Finite Element Simulation of Indentation Experiment on Branched Epoxy Novolac Resin

Andrejs Kovalovs¹, Andris Chate¹, Sergejs Gaidukovs², Arturs Medvids³

¹ Riga Technical University, Institute of Materials and Structures, Kipsalas St. 6A, LV-1048, Riga, Latvia
² Riga Technical University, Institute of Polymer Materials, Paula Valdena St. 3, LV-1048, Riga, Latvia
³ Riga Technical University, Institute of Technical Physics, Paula Valdena St. 3, LV-1048, Riga, Latvia
andrejs.kovalovs@rtu.lv

Abstract. The main purpose of this study was to investigate the micro-indentation test method on the branched epoxy novolac resin (ENR) and to validate it by means of finite element simulation. At the first stage, the micro-indentation test was carried out with the purpose of establishment of the Young’s modulus of ENR by the empirical relation to polymer materials. Experiments were performed by using the Vickers-type indenter. At the second stage, the finite element model was used to model the load-displacement curve of the material with the use of the initial values of elastic modulus and yield strength of material. The effectiveness of the use of these properties was investigated by using the planning of experiments and response surface technique. Full factorial design method was used to perform a series of numerical experiments. Then, a comparison of the results of micro-indentation test and finite element simulations was performed in order to compare and validate the values of Young’s modulus obtained from two different techniques. Good correlation between the experimental and numerical Young’s modulus was observed.

1. Introduction
The non-destructive experimental procedure is currently being widely studied by many researchers. The traditional test methods for measuring the mechanical properties of the materials, such as the Young’s modulus or yield strength, are often destructive and need numerous sample materials.

The indentation test is an alternative method to establish the mechanical properties of the materials. This method is widely used to establish the hardness of the material, which is obtained by dividing maximum indentation load by the contact area. The method is non-destructive and inexpensive [1].

Many studies were carried out in order to find an empirical relationship between the hardness and mechanical properties of the materials [2]. Load-displacement curve, which was recorded during the test, showed sensitivity to the mechanical properties [3].

Some researchers are studying indentation process by using Finite Element analysis. Application of FE analysis can be employed for investigation of the stress and strain field under indenter. The results obtained from numerical simulation can be compared with the experimental test to validate indentation process. The previous results show that the finite element simulation could be a good method for the establishment of the material hardness [4-6].

The main purpose of this study is to investigate the micro-indentation test method on branched epoxy novolac resin (ENR) and to validate it by finite element simulation. At the first stage, the micro-indentation test with Vickers indenter was carried out with the purpose of establishment of the
Young’s modulus of ENR. At the second stage, the finite element model was used to model the load-displacement curve of the material with the initial values of elastic modulus. Then, a comparison between the results of micro-indentation test and FE simulation was performed to compare and validate the values of Young’s modulus obtained from two different techniques.

2. Indentation Process

Microhardness was used as a very powerful applied technique in order to characterize the outcome of the finished coating photocuring process. Thereto, Vickers micro-indentation hardness (HV) of the polymer is related to the macroscopic mechanical properties, such as the empirical relation \(HV = E/10\) for polymer materials [7]. For validation of this relation the finite element simulation of the micro-indentation test was performed.

According to [8], the Vickers hardness value was calculated by using the following equation:

\[
HV = 0.189 \frac{F}{d^2}
\]  

(1)

where \(F\) – the indentation force, N; \(d\) – average length of the two diagonal left by the indenter, mm.

Correspondingly, by knowing the Vickers micro-indentation hardness one can solve the inverse problem. The identification of mechanical properties of coating materials was performed by employing numerical experimental method based on experiment design method and approximation technique. 3D finite element simulation was carried out by using specimens and diamond indenter. The indenter was penetrating to the calculated depth with various values of elastic properties of the test material. The indentation forces obtained from numerical simulation were compared with those obtained from the experiments to minimize the gap between the experimental and numerical parameters of response.

The depth of contact of the indenter in the test material was established from the following equation:

\[
h = \frac{d}{2 \tan \frac{136^\circ}{2}}
\]  

(2)

where \(d\) was calculated from the Vickers hardness value:

\[
d = \sqrt{\frac{0.189 F}{HV}}
\]  

(3)

3. Materials and Methods

3.1. Materials

The branched poly (phenyl glycidyl ether)-co-formaldehyde obtained from Advanced Polymer Coatings was used as ENR for the photopolymerization synthesis. The resin’s viscosity is 2,000 cPs, and the specific gravity is 1.21 g/cc. The UV 1242 cationic photoinitiator (PI) bis (4-dodecylphenyl) iodonium hexafluorantimonate was obtained from Deuteron GmbH. Hereafter, PI for the photoinitiator will be used in the text. As a reactive diluent, C12-, C14-glycidylether was applied. Its content is maintained at approx. 1 part per 1 part of PI. Acetone (>99.5%) obtained from Aldrich. For reference, a commercial two-component ENR formulation was used. The commercial mixture of amine and imidazole obtained from Advanced Polymer Coatings was used as a curing agent.

