays. Through an optimal correlation of technological parameters and constructive with characteristics electrospinning polymer solutions can be obtained complex nanofibrous structures, controlled assembly, self-assembly of electrospun nanofibers for the production of scaffolds used in various medical applications.
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