

# Electrospinning predictions using artificial neural networks



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## ABSTRACT

Electrospinning is a relatively simple method of producing nanofibres. Currently there is no method to predict the characteristics of electrospun fibres produced from a wide range of polymer/solvent combinations and concentrations without first measuring a number of solution properties. This paper shows how artificial neural networks can be trained to make electrospinning predictions using only commonly available prior knowledge of the polymer and solvent. Firstly, a probabilistic neural network was trained to predict the classification of three possibilities: no fibres (electrospraying); beaded fibres; and smooth fibres with >80% correct predictions. Secondly, a generalised neural network was trained to predict fibre diameter with an average absolute percentage error of 22.3% for the validation data. These predictive tools can be used to reduce the parameter space before scoping exercises.

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## 1. Introduction

Electrospinning is an increasingly important area of research as developments in biotechnology, electronics, catalysts, composites and sensors all call for increased flexibility and control over the manufacture of nano-scale fibres [1,2]. Specific applications include tissue scaffolds [3–11], membranes for fuel cells [12], separation layers for batteries [13], air and water filtration [14,15], reinforcement for nano-composites [2,16–18], piezoelectric fibres for energy harvesting and sensors [19–21] and conductive layers in solar cells and electronics [17,21,22]. Electrospinning compares favourably with other methods of nano-fibre manufacture such as drawing, template synthesis, phase separation and self-assembly because of its relative simplicity and low cost [23–26].

Needle or capillary electrospinning of polymer solutions typically works by drawing a small electrostatically charged jet from a droplet to a grounded collector (Fig. 1). The jet is initiated from the droplet when the repelling forces of the surface charges overcome the surface tension and viscous forces of the droplet. The jet diameter reduces as it is stretched and the solvent evaporates. At a certain point, known as the whipping instability, the electrostatic

repulsion overcomes the viscoelastic forces and the jet flight path changes to an expanding helix, causing further stretching and drying of the fibre. In most cases this leads to dry non-woven nanofibres depositing on the collector [27].

Many groups have studied the effects of electrospinning parameters on fibre characteristics with the aim of optimising fibres for specific applications. Koski and colleagues studied the effect of molecular weight and polymer concentration for poly(vinyl alcohol) (PVOH) and water systems. Electrospinning was found to occur if the product of the intrinsic viscosity and polymer concentration was over a certain threshold [28]. This supports the theory that the degree of polymer chain entanglement dictates whether electrospinning or electrospraying dominates. Son and co-workers studied the effect of various solvents and polyelectrolytes on poly(ethylene oxide) (PEO) fibre diameter and morphology. A nonlinear inverse relationship was found between the dielectric constant of the solvent and the fibre diameter [29]. A number of studies have all carried out parameter measurements for various polymer/solvent systems [30–33]. While it is difficult to compare findings directly, certain trends seem to be universal i.e. the qualitative relationships between polymer concentration and resulting fibre diameter.

Building on from similar parameter studies, empirical models have been created with good predictive capabilities, albeit for specific polymer/solvent and equipment combinations. Two studies used response surface methodology (RSM) to predict poly(acrylonitrile) (PAN) fibre diameters based on chosen

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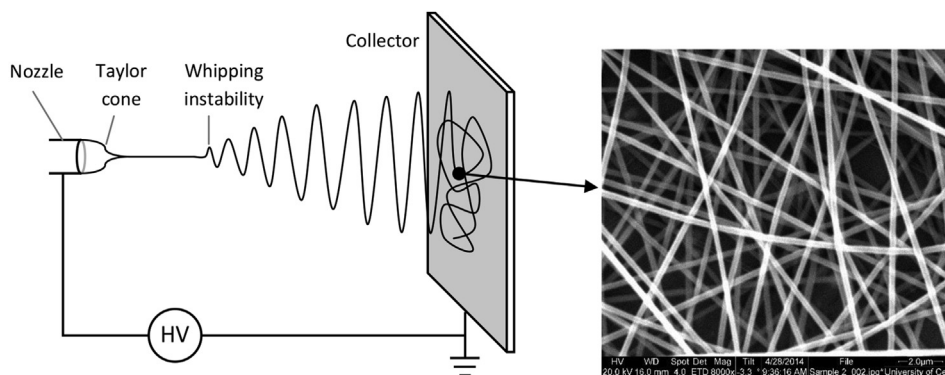


Fig. 1. Schematic of a typical electrospinning setup and an SEM of electrospun nanofibres.

parameters. They found polymer concentration to have a significantly larger effect than voltage and distance [34,35]. These studies aim to optimise polymer/solvent systems that were initially found using trial and error. These trial and error scoping exercises are time intensive, costly, and ideally could be replaced with generalised predictive empirical or analytical models. Currently there is no way to predict the characteristics of new electrospun materials based on prior knowledge of the polymer and solvent alone.

Accurate analytical modelling of fibre formation is complicated because of the nonlinear relationships in electrohydrodynamics with solvent loss. In addition, many material properties such as the intrinsic viscosity and bulk/surface charge density need to be determined experimentally before being incorporated into the models [32,36–41]. Notwithstanding these difficulties, important models have been validated that describe different portions of the jet for selected materials. Taylor introduced the “Leaky Dielectric Model” in the 1960s which describes how the surface charges of the droplet are balanced with the tangential viscous forces of the liquid, resulting in what is known as the Taylor cone [42]. In the 1970s Saville studied the stability of fluid cylinders in a longitudinal electric field [43]. Reneker and colleagues created a mathematical model to explain the whipping instability based on the rheological properties of the polymer solution and lateral perturbations [32,36,41,44]. Hohman and co-workers developed mathematical models for three different instabilities and used the models to create operating diagrams for when electrospinning occurs [37–39]. Feng created a 1D model based on slender body theory to examine the role of nonlinear rheology on the electrically stretched jet [40].

