RESEARCH OF SWELLING KINETICS OF ELASTOMERS FILLED WITH POLYACRYLAMIDE USING NEURAL NETWORK FRAMEWORK

Anatoly K. Pogodaev[1], Olga V. Karmanova[2], Sergey G. Tikhomirov[2], Semen L. Podvalny[3], Elena A. Balashova[2], Alexander S. Moskalev[2]

[1] *Lipetsk State Technical University*
30 Moskovskaya St. 398600, Lipetsk, Russian Federation
[2] *Voronezh State University of Engineering Technologies*
19 Revolution Av. 394036, Voronezh, Russian Federation
E-mail: karolga@mail.ru
[3] *Voronezh State Technical University*
14 Moskovsky Av. 394016, Voronezh, Russian Federation

Received 17 June 2020
Accepted 20 October 2020

ABSTRACT

The kinetics of the swelling process of crosslinked elastomeric materials filled with polyacrylamide (PA) in water is studied. The analysis of the dependence of the swelling degree and technological properties in the studied samples on the content of polyacrylamide and vulcanization conditions is carried out. It was found that the rate and maximum swelling degree increase with increasing polyacrylamide content. To ensure a high swelling degree of elastomeric materials ~ 500 %, the introduction of 80 phr PA is recommended, the vulcanization time at a temperature of 120°C should be 30 minutes.

Due to the fact that the swelling process dynamics of samples has a pronounced nonlinearity, an artificial neural network framework was used to model the properties of the composites. It was revealed that it is not practical to use the multilayer architecture of the neural network to simulate the swelling of elastomers, and preliminary classification of the object properties using the Kohonen networks (self-organizing map) should be performed, and then the properties for each distinguished class should be separately simulated. The average relative error in calculating the polymer swelling degree according to the proposed model with a two-step approach (at the first stage, objects are classified, and at the second, the polymer properties of each class are simulated) does not exceed 5 %.

Keywords: waterproofing, elastomers, swelling, polyacrylamide, vulcanization, modeling, neural networks, Kohonen network.

INTRODUCTION

Elastomeric materials and products based on them are widely used in various fields of industrial production due to their low specific density, high strength, wear resistance and resistance to aggressive environment. A significant amount of elastomeric materials is used as waterproofing in the construction industry. Waterproofing is mainly carried out in the underground parts of buildings and structures, which are affected by a humid environment, groundwater or precipitation (foundations, basements, tanks, etc.). Therefore, reliable insulation of building and structure elements in contact with an aggressive environment is an important task both in the design and construction [1, 2].

At present, water-swelling cords, tapes and sealants based on polymers are used for waterproofing. Water penetrating the joint leakage leads to swelling of the material filling the filtration zone, which stops leakage. The swelling degree of materials of various nature and composition is from 100 % to 600 %. In addition to the requirements for providing elastic-strength characteristics, resistance to weathering, such materials are required to achieve a given swelling degree during operation [3].

In this regard, the urgent task is to develop formulations of elastomeric compositions for the production of boundedly water-swellable seals that provide the required range of operational characteristics.

An analysis of the references [4-6] showed that ethylene propylene, nitrile butadiene, and styrene butadiene rubbers are used as the basis for water-swellable elastomeric materials. Oligomeric resins, polyacrylates, cellulose derivatives, starch derivatives, acrylamide polymers, polyvinyl alcohol and other compounds [7, 8] are used as hydrophilic additives. Acrylamide-based polymers are the most promising as additives that provide the necessary water-swelling of elastomeric waterproofing materials [8, 9].

The aim of this study is to develop water-swellable elastomeric compositions based on styrene butadiene rubber and polyacrylamide.

EXPERIMENTAL

When creating water-swellable seals, it is important to ensure their integrity throughout the entire life cycle. One of the technological methods providing improved operational characteristics is vulcanization. However, it should be borne in mind that the crosslinking of rubber macromolecules, in the matrix of which a hydrophilic component is distributed, reduces its ability to swell.

The polymer basis of water-swellable elastomeric materials (WSEM) was styrene butadiene rubber SBR-1705 manufactured by SIBUR. AK631 polyacrylamide (PA) was used as a hydrophilic additive, the content of the additive ranged from 50 phr to 80 phr. Fillers, plasticizers, vulcanizing group (sulfur, 2,2'-dibenzothiazolyl disulfide, zinc oxide, stearic acid) were also included in the recipe of the composition. The compositions were prepared on laboratory roller mill LB 320 160/160 with a friction of 1:1.14 at a temperature of 60 ± 5ºC. Washers were cut from the obtained rubber mixture samples and vulcanized without pressure at a temperature of 120°C.

