# МЕХАНИКА
# MECHANICS

I.G. Dobrotvor[1], D.P. Stukhlyak[1], A.V. Buketov[2],
A.G Mykytyshyn[1], R.Z. Zolotyi[1], O.V. Totosko[1]

[1]*Ivan Puluj Ternopil National Technical University, Ukraine;*
[2]*Kherson State Maritime Academy, Ukraine*
*(E-mail: zolotyy@gmail.com)*

# Automation research of thermal and physical characteristics of particulate-filled epoxy composites

Thermal-physical characteristics of filled epoxy composites were investigated by means of the developed device which allowed to do computer-aided research within defined temperature range. Forward and reverse run of relative linear elongation factor of the sample $\epsilon(T,q)=\Delta l/l$ at heating rate 1,5-2 K/min. and cooling rate 2,5-3 K/min. for the composites with particulate filler has been studied. Laplacian operator to scalar field *eu* and *ed* was used, which enables to derive stationarity or quasi-stationarity 2D areas of relaxation of macromolecular stress. Automation research will allow to determine the parameters of temperatures and filler content corresponding to minimal stress values. In the first approximation these are the following areas: $30<q<50$, $385<T<395$ K and $60<q<90$, $340<T<380$ K. Relaxation processes at the above-mentioned parameters are the most intensive in the composite. Such composites have the lowest residual stress.

*Keywords:* composite, epoxy matrix, thermal coefficient of linear expansion, relaxation processes, automation research.

*Introduction*

Nowadays, composite polymer materials are used not only as constructional materials but as coatings of various functions as well [1]. For the latter an important indicator of operational characteristics of composition coatings is thermal coefficient of linear expansion (TCLE). In most epoxy binders-based materials particulate fillers of various nature, shape and size are used [2]. Mechanism of particulate fillers influence on heat characteristics of polymer composites hasn't been investigated properly. A particulate filler when being put into epoxy composite causes the structural change due to the external surface layer formation between the filler hard surface and the binder [3]. It results in change of thermal-physical characteristics of epoxy binders-based heterogeneous systems, including thermal coefficient of linear expansion, thermal resistance and heat conductivity. While the material is being formed the filler hard surface (it depends how active it is with the binder) makes a big difference in structure and volume of external surface layers. When a composite material is being heated the particulate filler restrains its expansion that is important in operation under thermal cyclic load conditions. The relationship between physical nature of a filler in a composite material and TCLE value has been proved [4]. Taking into consideration the difference in thermal-physical characteristics of matrix and filler and activity coefficient of particulate fillers, it's possible to control the whole heat- physical characteristics of composites to change the volume and structure of surface layers. According to modern beliefs of some authors, boundary inlays may redistribute the stress in the system «matrix-filler» [5, 6]. Stress state formation of meta-polymer systems on

the phase division boundary both during and after the composite formation affects heat- physical characteristics of the material. High heat load in such layers may cause micro cracks in the material [5]. While using particles of the same dispersion the probability of cracks spread under performance conditions is increasing. Stress on the crack top can be lower due to plasticizers use. As we know, matrix-filler relation increase provides the improvement of physical-mechanical materials, including the case when dispersed magnetic fillers are added [6].

Thus, reasonable choice of plasticizers and structurally active filler (ferromagnetic, paramagnetic, diamagnetic materials) is one of the most efficient ways to solve the problem of properties of polymer composite materials being used as coatings. The use of epoxy binders of high adhesive strength to metal base, physical-mechanical characteristics and workability at coatings forming on long surfaces of complex contour is a promising direction for the development of epoxy composites of various functions operating under thermal scraping conditions.

*Results and discussion*

Low molecular epoxide resin ЕД-16 (ГОСТ 10687-76) was used as a binder for polymer composites formation. Aliphatic resin ДЕГ-1 (ТУ 6-05-1645-73) was added into the binder as a plasticizer to improve physical-chemical and technological properties of the matrix. Polyethylene polyamine (ТУ 6-02-594-73) was used as a hardener enabling to form the material at room temperature. Dispersed ferromagnetic (red mud), paramagnetic (TiC) and diamagnetic ($Al_2O_3$) powders were used at polymer systems filling. Polymer composites formation was taking place at temperature $293\pm2$ K for 24 hours followed by further thermal processing at $433\pm2$ K for 2 hours. After that the samples were kept for 48 hours at temperature $293\pm2$ K. Then the study of thermal properties of composite materials was conducted.