Despite the aforementioned empirical and analytical models, there has been little work into predictive methods that are generalised and do not require *a priori* measurements of the solution properties. Helgeson and Wagner developed a generalised predictive model for fibre diameters using dimensional analysis and measured parameters such as solution conductivity and dry fibre diameter. The predictions worked well for electrospinning parameters resulting in smooth fibres but deviated slightly for parameters where beading is known to occur [45].

Sarkar and co-workers trained artificial neural networks (ANNs) to predict fibre diameters for PEO and poly(methyl methacrylate) (PMMA) respectively [46,47]. Sarkar and colleagues were able to predict fibre diameters over several datasets taken from literature; however, these data contained *a priori* solution measurements. Nasouri and colleagues used a response surface methodology (RSM) and artificial neural networks (ANNs) to predict the highest production rate for PAN nanofibres within 4% of the measured value [48]. In all cases where RSM and ANNs were compared, the

authors noted ANNs were better able to predict electrospinning characteristics [47,48].

In this study ANNs were used to create generalised empirical models for the prediction of electrospun fibre diameters and morphology based on literature data. Specifically this included function approximation for fibre diameter prediction and classification to predict which category (fibre, beaded fibre, or no fibre) would prevail given a specific polymer/solvent combination. The primary aim was to determine whether an ANN trained only with data containing *a priori* information can compete with ANNs trained with data containing solution measurements for diameter and morphology prediction.

## 2. Process parameters

Studies of the electrospinning process have shown that to understand the phenomenon a large number of material and experimental parameters are required. Table 1 shows a list of parameters that affect the electrospinning process, including a qualitative measure of their influence on the fibre diameter and whether that parameter is known *a priori*, is an independent variable, or needs to be measured. Where conflict exists between findings in the literature, preference was given to experimental findings over theoretical models.

From the number of parameters shown in Table 1, which is not exhaustive, it is clear why analytical modelling of the electrospinning process is difficult. In contrast with this is the relative ease in which electrospun fibres can be produced in the laboratory for a wide range of polymer/solvent systems. Insight into this contradiction can be gained by focussing only on the few parameters that have the strongest influences, and by knowledge of the interconnected nature of the parameters.

The following sections provide rationale for which parameters could be included in the training data. An attempt is made to suggest which ‘hard to measure’ parameters could be replaced with parameters that are known *a priori* or are independently variable.

The method proposed in this paper is to find empirical models using statistical techniques. Quantitative relationships between the solution properties and the polymer or solvent properties will not be known in advance as it is hypothesised that the strongest interdependencies will be encapsulated in the trained ANNs.

### 2.1. Polymer properties

Arguably, the most important polymer parameter affecting electrospinning is the molecular weight. The length and size of the molecules directly influence the intrinsic viscosity of the polymer

**Table 1**  
Electrospinning parameters including their influence of fibre diameter and morphology.

Parameter group	Parameter	Impact inferred from literature	Easily known or controlled
Polymer	Molecular weight	Strong [28,30,31,49].	✓
	Polydispersity	Moderate [50–52]	×
	Intrinsic viscosity	Strong [45,51]	×
	Density	Weak [32]	✓
	Glass transition temperature	Unknown	✓
	Dielectric constant	Unknown	–
	Solubility parameters	Strong [53]	×
Solvent	Density	Weak [32]	✓
	Zero shear viscosity	Moderate [45]	✓
	Vapour pressure	Moderate [32,54]	✓
	Dielectric constant	Strong [29,53]	✓
	Conductivity	Strong [30,33,46,55]	✓
	Dipole moment	Strong [54]	✓
	Surface tension	Weak [30,32,44]	✓
Solution	Concentration	Strong [28,30,33,34,49,51]	✓
	Relaxation time	Strong [32]	×
	Viscosity	Strong [30,32,40,41]	×
	Vapour pressure	Moderate [32]	×
	Conductivity	Strong [30,33,46,55]	×
	Surface tension	Weak [30,32,44]	×
	Charge density	Strong [30,32,38]	×
	Feed rate	Weak [39,45,49,56]	✓
	Nozzle geometry	Weak [56–58]	✓
Experimental	Distance	Weak [30,33,49,56]	✓
	Voltage	Moderate [34,35,56,59]	✓
	Atmospheric humidity	Weak [32,50,60]	×
	Atmospheric temperature	Weak [60]	×
	Collector geometry	Moderate [56,59]	✓

in solution, as described by the Mark–Houwink equation [28,51]. The higher the molar mass, the lower the critical concentration needs to be before electrospinning can occur. The molecular weight has also been shown to have power law relationships with fibre diameter. Because the molecular weight is largely known for a given polymer and intrinsic viscosity is not, it is suggested that molecular weight may be used to supplant viscosity measurements in an empirical predictive model.

The influence of polydispersity (or molecular weight distribution) on viscosity (and hence fibre diameter) is moderate [50–52]. As this information is often not known, it was not directly included in this work and provided a known source of error.

In an attempt to provide the ANN with information about the polymer chain structure, the inclusion of glass transition temperature data was assessed. The glass transition temperature for a given polymer with a constant molecular weight is known to vary with the degree of branching [61]. The glass transition temperature combined with the molecular weight and the polymer concentration may help to improve the predicted degree of polymer chain entanglement and thus solution viscosity.

To the authors' knowledge, the influence of the polymer's dielectric constant in relation to electrospinning has not been studied. The dielectric constant is a measure of the polymer's ability to polarise in the presence of an electric field. It is therefore also an indirect measure of the polymer chain's polarity. The polarity of a polymer strongly influences whether it is soluble in a solvent, following the well-known rule of thumb 'like dissolves like'. The effect of solubility on the critical entanglement and fibre diameter is related to the hydrodynamic volume of the solute, which is larger in good solvents [51]. Therefore the solubility of any polymer/solvent system should be incorporated (directly or indirectly) into any

predictive model. Polymer and solvent solubility parameters, such as Hildebrand or Hansen solubility parameters, could also be useful, but were not used in this study.