The change in the mass of rubbers during their swelling in water was carried out according to GOST 9.030-74. The swelling degree of samples in water was evaluated by the change in their mass:

$$\alpha = 100(m - m_0)/m_0$$

where m_0, m are the masses of the sample before and after swelling, respectively.

RESULTS AND DISCUSSION

The swelling kinetics of elastomeric samples with different PA contents obtained under different vulcanization conditions is shown in Fig. 1.

An analysis of the dependences presented in Fig. 1 indicates that an increase in the duration of vulcanization contributes to a decrease in the swelling degree. The swelling degree increases with increasing PA content. However, the shape of the curves indicates an ambiguous effect of the vulcanization time on the swelling kinetics of WSEM with different PA contents.

Selecting the right combination of the PA content and the vulcanization time of the composition, which meets the operation requirements, is a difficult task. Therefore, it is necessary to conduct research on modeling the WSEM properties in a wide range of changes in their composition and production conditions, evaluate the accuracy of the proposed modeling methods and interpolate the polymer properties when changing the mode of their production [9, 10].

Since the dynamics of the composition swelling process has a pronounced nonlinearity (Fig. 1), in which two significant periods can be distinguished, one should expect high accuracy in modeling the properties of polymer compositions using neural networks, which are a non-linear transformation of input data into output values. For interpolation of data, multilayer feed forward neural networks, trained on the basis of the back propagation algorithm, have proven themselves well. An overwhelming number of works in this area relates to the processing of large amounts of statistical data, while in

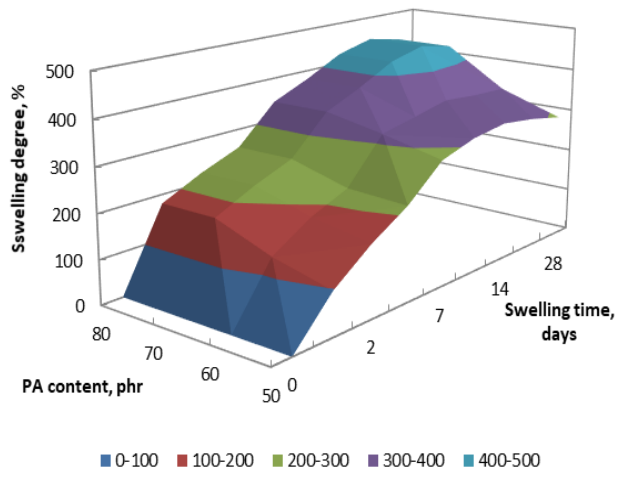

Fig. 1. WSEM swelling kinetics depending on the PA content and the vulcanization time.

the case under consideration modeling of the dynamics of multicomponent composition properties is required. Modeling of the dynamic properties requires special approaches to building a training set and training a network. It is possible to use a multialternative approach [11] for these purposes, involving the use of several simple models instead of one complex model with a large number of identifiable parameters. In addition, the processing of dynamic information often requires the preliminary feature extraction and classification for subsequent processing.

Classification organizes objects into homogeneous groups. The identification of the data structure in the training set can be performed using Kohonen networks when the number of classes is not known in advance. Neurons of a self-organizing network are trained to distinguish objects that are similar to each other from a set of n-dimensional objects. Kohonen networks exist in two versions: Kohonen layers (with disordered neurons) and Kohonen maps (arranged in such a way that close neurons correspond to close data clusters) [12, 13].

The training sample consisted of 12 different combinations of the mass parts of the water-swellable PA additive and the vulcanization time. The PA content ranged from 50 phr to 80 phr in increments of 10, the vulcanization time varied from 30 to 40 minutes in increments of 5 minutes. The dynamics of the polymer swelling degree (α, %) was estimated at 1, 2, 3, 7, 10, 14, 21 and 28 days. Each series of experiments consisted of 2 to 4 parallel experiments. The input data were the PA content (phr), the vulcanization time (min) and the swelling time (day). The output value is the polymer swelling degree (α, %).

The modeling of the properties of polymer water-swellable compositions was carried out using two-layer, three-layer and four-layer neural networks. An analysis of the results showed that the two-layer network cannot restore the results of all experiments with acceptable accuracy: the average error is 166.7 %. An increase in the number of layers to three makes it possible to reduce the average error to 9.68 %, however, individual points of the experiment are restored with an error reaching 250 %. The complexity of the network architecture to 4 layers allows you to reduce the average error to 5.95 % and the maximum error to 63.14 %.