Dilatometer investigation of polymer composites has demonstrated that TCLE value of composite material depends on the filler concentration and nature. It was determined that at temperature rise the relaxation of residual stress in materials is taking place due to the change of macromolecules conformation set in the composite during surface layers' formation near the filler hard surface. Relaxation behavior of the composite at thermal expansion corresponds to hysteresis form at heating and cooling cycle. In this way redistribution of internal stress has been determined which takes place in the system «epoxide matrix-particulate filler» that is in accord with the results of work [7, 8].

TCLE was studied by means of the developed device which allowed to do computer-aided research within defined temperature range (Fig. 1). Thermal-physical characteristics of epoxy composites were determined according to the sample length change at temperature variation under stationary conditions (GOST 15173-70). The device consists of heater (1). In the heating area of the sample (2) the temperature was regulated by temperature controller (3), which provided the defined heating rate. The sample stretching was recorded by motion sensor (4). The temperature in the heater was measured by thermocouple (5) and was sent by analog-to-number converter (ANC) to PC (7). Heating control signal was transmitted to the thermal regulator by input/output port (8). The operation of the whole input/output board of discrete signals (9) was synchronized by the timer (10).
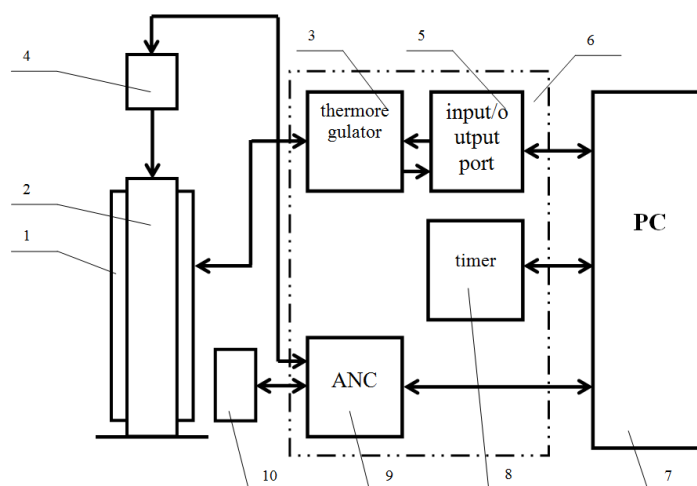


*1 – heater; 2 – sample; 3 – sample stretching meter; 4 – displacement transducer; 5 – thermocouple; 6 – analog-to-number converter; 7 – personal computer; 8 – input/output port; 9 – timer; 10 – input/output board of discrete signals*

Figure 1. Device design for the TCLE samples study

According to modern views on physics and chemistry of polymers, active dispersed particles adding whilst epoxy composites formation resulted in rigid surface layers building on the phase boundary. The parameters of such layers depend on the number of physical nodes, i.e. physical joining level [9, 10]. If temperature rises higher the vitrification temperature, physical nodes are ruined [11]. It should be admitted that in this case macromolecules which are near the hard surface of the filler may form migrating physical nodes. Flexibility increase of the latter takes place due to the change of macromolecules conformation set of the binder. According to Sperling research the above-mentioned nodes failure has been observed between transversal links of macromolecular chains. Such nodes are restored when being cooled at the temperature lower vitrification matrix temperature. It should be noted that material generation is taking place thanks to physical joints formation with catalytically-active centers on the dispersed fillers surface due to macromolecules recombination in the binder material. Thus, polymer cross-linking yield is increasing [12].

Forward and reverse run of relative linear elongation factor of the sample $\epsilon(T,q)=\Delta l/l$ at heating rate 1,5-2 $\square$K/min and cooling rate 2,5-3 $\square$K/min for the composites with particulate filler TiC has been studied. Time variation of temperature T most probably can be considered steady (quasi-steady). That's why variable T is time like. Factor q is a priori considered the one that is subjected to steady trend, in case of dispersions equal distribution on the matrix volume.