## 2.2. Solvent properties

The zero shear viscosity is known for most common solvents and together with the intrinsic viscosity of the polymer and the polymer concentration, contributes to the solution viscosity.

Solvent evaporation has an important influence on fibre morphology and is related to parameters such as vapour pressure and boiling point.

The conductivity of a solution depends on complicated interactions between the solute and the solvent. The presence of small quantities of impurities or electrolytes can have a significant effect on conductivity. However, as this parameter needs to be measured, the solvent's conductivity was used to provide a predictor of solution conductivity.

The surface tension of the solvent is considered a critical parameter in many theoretical models; however, experimentally it has been found to have a weak influence on fibre diameter [30,32], while it may have a large impact on fibre morphology [44]. For this reason the ANN was trained with solvent surface tension values to verify their impact.

## 2.3. Solution properties

The most important solution property is concentration, as together with molecular weight it determines whether there is a critical degree of molecular entanglement for electrospinning to occur [28,30,33,35,51]. The concentration also follows power law relationships with fibre diameter and can easily be varied before electrospinning.

It is hypothesised that many of the other solution properties such as surface tension, vapour pressure, dielectric constant, charge density, conductivity and viscosity can be derived (at least in principle) from the polymer and solvent parameters.

## 2.4. Experimental parameters

The electrospinning voltage has a moderate effect on fibre diameter as it is responsible for the drawing of the jet from the Taylor cone. In literature this is often reported as nominal electric field strength in kV/cm, the distance referring to the gap between the nozzle tip and collector [39,40,46].

There are conflicting reports on the significance of flow rate on the fibre diameter in the literature. Some report the flow rate is a parameter that can be used to control the final fibre diameter, whilst others report it has no statistical significance [30,39,46,49,62]. In the case of stable electrospinning where the meniscus at the spinning tip maintains constant shape, the flow rate into the Taylor cone must balance the rate of drawing from the jet. Over-feeding causes the meniscus to swell and fall off the nozzle tip, while lower flow rates result in starvation and stuttering of the jet. Unstable flow is likely to produce a wider variation in fibre diameters; however, there is little evidence to suggest a significant effect on the average fibre diameter [49,63]. In this study the effect of flow rate was assumed to be weak and thus not essential for the predictive model. Experiments have shown the nozzle diameter to have a statistically weak effect on fibre diameter and so this was not included in this study [56–58].

Typically single needle electrospinning is carried out at room temperature at atmospheric pressure and humidity. Whilst atmospheric conditions are known to contribute to fibre diameter and morphology, unless these conditions vary significantly from

standard atmospheric conditions, the effects are minimal [32,50,60]. Additionally, these parameters are often not quoted in the literature and are difficult to control with most laboratory setups. For these reasons, standard laboratory atmospheric conditions were assumed in this study.

### 2.5. Selected parameters

Table 2 summarises which parameters received focused attention for the development of the predictive models. All effort was made to focus on parameters known to have strong or moderate impacts on electrospinning characteristics, known in advance or that could be independently varied. Some parameters thought to have weak impacts were included for confirmation.

## 3. Experimental data

The data used to create the predictive models were gathered from the literature, as shown in Tables 3 and 4. Only data relating to single needle/capillary electrospinning of primary solvent systems were used. Differences in needle geometry, flow rate, collector geometry and atmospheric conditions were not taken into account, and are known sources of error. Effort was made to avoid including tests that involved the effects of electrolytes to change solution conductivity. Common solvent properties were taken from the literature [65].

Numerical data for fibre diameters were taken directly from the references. Where ranges or histograms were given, mid points or distribution peaks were used to approximate averages. Some of the data included fibres with a variety of morphologies, including beads and ribbons; however, the majority were reported to be smooth fibres. Because of the wide variety of measurement methods [72], and the relatively high standard deviations and skewed distributions typical in electrospun fibre mats [63], the errors associated with the measured average diameter data were expected to be high.

A recent study by Tucker and colleagues [72] investigated different fibre diameter measurement methods and may help to put the input errors into perspective. The study found that when six different people took measurements of the same 10 images their reported averages varied by up to 35 nm, which in this case is an average % error of 12.4%. This variation occurred even though the measurements were carried out using the same operating practice

**Table 2**

Parameters selected for trial in the predictive models.

Parameter group	Parameter	Impact inferred from literature	Easily known or controlled
Polymer	Molecular weight, ( $M_w$ )	Strong [28,30,31,49]	✓
	Glass transition temperature, ( $^{\circ}\text{C}$ )	Unknown	✓
	Dielectric constant at 1 MHz	Unknown	–
Solvent	Density, (g/mL)	Weak [32]	✓
	Zero shear viscosity, (cps)	Moderate [45]	✓
	Vapour pressure, (Torr)	Moderate [32,54]	✓
	Dielectric constant	Strong [29,53]	✓
	Dipole moment, (D)	Strong [54]	✓
	Boiling point, ( $^{\circ}\text{C}$ )	Unknown	✓
	Conductivity, (S/cm)	Strong [30,33,46,55]	✓
	Surface tension, (mN/m)	Weak [30,32,44]	✓
Solution	Concentration, ( $\%w/v$ )	Strong [28,30,33,34,49,51]	✓
	Electric field strength, (kV/cm)	Moderate [39,40,43,46,64]	✓

**Table 3**

Data from the literature used to train and test the artificial neural network diameter prediction models.

Polymer	Solvent	Fibre diameter (nm)	Data points	Reference
PS	DMF	292–4284	21	[55]
PVOH	Water	192–731	31	[33,66]
PEO	Water	80–540	18	[46]
PLA	DCM	200–1600	16	[67]
PVB	Acetone	86–1560	6	[31]
	Ethanol	130–1000	6	[68]
	Isopropanol	130–350	5	[68]
PA 66	n-Butanol	110–130	3	[68]
	Formic acid	117–198	38	[69]
PAN	DMF	83–2782	51	[34]
PMMA	DMF	143–1505	18	[47]
	Ethyl acetate	3840–3890	2	[70]
PVDF	DMAA	400	1	[71]

and an average of 171 measurements per person. The likelihood of the data, obtained from literature for this paper having significantly less measurement error, is very low.