3.2. Methods

3.2.1. Sample Preparation

Samples were prepared by solution casting and UV curing method. First, liquid mixtures of poly (phenyl glycidyl ether)-co-formaldehyde with a pre-calculated amount of bis (4-dodecylphenyl) iodonium hexafluorantimonate and toluene were blended in a glass flask. The bis (4-dodecylphenyl)
iodonium hexafluorantimonate blend with 50 wt% C12-/C14-glycidilether was used in the preparations. The real weight content of the photoinitiator in the epoxy compound was adjusted to 0.5, 1.5 and 3 and 6 wt%. The mixtures of photoinitiator (PI), ENR were coated on a commercially available glass slides and steel plates. No special preparations of the substrate surface were applied to the specimens. Finally, the coating films were cured at room temperature for 1, 3, 5, 10, 20, and 50 min under UV irradiation with 96.5 W/cm² intensity and at a distance of 13 cm from the irradiation source. The coating thickness was in the range 120-170 μm. Accordingly, 1000 W Hg deep UV exposure lamp with broad emission spectra range from 200–600 nm was utilized for photopolymerization curing experiments in air environments.

3.2.2. Micro-Indentation Experiment
The micro-indentation tests was carried out by using a Vickers-type indenter according to the standard procedure (ISO 14577-4). The microhardness was measured on a Vickers M-17 1021 device equipped with optical microscope with a 50-g (0.49 N) load and loading time of 20 s. At least five indentations were made in order to check the repeatability of the measurements.

3.2.3. Response Surface Technique and Plan of the Experiment
The basic idea of this approach is that simple mathematical models (response surfaces method - RSM) are established only by using the finite element solutions in the reference points of the experimental design. At the first stage, a plan of experiments was produced depending on the number of design variables and number of the experiments. At the second stage, the numerical model was created in order to model the response of a structure, and then the finite element analysis was carried out in the reference points of the experimental design. At the third stage, the numerical data obtained by the finite element calculations in the sample points were used in order to build the approximating functions by using the response surface method.

The objective of the present study was to establish the functional relationships between the research parameters and indentation force. The numerical experiments were designed according to Full Factorial design. The 4k factorial design was used to supply a relationship for indentation force as mathematical function of research parameters, where k is the number of parameters and the base 4 represents the level of treatment for each considered parameter [9]. The plan of experiments was formulated for 2 parameters, Young’s modulus (X1), Yield strength (X2), and 16 experiment points. The levels are reported in Table 1.

| Parameters               | Levels  |
|--------------------------|---------|
| Young’s Modulus [MPa]    | 7000    |
|                          | 7100    |
|                          | 7200    |
|                          | 7300    |
| Yield Strength [MPa]     | 420     |
|                          | 440     |
|                          | 460     |
|                          | 480     |

3.2.4. Finite Element Model
For comparison and validation of the micro-indentation test, 3D quarter-symmetry finite element (FE) model with Vickers indenter was used. In order correctly to simulate the indentation phenomenon, a contact analysis was conducted. 3D Finite element model was modelled by using SOLID185 elements. Target elements (TARGE170) were placed along the top surface of the resin and the contact elements (CONTA174) were used along the bottom surface of the indenter. The indentation was simulated by applying displacement boundary conditions in the y-direction to the nodes along the upper surface of the indenter. All nodes at the bottom of material were constrained to prevent them from moving in the x and z directions. Fragment of FE model is shown in Figure 1.

A pyramidal indenter with 68° face angle was used. This design provided the same projected area to depth as a Vickers indenter. The simulation of contact was carried out in two steps: loading and unloading. In the first step, the indenter was moved downwards along y-axis to calculate the depth (Figure 2). In the second step, the indenter was moved upwards. The contact between indenter and material was removed.
The material of indenter is the Diamond with the elastic modulus of 1140 GPa and Poisson’s ratio of 0.07. The Poisson’s ratio of the research material is constant 0.2. The micro-indentation procedure was terminated at a calculated penetration depth of 2.31 µm. The friction coefficient between the indenter and the sample was equal to 0.15.

4. Results and Discussions

The experiments showed that the highest hardness value equal to 712 MPa of coating was received with 1.5% of PI, and 3 min of UV-irradiation due to the very dense cross-linking network structure was developed in the polymer. According to the empirical relation HV~E/10 for polymer materials, the Young’s modulus of the ENR was 7120 MPa.

The numerical data obtained by the finite element calculations in the points of plan of experiments was used to build the approximating function by using the program EdaOpt [10]. A second-order polynomial equation is the following:

\[ F(x) = 0.56 - 9.32 \times 10^{-5} x_1 + 1.09 \times 10^{-5} x_2 + 6.25 \times 10^{-6} x_1 x_1 - 3.13 \times 10^{-7} x_1 x_2 + 9.00 \times 10^{-8} x_1 x_2 \]  

(4)

where \( x_1 \) - Young’s modulus and \( x_2 \) – Yield strength.