Obviously, the difficulties of restoring the initial data are associated with a large heterogeneity of the properties of the considered groups of polymers. To improve the accuracy of modeling the properties of polymer compositions, it is necessary to identify classes of polymers with similar trends in the swelling degree from the initial data set.

Preliminary classification of the initial set in order to distinguish homogeneous groups was carried out using a self-organizing Kohonen network. The maximum number of distinguished classes varied from 12 to 6. The initial data for classification were the PA content, the vulcanization time, and the dynamics of changes in the polymer swelling degree at 9 time points, i.e. the number of inputs - 11. As a result, the following structure of the training set was obtained (Fig. 2).

The strength of relationship was established between classes (Fig. 3).

Since according to Fig. 3, illustrating the degree of the strength of relationship between classes using color intensity (white to black), the relationship between class 1 and class 4 is very weak (displayed in white), that is, the polymer swelling degree assigned to class 1 and class 4 has completely different the dynamics of the swelling degree.

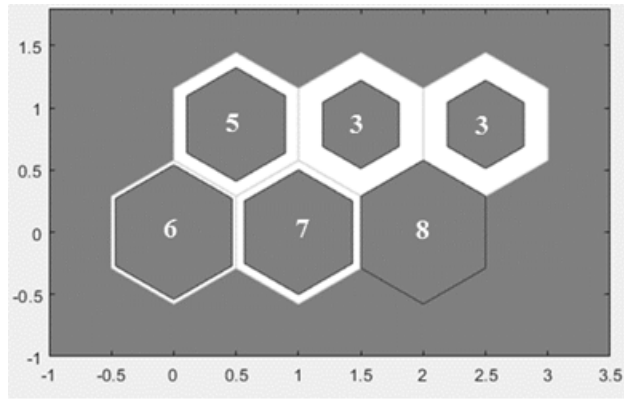

Fig. 2. An example of the training set structure.

Fig. 3. The strength of relationship between classes.

Table 1. The results of the WSEM classification.

The PA content, pts.wt.	Vulcanization time, min		
	30	35	40
50	Class 3		Class 2
60	Class 5-6		
70		Class 4	
80		Class 1	

Table 2. Modeling errors in the WSEM swelling degree.

Class	Relative errors, %					
	Two-layer network		Three-layer network		Four-layer network	
	Maximum	Average	Maximum	Average	Maximum	Average
1	9.5	2.65	12.34	2.58	10.29	2.48
2	24.07	3.75	26.25	3.75	–	–
3	32.69	3.64	30.10	3.75	–	–
4	32.69	4.79	28.26	5.08	–	–
5-6	49.5	6.22	21.12	3.98	–	–

Since objects of class 5 are close in properties to objects of class 4 (in Fig. 3, the relationship is indicated in gray), and the results of experiments combined in class 6 are close in properties to objects of class 3 (highlighted in dark gray), and between the objects of classes 2 and 5, the closest relationship is noted (highlighted in black), the experiments included in classes 5 and 6 are combined into one class. Thus, for the classification obtained (Fig. 1), it was decided to combine two classes that are similar in WSEM properties to one (Table 1).

Analysis of the degree of influence of individual inputs on the classification result showed that the PA content and the vulcanization time are important features. Considering their influence on the arrangement of class centers separately, the combination of classes 5 and 6 is justified, since their centers are very close (63.3; 30) for class 5 and (76.7; 30) for class 6 and are at a distance of 13, 4. For further modeling of the polymer composition properties, the initial set was divided into 5 classes. Modeling the WSEM swelling degree in each group was carried out separately. To restore their properties, multilayer feedforward neural networks, trained on the basis of the back propagation algorithm, were used. The number of layers ranged from 2 to 4.

Class 1 was composed of WSEMs, which contain 80 phr and were obtained at the vulcanization time of 35 min and 40 min. The output value is a vector of 9 elements corresponding to the swelling degree in a given time period. The modeling of WSEM properties was carried out using a two-layer neural network with 100 neurons in the inner layer, a three-layer network with 120 neurons in the first inner layer and 70 in the second, and four-layer network with 120 neurons in the first inner layer, 70 in the second, and 30 in the third. The output layer of all networks contained 9 neurons. With the complexity of the network architecture, an increase in the training time is observed, however, the modeling error decreases slightly (Table 2).