Three classes (fibres, beaded fibres and no fibres) were used to classify the data in Table 4. As it is impossible to define exact boundaries between what constitutes a beaded fibre and what constitutes a smooth fibre, there is inherently some error to these data. This is because of the difficulties of examining a truly representative quantity of fibre with the limited field of vision available using a microscope.

The data taken from these sources were also biased, as researchers are more likely to present conditions leading to ‘fibres’ rather than ‘no fibres’. To balance the bias, extra data points were generated for ‘no fibre’ conditions. This was easily done by combining non-compatible polymer/solvent combinations or by reducing the polymer concentration to zero.

The parameter data used to train the predictive models contained significant errors. A moderate source of error is polymer concentration. Most authors state the percentage weight to volume ( $\%w/v$ ), which is calculated to one significant figure. Solutes are usually dissolved with continuous mixing, allowing significant solvent loss, particularly if the solvent is volatile. The authors' own experiences have shown concentration routinely varies up to 0.5% during this process. It is also common to start with a high concentration master batch and dilute this to lower concentrations, compounding the error. As concentration is expected to play a significant role in the determination of fibre diameter, the effects of these errors will be particularly high. The dielectric constant data also contained significant errors, as it was not always possible to

**Table 4**

Data from the literature used to train and test the artificial neural network classification models.

Polymer	Solvent	Data points	Reference
PS	DMF	26	[55]
PVOH	Water	34	[33,66,73]
PEO	Water	22	[46]
PLA	DMF	4	[29]
	Ethanol	4	[29]
	Chloroform	4	[29]
	DCM	18	[67]
PVB	Acetone	6	[31]
	Ethanol	6	[68]
	Isopropanol	6	[68]
PA 6	n-Butanol	6	[68]
	Formic acid	21	[49]
PA 66	Formic acid	43	[69]
PAN	DMF	51	[34,35]



find values for polymers with the same molecular weight as those used in the experiments. In these cases, interpolation was used.

### 3.1. Data subsets

Within the data outlined above, subsets were also created to compare data containing solution measurements with the same data without the solution measurements. This provided an idea of how much accuracy was lost by using only *a priori* information.

Data subsets were also created to compare individual polymers with the generalised case.

To train, test and validate the predictive models, each data subset was randomly partitioned into training, testing and validation sets, with respective ratios of 70:20:10. The training data were used to train the ANNs, while the testing data were used to select the highest performing networks. The validation data were used to assess how well the model generalised for new 'unseen' electrospinning conditions.

## 4. Predictive models

Linear regression and ANNs were used to create the predictive models. The models were trained using Palisade's NeuralTools add-in for Microsoft® Excel®. Results from the highest performing models are discussed.

### 4.1. Probabilistic neural networks (PNNs) and generalised regression neural networks (GRNNs)

PNNs are used for classification, while GRNNs are often used for function approximation and prediction [74]. All PNNs and GRNNs consist of four layers, as described below and shown in Fig. 2.

- The input layer is where the number of neurons equals the number of input parameters.
- The pattern layer contains the radial centres for all the training points; it is also where the smoothing is applied to the radial basis function to minimise the predicted error.
- The third layer is different for PNNs and GRNNs. For PNNs, it is a class layer containing one neuron for each class. For GRNNs, it consists of two neurons: one neuron is the numerator, the other is the denominator. The denominator adds up the weight values coming from each of the hidden neurons. The numerator adds up the weight values multiplied by the actual target value for each pattern neuron.

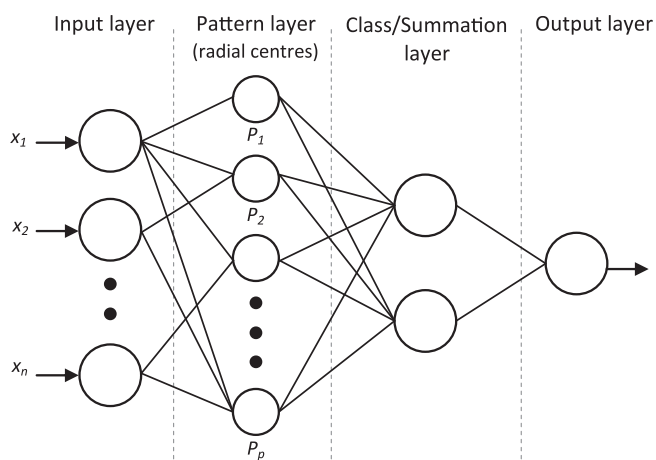


Fig. 2. Architecture for a generalised regression neural network (GRNN).

- For PNNs, the output is the probability for each class. For GRNNs, the output neuron divides the numerator by the denominator to produce the predicted value.

The PNN/GRNNs are trained by optimising the smoothing factor assigned to each training data point's radial basis function using the conjugate gradient descent method.

Unlike linear regression, ANNs can model highly nonlinear relationships. PNN/GRNNs train almost instantly and they do not require decisions about the system topology i.e. number of neurons or hidden layers. One disadvantage is that they do not extrapolate well outside the training data range. To mitigate this disadvantage, a wide range of polymers and solvents were included in the datasets.

### 4.2. Multi-layer feed-forward neural network (MLFN)

MLFN, also known as multilayer perceptrons, are also used for classification, function approximation and prediction [75]. The MLFNs used in this work contained one hidden layer, while the numbers of hidden neurons were varied to find an optimal topology (Fig. 3):

- The input layer contains a number of neurons equal to the number of input parameters. Weights are applied to the inputs as they are 'fed forward' to the hidden neurons.
- At the hidden layer, the weighted values are fed into transfer functions, which are then distributed to the output layer. Again weights are applied to the values fed to the output layer.
- Weighted values from the hidden layer arrive at the output neuron and are combined before entering another transfer function. The output of the transfer function is the output of the network.