Relaxation behavior of thermal expansion was observed at continuous cyclic heating and cooling of composite material. As a result of discrete measurements the table dependencies of relative linear elongation of the sample were observed (see Table). Such dependencies in reality due to well-known statements of mathematical analysis are of two-dimensional continuous smooth character for the set of all possible degrees of composite q filling with dispersions (Fig. 2). For each one-dimensional cross-section for q=const they are in the form of hysteresis loops. An irreversible component of the sample relative elongation is determined to be available. The reason of this phenomena is viscoelasticity of composites causing internal residual stress in the system which avoided relaxation due to their significant deviations from steady features of transient processes at composites formation.

Table

**Dilatometer investigation of forward (↗) and reversed (↙) run of the samples
relative elongation of composites filled with titanium carbide dispersions
(parameter q is a value of filling per 100 resin mass fraction)**

| q=30 mass fraction | | | | q=50 mass fraction | | | |
|---|---|---|---|---|---|---|---|
| $T^0K$↗ | $\varepsilon$↗ | $T^0K$↙ | $\varepsilon$↙ | $T^0K$↗ | $\varepsilon$↗ | $T^0K$↙ | $\varepsilon$↙ |
| 317 | 0,05 | 397 | 0,95 | 323 | 0,12 | 423 | 0,94 |
| 324 | 0,18 | 386 | 0,68 | 333 | 0,20 | 410 | 0,83 |
| 327 | 0,44 | 378 | 0,54 | 343 | 0,24 | 397 | 0,75 |
| 335 | 0,39 | 370 | 0,39 | 352 | 0,27 | 386 | 0,64 |
| 346 | 0,52 | 352 | 0,30 | 366 | 0,30 | 375 | 0,52 |
| 360 | 0,70 | 340 | 0,27 | 381 | 0,39 | 359 | 0,34 |
| 366 | 0,81 | 327 | 0,24 | 395 | 0,54 | 352 | 0,27 |
| 377 | 0,90 | 314 | 0,20 | 407 | 0,68 | 344 | 0,15 |
| 396 | 0,95 | 308 | 0,12 | 423 | 0,94 | 336 | 0,02 |

| q=80 mass fraction | | | | q=100 mass fraction | | | |
|---|---|---|---|---|---|---|---|
| $T^0K$↗ | $\varepsilon$↗ | $T^0K$↙ | $\varepsilon$↙ | $T^0K$↗ | $\varepsilon$↗ | $T^0K$↙ | $\varepsilon$↙ |
| 347 | 0,01 | 423 | 0,67 | 353 | 0,01 | 423 | 0,60 |
| 354 | 0,13 | 416 | 0,56 | 362 | 0,14 | 418 | 0,52 |
| 357 | 0,17 | 408 | 0,42 | 370 | 0,22 | 413 | 0,45 |
| 365 | 0,30 | 399 | 0,32 | 377 | 0,29 | 406 | 0,32 |
| 376 | 0,43 | 381 | 0,24 | 381 | 0,31 | 397 | 0,23 |
| 389 | 0,51 | 370 | 0,21 | 393 | 0,40 | 382 | 0,16 |
| 396 | 0,55 | 357 | 0,17 | 400 | 0,45 | 363 | 0,14 |
| 408 | 0,61 | 344 | 0,12 | 415 | 0,54 | 347 | 0,11 |
| 423 | 0,67 | 324 | 0,06 | 423 | 0,60 | 324 | 0,08 |

Sloping zones of trend response (Fig. 2) of two-dimensional dependencies graph $\epsilon$(T,q) change indicates the stationarity areas of residual stress relation. The necessary condition of extreme value of the function f=A(x, y) first derivative in a random point is that the second derivative is equal to zero in the same point, in this case the second derivative must have different signs on the opposite sides of the point.

For the two-dimensional option the analogue of the second derivative A(x, y) is Laplacian (1) — scalar operator like

$$\nabla^2(f) = \frac{\partial^2 A(x,y)}{\partial^2 + x^2} + \frac{\partial^2 A(x,y)}{\partial^2 + y^2}. \tag{1}$$

