JOURNAL OF OPTOELECTRONİCS AND ADVANCED MATERİALS Vol. 9, No. 6, June 2007, p. 1618 - 1622

Neural Network applied to multifunctional materials

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Progress in optoelectronic and electronic sensor technology has enabled new capabilities in the field of sensor construction, particularly miniaturized pH sensors (ion-sensitive field-effect transistors, ISFETS). The possibility of covalent anchoring of polyHEMA on the surface of silicon nitride, using photolithographic techniques for crosslinking of 2-hydroyethyl methacrylate (HEMA) has already demonstrated in the literature. 2-Hydroxyethyl methacrylate, HEMA, and 2-hydroxyethyl methacrylate / diethylene glycol dimethacrylate, HEMA / DEGDMA, systems were chosen as model systems due to their importance as possibly of chemical binding of the ion-selective membrane to the silicon nitride surface. In the manufacturing of durable ion-selective sensors, the mechanical stability of sensors is very important. This property is affected by the light intensity and crosslinking ratio of the polyHEMA and polyHEMA / EGDMA layers produced. The aim of the present work was to develop a nonlinear empirical model and in this way to overcome the difficulty of using the complex polymerization kinetic model. This paper focuses on using the back-propagation multi-layer perceptron (BPMLP) algorithm to predict the monomer conversion. The neural network proposed in this study is very suitable to achieve improved control over the photopolymerization process, in the manufacturing processes of durable ion-selective sensors with photolithographic techniques.

(Received November 14, 2006; accepted April 26, 2007)

Keywords: Biosensors, HEMA, DEGDMA, Neural Networks, ISFETS

1. Introduction

The development of micropatterned surfaces, especially biomaterial surfaces, is of considerable interest for the biotechnology industry. These developments, especially biomaterial surfaces, has been extensively explored for applications ranging from biosensor technology [1] to tissue engineering [2, 3].

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The possibility of covalent anchoring of polyHEMA on the surface of silicon nitride, using photolithographic techniques for crosslinking of 2-hydroxyethyl methacrylate (HEMA) has already been demonstrated in the literature. A fundamental understanding of the kinetics and mechanism of photopolymerization reactions is very important for the development of applications in the biotechnology industry.

The high cross-linking density of these networks provides increased dimensional stability, high glass transition temperatures and decreased solvent absorption. Applied light intensity in photopolymerization reactions is a very important parameter. Light intensity is directly proportional to the initiation rate and can be varied to control the polymerization rate. High light intensities to photopolymerization reactions cause a high radical concentration that leads to primary radical termination and short branching [4, 5]. The resultant polymer is not solvent resistant as desired for some applications, such as optoelectronics, optical fiber manufacturing, video disk coatings, and photolithographic resists [6]. On the other hand, at low light intensities, initiation and polymerization rates may not be high enough to overcome the oxygen inhibition effect completely. Due to low initial rate, photopolymerization may stop short of completion.

In this work, to overcome the difficulty arising from several dozens of differential and algebraic equations and to achieve improved control over the photopolymerization process and resulting polymer formation, a back-prapogation multi-layer perceptron (BPMLP) algorithm is used.

Since photopolymerization processes are highly non-linear processes, non-linear empirical models should be developed. Neural networks have been shown to be able to approximate any continuous non-linear functions, and have recently been applied to non-linear process modelling [7]. An advantage of neural network based modelling is that a complex non-linear process model can be developed from process data only.

2. Experimental

2.1 Model description

The BPMLP network is one of the most commonly used Artificial Neural Network (ANN) architecture [8]. It consists of one input layer with M inputs, one output layer with N outputs and one or more hidden layers with varying number of nodes. In this study, two hidden layers are used in the BPMLP network and the number of inputs was two which corresponds to light intensity and time. There was a single output that corresponds to conversion of photopolymerization. The BPMLP neural network is a type of supervised, error correction learning that calculates an error on the output layer and propagates that error backwards through the network to determine how each individual weight factor contributes to the output error. The steepest-descent gradient approach used by the BPMLP to minimize the mean square error function and the local error function [8] defined as:

$$E_p = \frac{1}{2}\sum_{j=1}^{n}\left(d_{jp} - y_{jp}\right)^2 = \frac{1}{2}\sum_{j=1}^{n} e_{jp}^2 \qquad (1)$$

where, d_{jp} is the desired output signal of the jth output neuron for the pth example, n is the length of each example and y_{jp} is the output signal. The total error function is defined as:

$$E_T = \sum_p E_p = \frac{1}{2}\sum_p\sum_j\left(d_{jp} - y_{jp}\right)^2 \qquad (2)$$