The MLFNs were trained by optimising the weights using the conjugate gradient descent method [76]. Training was stopped when the reduction in error dropped below 1% in a 6-min period.

## 5. Results and discussion

For each data subset, statistical methods were used to find empirical predictive models. For the MFLNs, optimal topologies were found by varying the number of neurons in the hidden layer between two and six. Trials using more than six neurons showed no improvement, because of over-fitting.

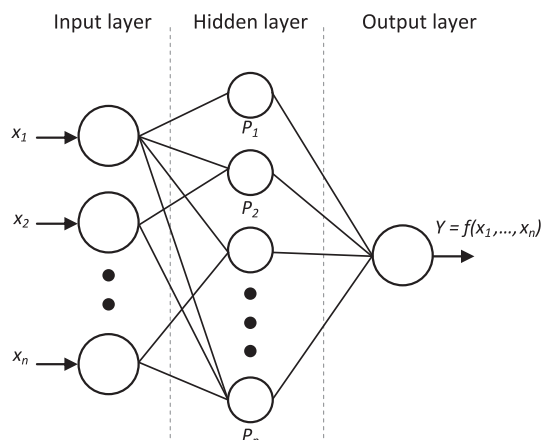


Fig. 3. Architecture of a multilayer neural network with one hidden layer.

**Table 5**  
Impact of parameters on classification from impact analysis.

Impact on classification	Parameter
Strong	Polymer concentration, Solvent density, Solvent viscosity, Polymer molecular weight
Moderate	Polymer glass transition temperature, Electric field strength, Solvent boiling point, Polymer dielectric constant
Weak	Solvent dielectric constant, Solvent vapour pressure, Solvent dipole moment

The results are split into two sections, spinning classification and fibre diameter prediction.

### 5.1. Spinning classification

The purpose of the classification model was to try to predict whether single needle electrospinning would occur given a polymer/solvent/concentration combination, and what morphology the fibre would have. The classes were categorised as ‘no fibres’ (electrospraying), ‘beaded fibres’ and ‘fibres’. PNNs and MLFNs were trained using three sets of data: one larger set containing only easily known parameters, and two smaller sets that were used to compare the performance of ANNs trained with and without solution measurements such as viscosity, surface tension and conductivity.

#### 5.1.1. Classification without solution measurements

Initially all the parameters in Table 2 were used to train 10 ANNs and a parameter impact analysis was carried out for each. Each ANN was trained with randomly selected testing data to mitigate data sensitivity on the impact analysis. The results were collated and the parameters were ranked in order of impact. The findings largely agreed with literature and are shown in Table 5.

To test the validity of the impact groupings shown above, the ANNs were retrained without the ‘weak’ parameters. The performance of the ANNs trained with and without the weak parameters is shown below. The results are from the highest performing ANNs, which were PNNs. The results (Table 6) show that removing the weak parameters slightly improved the performance of the PNN (although this was within the sensitivity range of the dataset).

A further breakdown of the classification results for the PNN without weak parameters is shown in Table 7. Most of the error came from trying to predict beaded fibres. This is likely to be because, in the parameter space, beading occurs on a continuum between electrospraying (no fibres) and smooth fibres.

The results show that it is possible to predict with over 80% accuracy whether smooth fibre electrospinning will occur, and if beading is present, using only readily available data known *a priori*. This result is significant, as it could reduce the amount of time spent doing scoping exercises for new polymer/solvent systems.

In the next section, a subset of data was used to test whether adding solution measurements such as viscosity improved the performance of the model.

**Table 6**  
Percentage of wrong predictions with and without weak parameters used in the data.

Data inputs	% of wrong predictions		
	Training	Testing	Validation
All parameters	7.4%	17.5%	23.3%
Weak parameters removed	5.2%	14.0%	19.2%

**Table 7**  
Breakdown of classification predictions.

		Predicted classes			Wrong predictions %
		Beaded	Fibres	None	
Observed classes	<i>Training data</i>				
	Beaded	35	3	2	12.5%
	Fibres	4	126	0	3.1%
	None	2	1	57	5.0%
	<i>Testing data</i>				
	Beaded	5	3	0	37.5%
	Fibres	2	26	0	7.1%
	None	1	2	18	14.3%
	<i>Validation data</i>				
	Beaded	2	1	0	33.0%
	Fibres	2	15	0	11.8%
	None	2	1	8	27.0%

**Table 8**  
Percentage of wrong predictions with and without solution measurements.

Data inputs	% of wrong predictions		
	Training	Testing	Validation
With solution measurements	0%	8.3%	7.7%
Without solution measurements	12.4%	16.7%	15.4%

#### 5.1.2. Classification with solution measurements

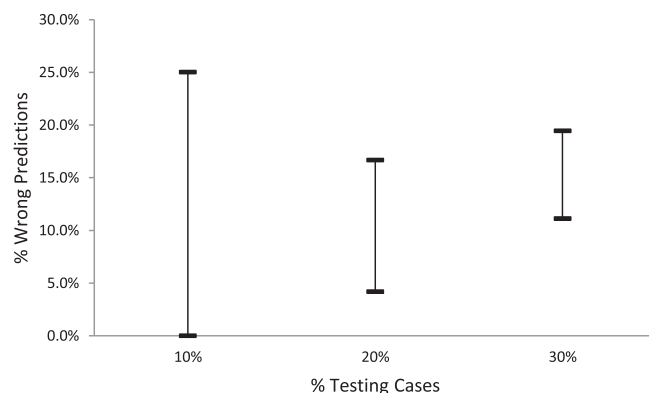
Table 8 summarises how the performance of the networks changed if solution viscosity, conductivity and surface tension measurements were included.

It can be seen that in this case there was a reduction in wrong predictions if solution measurements were used; however, this variation could easily be accounted for by the random selection of the testing data, as shown by the sensitivity analysis (Fig. 4). The sensitivity of this dataset was high, because of the low number of case points (134).