We use Laplacian operator to scalar field eu and ed (Fig. 2), which enables to derive stationarity or quasi-stationarity 2D areas [13], which are characterized by constant trend (mathematical expectation) and deviation (dispersion). Hysteresis dependence characterizes the rate of relaxation of macromolecular stress [14, 15]. This relaxation rate provides the residual stress minimization. The most optimal process of the composite hardening must have some features of stationarity to provide the least residual internal stress. Using numerical calculation methods of Laplacian operator (1) (listing 1) for discrete fields of measurements eu and ed of tests results (Fig. 2), we'll pay attention to the areas where the values of calculation are close to zero (Fig. 3). We must admit that the calculated field of Laplacian values lu and ld is smaller than the initial field due to specific discrete calculations. We build the field of vector product lu and ld (Fig. 3) to guarantee the stationarity of forward and reverse run. The obtained results prove the relaxation processes stationarity of forward and reverse run of dilatometer measurements for the composites filled with titanium carbide dispersions [16, 17]. Non stationary areas are found to be the ones close to those which satisfy the ratio: {340<T<350, 30<q<55} та {400<T<420, 30<q<40}.
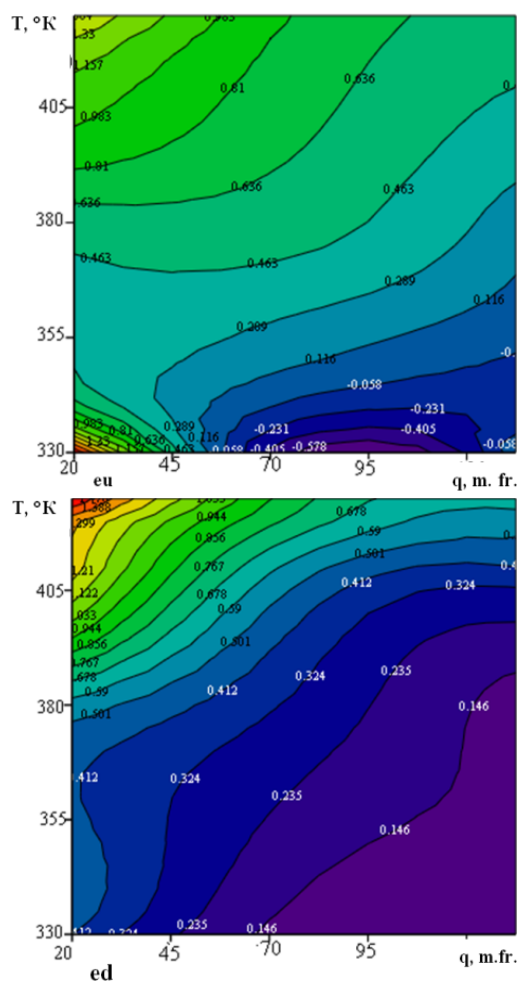


Figure 2. Two-dimensional dependencies graph $\epsilon$(T,q) for forward (eu) and
reverse (ed) run of dilatometer measurements of relative linear elongation of the sample
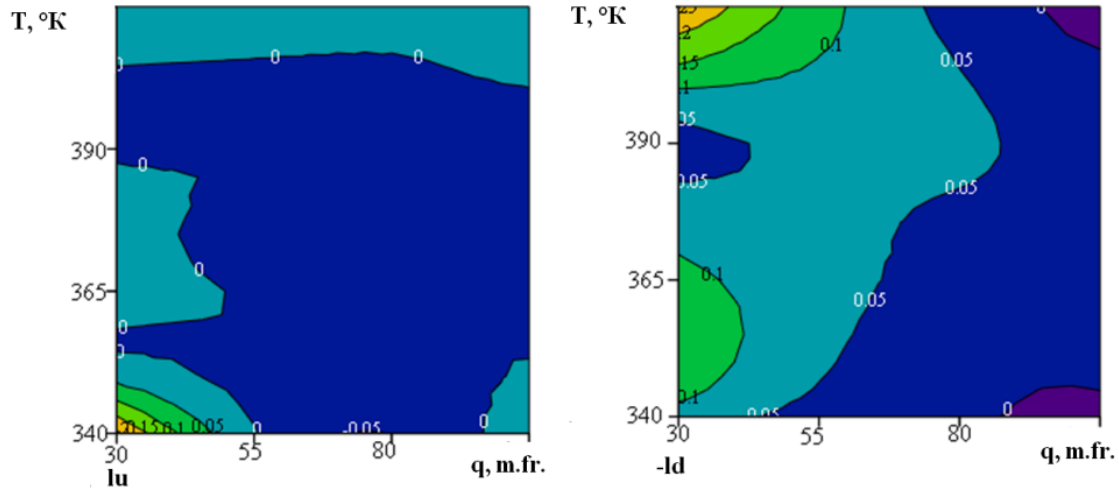
Figure 3. Result of Laplacian operator to the fields of forward (lu) and
reverse (ld) run of dilatometer investigation