The data from the input neurons is propagated through the network via the interconnections such that every neuron in a layer is connected to every neuron in the adjacent layers. Each interconnection has associated with it a scalar weight, which acts to modify the strength of the signal passing through it. The neurons within the hidden layer perform two tasks: they sum the weighted inputs to the neuron and then pass the resulting summation through a nonlinear activation function. The unipolar sigmoid activation function with its output in the range (0, 1) used in this study is as follows:

$$y_j = \varphi\left(u_j\right) = \frac{1}{1 + \exp\left(-\gamma_j u_j\right)} \qquad (3)$$

where, $\varphi(.)$ is the unipolar sigmoid activation function and u_j is defined in equation 4 with its bias value θ_j. In addition to the weighted inputs to the neuron, a bias is included in order to shift the space of the nonlinearity.

$$u_j = \sum_{i=1}^{n} w_{ji} x_i + \theta_j \qquad (4)$$

One way to improve the back propagation multiplayer learning algorithm is to smooth the weight changes by overrelaxation. In other words, by adding a momentum term defined as:

$$\Delta w_{ji}(k) = \eta \delta_j y_j + \alpha \Delta w_{ji}(k-1) \qquad (5)$$

where, $0 \le \alpha \le 1$

The second term in formula 5 is called the momentum term that makes the current (k-th) search directions. Momentum term enables the improvement of the convergence rate and the steady state performance of the back-propagation multi layer learning algorithm.

2.2. Experimental

The photoinitiator, 2,2-dimethoxy-2-pheny-lacetophenone (DMPA, 0.1 wt %, Ciba Geigy, Hawthorn, NY), 2-hydroxyethyl methacrylate (HEMA) and diethylene glycol dimethacrylate (DEGDMA) (Aldrich, Milwaukee, WI) were all used as received. Polymerizations at various light intensities were monitored using a FTIR spectrometer (Nicolet Model 760 Magna Series II FTIR, Nicolet, Madison, WI) equipped with DTGS - KBr and MCT/B - KBr detector-beam splitter combinations. Polymerizations were initiated by irradiation of the monomer/comonomer/initiator mixtures between sodium chloride crystals utilizing a UV light source (Ultra cure 100SS, 100 Watt Hg short-arc lamp, EFOS, Mississauga, Ontario, Canada) equipped with a liquid light guide. Light intensity was varied in the range of 0.5-45 mW/cm^2. Conversions were calculated from the decrease in the area of the double bond IR absorption band as a function of time.

3. Results

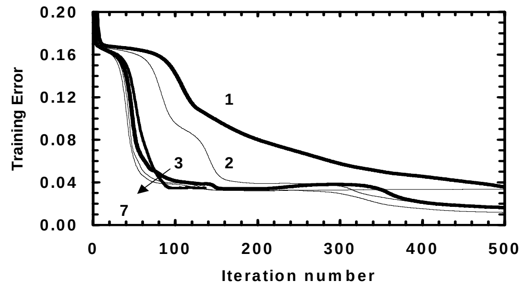
The prediction of the conversion in free-radical photopolymerization reactions of HEMA and HEMA/DEGDMA systems is important in the application of biosensor technology and it is highly nonlinear for different light intensities. In this paper, a back-propagation multi-layer perceptron (BPMLP), which is one of the artificial neural networks approach, is used to establish the relation between the conversion and light-intensity of both of the systems. To achieve this goal, for each system seven sets of data are used taken for different light intensities.

It is a well-known fact that for an insufficient number of training data, the training of a model may not be possible. In this study, the number of data at hand is not sufficient and hence the BPMLP network may not be trained in an adequate way. Knowing this constraint, we decided not to use any data for cross-validation. Hence, for this prediction problem, 5 sets of data are used in the training of BPMLP and the remaining 2 sets of data are used for testing phase. These data are given in Table 1.

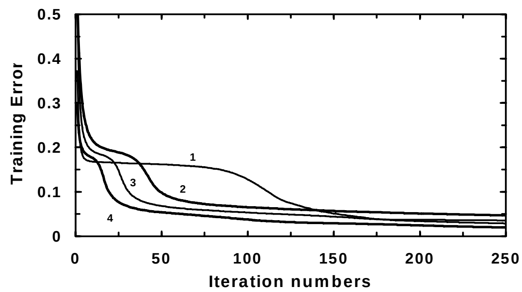
Table 1. Light intensities used in training and testing of neural networks for HEMA and HEMA/DEGDMA systems.