Overall we found that the performance gained by adding solution measurements was minimal and compromised the utility of a tool designed to reduce the time and cost of laboratory-based electrospinning scoping exercises.

### 5.2. Fibre diameter prediction

The next question that might be asked after “Will it spin?” is “What will the fibre diameter be?” This section presents the results of ANNs used for fibre diameter prediction.



**Fig. 4.** Sensitivity analysis showing the variation in prediction performance of artificial neural networks with solution measurements using randomly selected testing datasets and different dataset sizes.

The performance measure used here was the average absolute percentage error (average % error), as it provided a more intuitive measure than the root mean square error (RMSE). Again, the results were based on three data sets: one containing all the fibre diameter cases, and two smaller sets that were used to compare the performance of ANNs trained with and without solution measurements.

In all cases, the ANNs significantly outperformed linear regression, validating the claim that electrospinning phenomena are highly nonlinear relative to common input parameters.

### 5.2.1. Diameter predictions without solution measurements

The predictive model in this section was trained using polymer, solvent and experimental setup data with no knowledge of solution properties. Parameter impact analysis was used to determine the significance of the parameters in Table 2. The results are summarised in Table 9.

Interesting to note is the strong impact the polymer glass transition temperature and dielectric constant had on diameter prediction. The physical relationship between the dielectric constant and fibre diameter is unknown; however, it is unlikely to be related to solubility, as the solvent dielectric constant has negligible impact.

A comparison was made for networks trained with and without the weak parameters, confirming that the network could perform equally well without them. Fig. 5 shows the predicted versus measured fibre diameters for the best performing ANN (a GRNN without the weak parameters). The training, testing and validation data all showed a high degree of correlation over a wide range of diameters (note the logarithmic scale). The average % errors for the training, testing and validation sets were 19.9%, 35.0% and 22.3% respectively, which were comparable to the estimated errors of the input data.

Despite the exclusion of many parameters such as atmospheric conditions, needle diameter, flow rate, surface tension, vapour pressure, conductivity, collector geometry and more, it is striking to note that the average % error for the validation data was only 22.3% or a RMSE of 186.8 nm. This, combined with the knowledge that the diameter measurements came from over twenty different studies all using different measurement methods, shows that ANNs are a robust method for electrospinning predictions.

### 5.2.2. Diameter predictions using solution measurement data

Fig. 6 shows the predicted versus measured diameters for ANNs trained with a smaller dataset, with and without solution measurements such as viscosity, surface tension and conductivity. The validation data was taken from literature but was not used to train or test the models.

The average % errors of the validation data with and without solution measurements were 22.0% and 22.7% respectively. These results validate the hypothesis that good predictions can be made

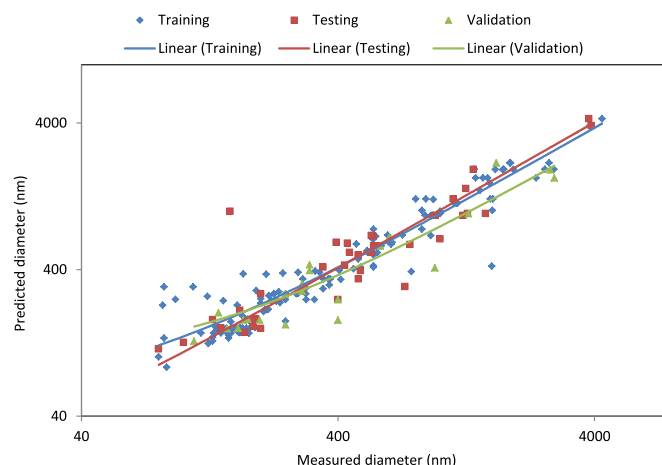


Fig. 5. Predicted versus measured diameter for a Generalised Regression Neural Network (GRNN) trained using data for 13 different polymer/solvent combinations from over 20 independent studies.

without the need for solution measurements, using only easily available prior knowledge.

## 6. Conclusion

The purpose of this work was to determine if ANNs could provide a generalised method of predicting electrospun fibre characteristics using only easily available, prior knowledge of the polymer, solvent, concentration and electric field strength. PNNs and GRNNs were found to outperform MLFNs and linear regression.

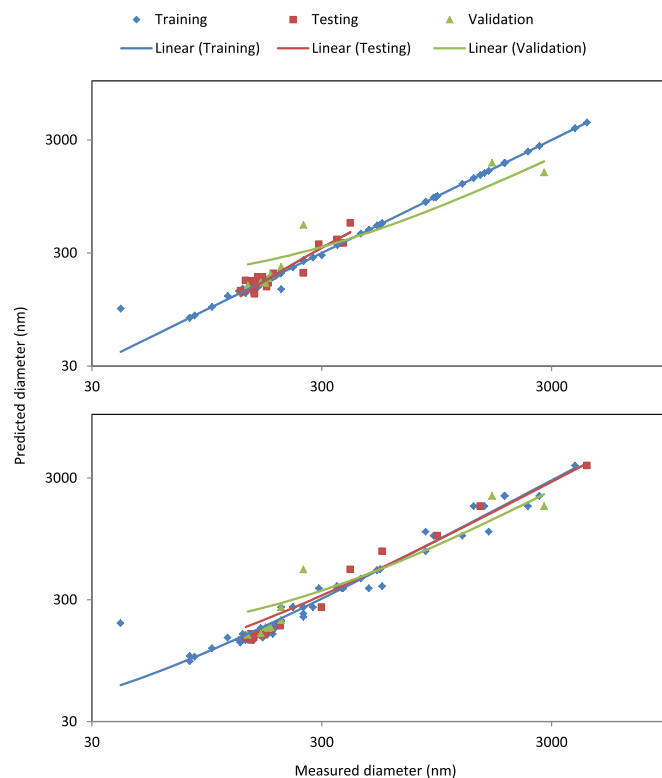


Fig. 6. Predicted versus measured fibre diameters for artificial neural networks (ANNs) trained with (top) and without (below) solution measurements.