Listing 1. Calculation with the help of MathCAD-14 program of two-dimensional Laplacian operator for the field of dilatometer measurements of dependencies $\epsilon$(T,q) for forward (eu) and reverse (ed) run:

$$j := 1..18 \quad i := 1..18$$

$$edq_{i,j} := 0.25 \cdot (ed_{i+1,j+1} - ed_{i-1,j+1} + ed_{i+1,j-1} - ed_{i-1,j-1});$$

$$edt_{i,j} = 0.25 \cdot (ed_{i+1,j+1} - ed_{i+1,j-1} + ed_{i-1,j+1} - ed_{i-1,j-1});$$

$$euq_{i,j} = 0.25 \cdot (eu_{i+1,j+1} - eu_{i-1,j+1} + eu_{i+1,j-1} - eu_{i-1,j-1});$$

$$eut_{i,j} = 0.25 \cdot (eu_{i+1,j+1} - eu_{i+1,j-1} + eu_{i-1,j+1} - eu_{i-1,j-1});$$

$$cols(euq) = 19 \quad rows(euq) = 19;$$

$$edq := submatrix(edq, 1, 18, 1, 18) \quad edt := submatrix(edt, 1, 18, 1, 18);$$

$$euq := submatrix(euq, 1, 18, 1, 18) \quad eut := submatrix(eut, 1, 18, 1, 18);$$

$$cols(euq) = 18 \quad rows(euq) = 18;$$

$$j := 1..16 \quad i := 1..16;$$

$$euq2_{i,j} = 0.25 \cdot (euq_{i+1,j+1} - euq_{i-1,j+1} + euq_{i+1,j-1} - euq_{i-1,j-1});$$

$$eut2_{i,j} = 0.25 \cdot (eut_{i+1,j+1} - eut_{i+1,j-1} + eut_{i-1,j+1} - eut_{i-1,j-1});$$

$$euq2_{i,j} = 0.25 \cdot (euq_{i+1,j+1} - euq_{i-1,j+1} + euq_{i+1,j-1} - euq_{i-1,j-1});$$

$$edt2_{i,j} = 0.25 \cdot (ed_{i+1,j+1} - ed_{i+1,j-1} + ed_{i-1,j+1} - ed_{i-1,j-1});$$

$$cols(euq2) = 17 \quad rows(euq2) = 17;$$

$$euq2 := submatrix(euq\,2, 1, 16, 1, 16) \quad eut2 := submatrix(eut\,2, 1, 16, 1, 16);$$

$$edq2 := submatrix(edq\,2, 1, 16, 1, 16) \quad edt2 := submatrix(edt\,2, 1, 16, 1, 16);$$

$$cols(euq2) = 16 \quad rows(euq2) = 16;$$

$$lu := euq2 + eut2 \quad ld := edq2 + edt2.$$

By means of the program MathCAD-14 we determine the stationarity fields of hysteresis loops passing (forward and reverse) forming the operators of vector product lu*ld for sampling investigation (at least one of them) and the sum of simultaneous study (the both). In the sum lu-ld the sign «-» appears due to the temperature countdown (Fig. 4).
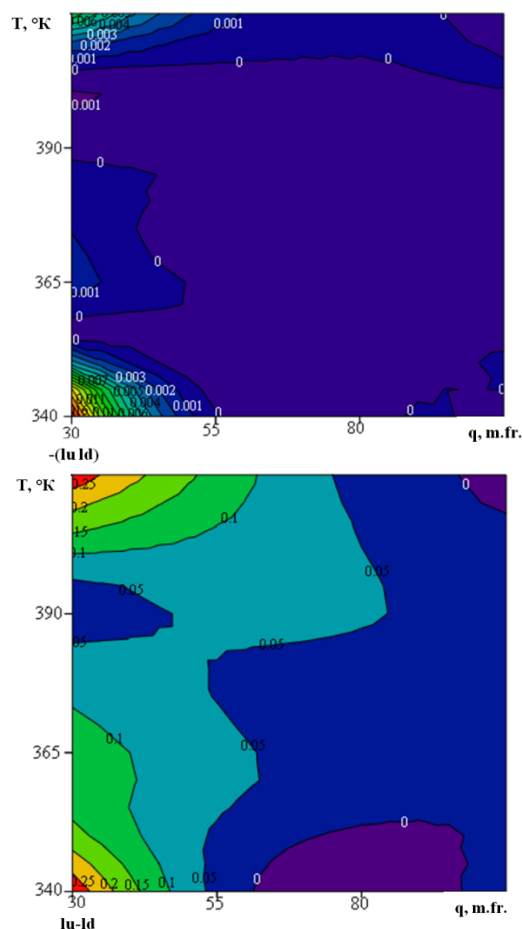
Figure 4. Fields of characteristics of sampling lu ld and simultaneous lu-ld stationarity performance of forward and reverse run of relaxation processes of epoxy composite filled with titanium carbide