SET 1		SET 2	
HEMA/DEGDMA	System	HEMA system	
Training data	Testing data	Training data	Testing data
0.5 mW/cm^2	3.0 mW/cm^2	0.5 mW/cm^2	1.0 mW/cm^2
1.0 mW/cm^2	1.9 mW/cm^2	2.5 mW/cm^2	12.0 mW/cm^2
2.63 mW/cm^2		12.6 mW/cm^2	
4.1 mW/cm^2		15.0 mW/cm^2	
9.6 mW/cm^2		5.0 mW/cm^2	

The first experiment was performed in order to select the optimum learning parameter (μ) for the HEMA and HEMA/DEGDMA systems. For this experiment, the momentum coefficient (mp), the number of neurons in the first hidden layer (n_1) and the number of neurons in the second hidden layer (n_2) were fixed as 0.85, 7 and 3, respectively. Total training errors are plotted as a function of number of iterations in Fig. 1a for the HEMA system and Fig. 1b for the HEMA/DEGDMA system. In this paper, the weights are initialized randomly to some small real numbers. Because of this random initialization, the results may stack in different local points. In order to make our results more reliable, all the data presented in Figs. 1a/1b are obtained by averaging the 20 results obtained by running the same program for each case 20 times.



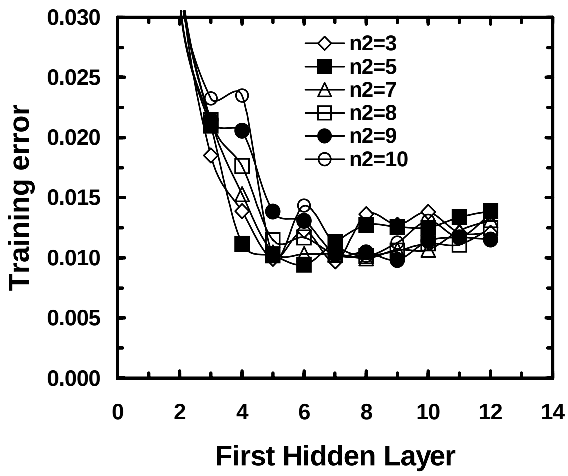
a



b

Fig. 1a. Total training errors plotted as a function of number of iterations for HEMA system (1: lp=0.4, 2: lp=0.35, 3: lp=0.3 …., 7: lp=0.1). Fig. 1b: Total training errors plotted as a function of number of iterations for HEMA/DEGDMA system (1: lp = 0.1, 2: lp = 0.2, 3: lp=0.3, 4: lp=0.4). (Iteration number x 20).

Another experiment was performed using the selected learning parameter values and keeping the momentum coefficient as 0.85 for both of the systems. The aim was to observe the significance of the number of neurons in both of the hidden layers. To do this, the number of neurons in the first hidden layer was changed from 1 to 12. The total training error was determined at each value of the first layer, by considering different number of neurons (3, 5, 7, 8, 9, 10) in the second hidden layer. Results are presented in Fig. 2a and Fig. 2b for HEMA and HEMA/DEGDMA systems, respectively.



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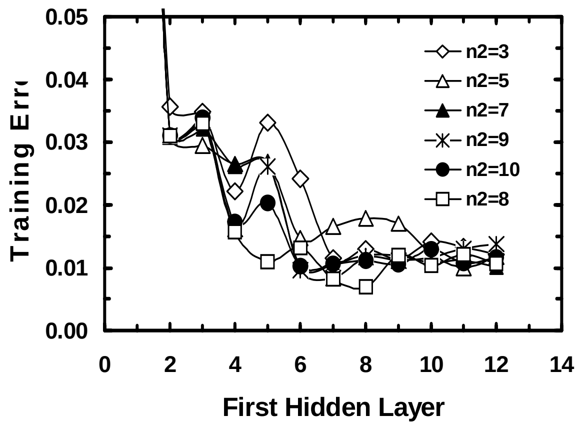
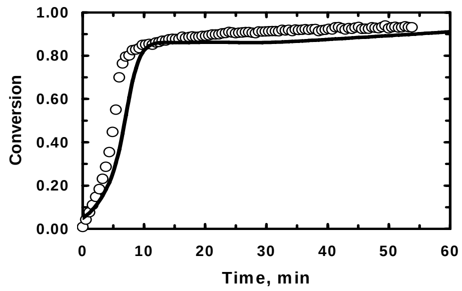


b

Fig. 2a. The total training error versus the number of neurons at the First Hidden Layer, by considering different number of neurons (3, 5, 7, 8, 9, 10) in the Second Hidden Layer for HEMA system. Fig. 2b: The total training error versus the number of neurons at the first Hidden Layer, by considering different number of neurons (3, 5, 7, 8, 9, 10) in the Second Hidden Layer for HEMA/DEGDMA system.