Table 9  
Impact of parameters on fibre diameter from impact analysis.

Impact on diameter prediction	Parameter
Strong	Polymer concentration, Solvent viscosity, Polymer glass transition temperature, Polymer dielectric constant, Polymer molecular weight
Moderate	Electric field strength, Solvent density, Solvent boiling point
Weak	Solvent vapour pressure, Solvent surface tension, Solvent dielectric constant, Solvent conductivity, Solvent dipole moment

Trained PNNs were found to be effective in classifying fibre morphology and spin-ability, with the validation data making >80% correct predictions. Trained GRNNs were also effective in predicting fibre diameters, with an average % error of 22.3% for the validation data.

The inclusion of solution measurements was found to have a minimal impact on prediction performance. These parameter impact analyses for classification and diameter prediction constitute the largest quantitative study of this type known to the authors. The analysis helped us to focus on which parameters influence important electrospinning characteristics, namely fibre morphology and diameter. In addition to this, the success of the model goes some way to validate the exclusion of parameters such as feed rate and nozzle diameter.

The development of these tools will allow researchers to narrow down polymer, solvent and concentration combinations before carrying out time-consuming scoping exercises. Further development of the tools, by increasing the dataset size and reducing the errors associated with the input data, may also allow for direct selection of optimised parameters for a given purpose. An interesting extension of this work would be the inclusion of fibre diameter standard deviation as a dependent value.

This paper used data from single needle setups with primary solvent systems and therefore cannot be used to predict needleless electrospinning or multiple solvent systems. Adapting the method for binary or ternary solvents will require further research.