![Figure 1. 3D Quarter-Symmetry Finite Element Model.](image1)

![Figure 2. Distribution of Displacement.](image2)

**Figure 1.** 3D Quarter-Symmetry Finite Element Model.

**Figure 2.** Distribution of Displacement.

![Figure 3.](image3)

**Figure 3.** Accordance Between Approximation Function and Control Points: a-Young’s Modulus is 7300 MPa; b- Yield strength is 46 MPa
The approximating function was verified by the finite element solutions in the points different from
the points taken in the plan of experiment. Examples of finite element verification of the response
surfaces are presented in Figure 3, where a very good correlation is observed for the approximations
and finite element solutions.

After selection of equation of regression the parametric studies were carried out to study the
influence of the research parameters on the indentation force. The view of the response surfaces are
shown in Figures 4. This figure shows the indentation force dependency on the Yield stress and
Young’s modulus. It can be seen that the value of the indentation force significantly increases with an
increase of strength and modulus.

It is assumed that the indentation force does not exceed 0.49 N in order to detect the Young’s
 modulus and Yield strength. It is presented in Figure 4 that there are many optimal solutions that
satisfy the condition under a constant load of 0.49 N. Therefore, to establish the elasticity modulus of
the material and to compare it with the results obtained from the experiment, the minimum and
maximum values of the elasticity modulus from the possible range of the obtained solutions were
selected.

The results of finite element simulations and the experimental test are presented in Table 2. It is
seen that the minimal Young’s modulus (7177 MPa) has the biggest Yield stress (48 MPa). Difference
between the experimental and numerical results is 0.8%.

![Figure 4. Dependency on the Indentation Force.](image)

Similarly, the optimal result was obtained for minimal the Yield strength. The minimal Yield
strength is 47 MPa, when the maximum Young’s modulus is 7293 MPa. The difference between
experimental and numerical modulus reaches 2.4%. It is seen that these differences are very little.
Mostly residuals do not exceed 3%, which shows the good correlation between the approximating
functions and numerical model. The load displacement curve obtained from the finite element
simulation of the micro-hardness test with a Vickers indenter is shown in Figure 5.

| Table 2. Young’s modulus and Yield stress |
|------------------------------------------|
| Experiment | RSM 1 | Δ,% | RSM 2 | Δ,% |
| Young’s Modulus [MPa] | 7120 | 7177 | 0.8 | 7293 | 2.4 |
| Yield Strength [MPa] | – | 480 | – | 470 | – |
5. Conclusions

The micro-indentation experiment was performed on the branched epoxy novolac resin (ENR) by using Vickers indenter with the purpose of the establishment the Young’s modulus of material. According to empirical relation for polymer materials, the value of Young’s modulus was obtained. Then the finite element model was used to simulate the micro-indentation process in order to check the elastic properties.

Based on the indentation load and displacement data extracted from the finite element simulation and from the micro-indentation experiment, the Young’s modulus of branched epoxy novolac resin (ENR) was compared. As it can be seen, there is only a little difference between the Young’s modulus obtained from finite element model and from the results of the experiments. The difference between the Young’s modulus values obtained from the finite element simulation and from the micro-indentation experiment is negligible (0.8%). Additionally Yield strength of the material was defined.

References
[1] Z. Chen, K. Zhou, X. Lu, Y.C. Lam, “A review on the mechanical methods for evaluating coating adhesion,” Acta Mech, Vol. 225, pp. 431–452, 2014.
[2] A.C. Fisher-Cripps, Introduction to Contact Mechanics. – Springer Verlag, New York, 2000.
[3] K. Komvopoulos, “An elastic–plastic analysis of spherical indentation: Constitutive equations for single-indentation unloading and development of plasticity due to repeated indentation,” Mech Mater, Vol. 76, pp. 93–101, 2014.
[4] L. Min, C. Wei-Min, U. Demirler, L. Kayali, W. Ling-Dong, “A numerical study of indentation using indenters of different geometry,” J Mater Res, Vol. 19, pp. 73–78, 2014.
[5] X. Hernot, C. Moussa, O. Bartier, “Study of the concept of representative strain and constraint factor introduced by Vickers indentation,” Mech Mater, Vol. 68, pp. 1–14, 2014.
[6] A. Karimzadeh, M.R. Ayatollahi, M. Alizadeh, “Finite element simulation of nano-indentation experiment on aluminum 1100,” Comput Mater Sci, Vol. 81, pp. 595–600, 2014.
[7] G. H. Michler, F. J. Balta-Calleja, Mechanical properties of polymers based on nanostructure and morphology. – CRC Press, London, 2016.
[8] ISO6507-1, Metallic materials – Vickers hardness test.
[9] R. H. Myers, D. C. Montgomery, Response surface methodology: Process and product optimisation using designed experiments. – John Wiley & Sons, New York, 1976. – 714 p.
[10] J . Auzins, A. Janushevskis, J. Janushevskis, E. Skukis, “Software EdaOpt for experimental design, analysis and multiojective robust optimization” 2014 Proc. OPT-i Int. Conf. Engineering and Applied Science Optimization, pp 101-123