The results in Fig. 2a and Fig. 2b are obtained by averaging 20 trials for each case. Hence, the total training errors in these figures are the average errors that were obtained at the end of 10000 iterations. The training error shows an exponential decrease with the number of neurons in the first hidden layer up to about a certain value (5-6 neurons). It then starts to increase slightly. For the HEMA system (Fig. 2a), the best performance was obtained with six neurons in the first hidden layer and five neurons in the second hidden layer. The training error is minimum for this case. Fig. 2b revealed that, in the HEMA/DEGDMA system the best performance could be achieved when the number of neurons in both layers was eight.



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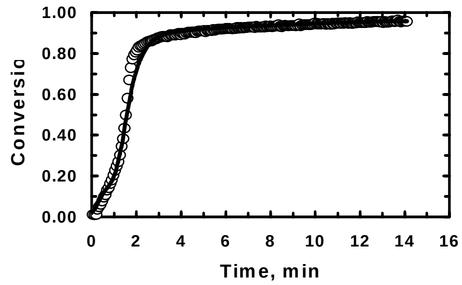

b

Fig. 3a. Experimental data points and the predicted conversion-time curve obtained for the HEMA system with neuron set 6:5, lp=0.1 and mp=0.85. Fig. 3b. Experimental data points and the predicted conversion-time curve obtained for the HEMA/DEGDMA system with neuron set 8:8, lp=0.4 and mp=0.85.

The optimized BPMLP was used to predict the polymerization behaviour of the HEMA and HEMA/DEGDMA systems in the last step. Figure 3a and Figure 3b present the comparisons of the experimental and predicted conversions as a function of time for both systems (ooo: Experimental data points, — : predicted data points).

4. Discussion

From the results obtained in Fig. 1a and Fig. 1b, one can observe that at low iteration numbers, the training errors are very high. The performance of the ANN increases as the number of iterations increases. The variation in total training errors is sufficiently small when the number of iterations is 10000 for different learning parameters. It may be deduced that, for a sufficiently large number of iterations, the importance of the learning parameter selection decreases. Hence, we selected the learning parameter as 0.1 and 0.4 for the HEMA and the HEMA/DEGDMA system, respectively. These parameters gave the minimum total training error for 10000 iterations.

The total training errors that are shown in Fig. 2a and Fig. 2b for the HEMA system and the HEMA/DEGDMA system respectively, shows the number of neurons in the first and second layers is very important in terms of performance assessment. Moreover, the importance of the number of neurons in the second hidden layer decreases as the number of neurons in the first hidden layer increases. In order to decrease the run-time and the storage of the weights, a BPMLP composed of two hidden layers seems to be the best choice.

We observed the performance of the ANN for the testing data. We plotted one result for the HEMA system and one result for the HEMA/DEGDMA system. The results showed that the performance of our model is very good in general for both of the systems. However, from the figures, the performance of our model is better for the HEMA/DEGDMA system. It has predicted the polymerization behavior with great accuracy especially in the autoacceleration region which is the most important regime. In this region, a great amount of monomer converts into polymer in a short time compared to other regimes making it crucial to know the exact behavior. The neural network is less successful in modelling the HEMA polymerization on this region. The reason may be attributed to less efficient training. The number of the experimental data points obtained from the polymerization of HEMA at each condition was less than the HEMA/DEGDMA system due to less frequent data collection. A more precise prediction can be achieved by increasing the number of training sets.

5. Conclusions

Back-propagation multi-layer perceptron is used to predict the monomer conversion in free-radical photopolymerization reactions of HEMA and HEMA/DEGDMA systems. BPMLP is obtained by developing the forward propagation phase and the sensitivity network. It is shown that, the generalization capability can be significantly improved by optimizing some of the parameters such as the learning parameter, momentum coefficient and neuron set. This technique requires only a limited amount of conversion versus time data sets obtained experimentally at certain polymerization conditions. Since UV light intensity is one of the most important parameters in the photolithographic technique, we have shown in this work that it is possible to predict the photopolymerization behavior at different light intensities. The same network can be trained for predictions in other experimental conditions and it is possible to have a single network feed by several inputs as polymerisation conditions. Therefore, by using ANNs, the development of complex polymerization kinetic models and the problem of numerical integration of a large number of complex differential equations were avoided. The neural network proposed in this study is very suitable to achieve improved control over the photopolymerization process, in the manufacturing processes of durable ion-selective sensors with photolithographic techniques.

Acknowledgement

The authors would like to thank to Prof. C.N. Bowman for the opportunity of using the data obtained in his laboratory.

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