## References

- [1] Doshi J, Reneker DH. *J Electrostat* 1995;35(2–3):151–60.
- [2] Huang Z-M, Zhang YZ, Kotaki M, Ramakrishna S. *Compos Sci Technol* 2003;63(15):2223–53.
- [3] Yang F, Murugan R, Wang S, Ramakrishna S. *Biomaterials* 2005;26(15):2603–10.
- [4] Ayres C, Bowlin GL, Henderson SC, Taylor L, Shultz J, Alexander J, et al. *Biomaterials* 2006;27(32):5524–34.
- [5] Barnes CP, Sell SA, Boland ED, Simpson DG, Bowlin GL. *Adv Drug Deliv Rev* 2007;59(14):1413–33.
- [6] Sun T, Norton D, McKean RJ, Haycock JW, Ryan AJ, MacNeil S. *Biotechnol Bioeng* 2007;97(5):1318–28.
- [7] Ghasemi-Mobarakeh L, Prabhakaran MP, Morshed M, Nasr-Esfahani M-H, Ramakrishna S. *Biomaterials* 2008;29(34):4532–9.
- [8] Cantón I, McKean R, Charnley M, Blackwood KA, Fiorica C, Ryan AJ, et al. *Biotechnol Bioeng* 2010;105(2):396–408.
- [9] Deshpande P, Ramachandran C, Sefat F, Mariappan I, Johnson C, McKean R, et al. *Biomaterials* 2013;34(21):5088–106.
- [10] Mota C, Puppi D, Gazzarri M, Bártolo P, Chiellini F. *Polym Int* 2013;62(6):893–900.
- [11] Sefat F, McKean R, Deshpande P, Ramachandran C, Hill CJ, Sangwan VS, et al. *Procedia Eng* 2013;59(0):101–16.
- [12] Tamura T, Kawakami H. *Nano Lett* 2010;10(4):1324–8.
- [13] Miao Y-E, Zhu G-N, Hou H, Xia Y-Y, Liu T. *J Power Sources* 2013;226(0):82–6.
- [14] Gopal R, Kaur S, Ma Z, Chan C, Ramakrishna S, Matsuura T. *J Membr Sci* 2006;281(1–2):581–6.
- [15] Barhate RS, Ramakrishna S. *J Membr Sci* 2007;296(1–2):1–8.
- [16] Li D, Wang Y, Xia Y. *Adv Mater* 2004;16(4):361–6.
- [17] Chronakis IS. *J Mater Process Technol* 2005;167(2–3):283–93.
- [18] Teo WE, Ramakrishna S. *Nanotechnology* 2006;17(14):R89.
- [19] Cavaliere S, Subianto S, Savych I, Jones DJ, Roziere J. *Energy Environ Sci* 2011;4(12):4761–85.
- [20] Raghavan P, Lim D-H, Ahn J-H, Nah C, Sherrington DC, Ryu H-S, et al. *React Funct Polym* 2012;72(12):915–30.
- [21] Agarwal S, Greiner A, Wendorff JH. *Prog Polym Sci* 2013;38(6):963–91.
- [22] Mi Yeon S, Do Kyun K, Kyo Jin I, Seong Mu J, Dong Young K. *Nanotechnology* 2004;15(12):1861.
- [23] Ondarçuhu T, Joachim C. *EPL (Europhysics Letters)* 1998;42(2):215.
- [24] Theron SA, Yarin AL, Zussman E, Kroll E. *Polymer* 2005;46(9):2889–99.
- [25] Zhou F-L, Gong R-H, Porat I. *Polym Int* 2009;58(4):331–42.
- [26] Technology IoN. Global markets and technologies for nanofibers. 2013.
- [27] Yokoyama Y, Hattori S, Yoshikawa C, Yasuda Y, Koyama H, Takato T, et al. *Mater Lett* 2009;63(9–10):754–6.
- [28] Koski A, Yim K, Shivkumar S. *Mater Lett* 2004;58(3–4):493–7.
- [29] Son WK, Youk JH, Lee TS, Park WH. *Polymer* 2004;45(9):2959–66.
- [30] Theron SA, Zussman E, Yarin AL. *Polymer* 2004;45(6):2017–30.
- [31] Cui W, Li X, Zhou S, Weng J. *J Appl Polym Sci* 2007;103(5):3105–12.
- [32] Thompson CJ, Chase GG, Yarin AL, Reneker DH. *Polymer* 2007;48(23):6913–22.
- [33] Coles SR, Jacobs DK, Meredith JO, Barker G, Clark AJ, Kirwan K, et al. *J Appl Polym Sci* 2010;117(4):2251–7.
- [34] Yördem OS, Papila M, Menceoğlu YZ. *Mater Des* 2008;29(1):34–44.
- [35] Gu SY, Ren J, Vancso GJ. *Eur Polym J* 2005;41(11):2559–68.
- [36] Reneker DH, Yarin AL, Fong H, Kooomhongse S. *J Appl Phys* 2000;87(9):4531–47.
- [37] Hohman MM, Shin M, Rutledge G, Brenner MP. *Phys Fluids* (1994–present) 2001;13(8):2201–20.
- [38] Hohman MM, Shin M, Rutledge G, Brenner MP. *Phys Fluids* 2001;13(8):2221–36.
- [39] Shin YM, Hohman MM, Brenner MP, Rutledge GC. *Polymer* 2001;42(25):09955–67.
- [40] Feng JJ. *J Newt Fluid Mech* 2003;116(1):55–70.
- [41] Han T, Yarin AL, Reneker DH. *Polymer* 2008;49(6):1651–8.
- [42] Rayleigh L. *Proc R Soc Lond* 1879;29:71–97.
- [43] Saville DA. *Annu Rev Fluid Mech* 1997;29(1):27–64.
- [44] Fong H, Chun I, Reneker DH. *Polymer* 1999;40(16):4585–92.
- [45] Helgeson ME, Wagner NJ. *AIChE J* 2007;53(1):51–5.
- [46] Sarkar K, Ghahla MB, Wu Z, Bose SC. *J Mater Process Technol* 2009;209(7):3156–65.
- [47] Khanlou H, Sadollah A, Ang B, Kim J, Talebian S, Ghadimi A. *Neural Comput Appl* 2014;1–11.
- [48] Nasouri K, Shoushtari A, Khamforoush M. *Fibers Polym* 2013;14(11):1849–56.
- [49] Ojha SS, Afshari M, Kotek R, Gorga RE. *J Appl Polym Sci* 2008;108(1):308–19.
- [50] Casper CL, Stephens JS, Tassi NG, Chase DB, Rabolt JF. *Macromolecules* 2003;37(2):573–8.
- [51] Gupta P, Elkins C, Long TE, Wilkes GL. *Polymer* 2005;46(13):4799–810.
- [52] Palangetic L, Reddy NK, Srinivasan S, Cohen RE, McKinley GH, Clasen C. *Polymer* (0).
- [53] Luo CJ, Stride E, Edirisinghe M. *Macromolecules* 2012;45(11):4669–80.
- [54] Kim K, Kang M, Chin I-J, Jin H-J. *Macromol Res* 2005;13(6):533–7.
- [55] Uyar T, Besenbacher F. *Polymer* 2008;49(24):5336–43.
- [56] Biber E, Gündüz G, Mavis B, Colak U. *Appl Phys A* 2010;99(2):477–87.
- [57] Macosay J, Marruffo A, Rincon R, Eubanks T, Kuang A. *Polym Adv Technol* 2007;18(3):180–3.
- [58] Heikkilä P, Harlin A. *Express Polym Lett* 2009;3(7):437–45.
- [59] Stanger J, Tucker N, Wallace A, Larsen N, Staiger M, Reeves R. *J Appl Polym Sci* 2009;112(3):1729–37.
- [60] Hardick O, Stevens B, Bracewell D. *J Mater Sci* 2011;46(11):3890–8.
- [61] Luo X, Xie S, Liu J, Hu H, Jiang J, Huang W, et al. *Polym Chem* 2014;5(4):1305–12.
- [62] Deitzel J, Kleinmeyer J, Harris D, Beck Tan N. *Polymer* 2001;42(1):261–72.
- [63] Yan X, Geveler M. *J Electrostat* 2010;68(5):458–64.
- [64] Nurfaizey AH, Stanger J, Tucker N, Buunk N, Wallace A, Staiger MP. *J Mater Sci* 2012;47(3):1156–63.
- [65] Smallwood IM. In: Smallwood IM, editor. *Handbook of organic solvent properties*. Oxford: Butterworth-Heinemann; 1996.
- [66] Tao J, Shivkumar S. *Mater Lett* 2007;61(11–12):2325–8.
- [67] Zeng J, Haoqing H, Schaper A, Wendorff Joachim H, Greiner A. Poly-L-lactide nanofibers by electrospinning – influence of solution viscosity and electrical conductivity on fiber diameter and fiber morphology. *e-Polymers* 2003;3:102.
- [68] Yener F, Jirsak O. *Iranian J Chem Chem Eng (IJCCCE)* 2012;31(4):49–58.
- [69] Guerrini LM, Branciforti MC, Canova T, Bretas RES. *Mater Res* 2009;12:181–90.
- [70] Li L, Li R, Li M, Rong Z, Fang T. *RSC Adv* 2014;4(53):27914–21.
- [71] Choi S-S, Lee YS, Joo CW, Lee SG, Park JK, Han K-S. *Electrochim Acta* 2004;50(2–3):339–43.
- [72] Stanger JJ, Tucker N, Buunk N, Truong YB. *Polymer Test* (0).
- [73] Keun Son W, Ho Youk J, Seung Lee T, Park WH. *Mater Lett* 2005;59(12):1571–5.
- [74] Specht DF. *Neural Netw IEEE Trans* 1991;2(6):568–76.
- [75] Hornik K. *Neural Netw* 1991;4(2):251–7.
- [76] Hestenes MR, Stiefel E. *Methods of conjugate gradients for solving linear systems*. Washington, DC: National Bureau of Standards; 1952.