Modeling the WSEM swelling degree in each group was carried out separately. In the inner layers, the sigmoid activation function (the hyperbolic tangent) was used. In the outer layer, the linear activation function was used.

Table 2 shows the calculation results of errors in modeling the WSEM properties for each distinguished class.

The results of modeling the properties of the Class 1 objects obtained using a two-layer network are superior in accuracy to the values obtained using a four-layer network for the entire training set. The average relative error in absolute value is two times lower - 2.65 % for the Class 1 objects versus 5.95 % for the entire set. The maximum error was 9.5 %, which is several times lower than for the entire set - 63.14 %. This circumstance allowed us to make the assumption that there is no need to complicate the network architecture, which was confirmed experimentally by comparing the simulation results of a two- and three-layer network (similar to the considered case of architecture) to restore the properties of the remaining four classes.

CONCLUSIONS

To simulate the properties of water-swelling elastomeric materials based on an high-accuracy neural network, a larger number of measurements are required within each experiment. High accuracy of modeling the properties of water-swellable elastomeric materials can be ensured by performing a preliminary classification of the object properties using the Kohonen network, and then performing the property recovery procedure for each class separately. Moreover, to describe the properties of water-swelling elastomeric materials, it is sufficient to use a two-layer neural network for simple classes and a three-layer network for an enlarged class. The average relative error in modeling the polymer swelling with the proposed two-stage approach does not exceed 5 %.

REFERENCES

1. B.M. Rumyantsev, O.B. Lyapidevskaya, A.D. Zhuko, Systems of insulation of building structures, Publishing House Mosk. State Builds. University, 2016.
2. S.D. Sokova, The use of innovative technologies in the repair of buildings: a monograph, Publishing House MGSU, 2011.
3. T.M. Bochkareva., R.D. Urmancheev, Analysis of the use of traditional and modern waterproofing technologies in the reconstruction of buildings and structures, Modern technologies in construction, Theory and Practice, 12018C, 207-217.
4. S.N. Prokhorova, E.E. Potapov, F.P. Iordansky, V.V. Ivanov, I.S. Pyatov, The study of the processes of water swelling of packer rubbers, Rubberand R, 77, 1, 2018, 30-32.
5. N.F. Ushmarin, S.I. Sandalov, D.V. Pelipenko, E.N. Egorov, N.I. Koltsov Development of rubber-limited swellable rubber for packers, Rubber and Rubber, 78, 4, 2019, 214-219.
6. O.V. Karmanova, A.S. Moskalev, S.G. Tikhomirov, Yu. F. Shutilin, The composition and technological aspects of obtaining water-swelling elastomeric materials, Advanced Materials& Technologies, 1, 2019, 45-48.
7. V.F Kablov., N.A. Keybal, T.V. Krekaleva, E.V. Shaporov, E.E. Sitnikov, Development of oil-swellable elastomers for sealing elements of packer equipment, Bulletin of the Volgograd State Technical University, 228, 5, 2019, 63-66.
8. O.V. Evsikova, S.G. Starodubuev, A.R. Khokhlov, Synthesis, swelling and adsorption properties of composites based on polyacrylamide gel and sodium bentonite, Polymer science, Series A, 44, 5, 2002, 802-808.
9. A.K. Pogodaev, S.G. Tikhomirov, A A. Khvostov, O.V. Karmanova, S.L. Podvalny, Enhancing the composition of the polymer compound on the basis of the «Composition-property» model, J. Chem. Technol. Metall., 52, 4, 2017, 751-756.
10. A.K. Pogodaev, S.G. Tikhomirov, O.V. Karmanova, E.A. Balashova, S.L. Podval'ny, A. Yu. Fatneva, Modeling elastomer properties in presence of a composite vulcanization activator, J. Chem. Technol. Metall., 53, 5, 2018, 807-815.
11. S.L. Podval'ny, T.M. Ledeneva, Intelligent modeling systems; design principles, Automation and Remote Control, 74, 7, 2012, 1201-1210.
12. M.V. Pavlyuchenkova, N.P. Prokudenkov, A way to control industrial facilities based on a recurrent neural network, International Journal of Information Technology and Energy Efficiency, 2019, 4, 2, 18-24.
13. G.I. Rudoy, The choice of the activation function in forecasting by neural networks, Machine Learning and Data Analysis, 1, 1, 2011, 16-39.