*Conclusions*

Automation research will allow to determine the parameters of temperatures and filler content corresponding to minimal stress values. In the first approximation these are the following areas: $30<q<50$, $385<T<395$ K та $60<q<90$, $340<T<380$ K. Relaxation processes at the above-mentioned parameters are the most intensive in the composite. Such composites have the lowest residual stress. To make more accurate recommendations one must turn to the theory of higher orders operators and the theory of pattern recognition that will be the matter of further research [18]. The research method of composites dilatometer properties under discussion can be used for another composition of composite materials.

References

1 Стухляк П.Д. Эпоксидные композиты для защитных покрытий / П.Д. Стухляк. — Тернопіль: Збруч, 1994. — 177 с.

2 Кац Г.С. Наполнители для полимерных композиционнных материалов / Г.С. Кац, Д.В. Милевский. — М.: Химия, 1981. — 736 с.

3 Dobrotvor I.G. Investigation of the formation of external surface layers in epoxy composites / I.G. Dobrotvor, D.P. Stukhlyak, A.V. Buketov // Materials Science. — 2009. — Vol. 45. — No. 4. — P. 582–588.

4 Teraoka I. Polymer solutions. An Introduction to Physical Properties / I. Teraoka. — N.Y.: Wliey – Interscience, 2002. — 332 p.

5 Buketov A. Investigation of thermophysical properties of epoxy nanocomposites / Ё.Buketov, P. Ma-ruschak, O. Sapronov, M. Brailo, O. Leshchenko, L. Bencheikh, A. Menou // Molecular Crystals and Liquid Crystals. — 2016. — Vol. 628. — P. 167–179.

6 Stukhlyak P. Influence of ultraviolet radiation on the properties of epoxy composites / P. Stukhlyak, A. Buketov, I. Dobrotvor, N. Dolgov // 9th International symposium of Croatian metallurgical society Material and metallurgy, Summaries of lecture, Sibenic, Croatia, June, 20-24. — 2010. — P. 237.

7 Rohwer K. Application of higher order theories to the bending analysis of layered composite shells / K. Rohwer // Iut. J. Solids and Struct. — 1992. — Vol. 28. — No. 1. — P. 105–119.

8 Стухляк П.Д. Дослідження впливу природи наповнювачів і товщини покриттів на зміну градіэнта кольорів на внутрішні напруження в епоксикомпозитах / П.Д. Стухляк, І.Г. Добротвор, А.В. Букетов, Р.З. Золотий // Вісник Київського національного університету технологій та дизайну. — 2006. — № 5(31). — С. 82–87.

9 Кунце Х.-И. Методы физических измерений / Х.-И. Кунце. — М.: Мир, 1989. — 216 с.

10 Саченко А.А. Специализированная информационно-измерительная система на базисе микро-ЭВМ / А.А. Саченко, И.Г. Добротвор, А.Ф. Карачка, М.И. Чирка // Электронная техника. Сер. 8, Управление качеством, стандартизация, метрология, испытания. — 1987. — Вып. 2(125). — С. 127–132.

11 Мэнмон Дж. Полимерные смеси и композиты / Дж. Мэнмон, Л. Сперлинг. — М.: Химия, 1979. — 440 с.

12 Handbook of composites. G. Lubin van Nostrand Reinhold Company inc. — N.Y., 1982. — 582 p.

13 Саченко А.А., Добротвор И.Г. Выделение квазистационарных интервалов нестационарных временных процессов // Применение вычислительной техники и математики в научных исследованиях: тез. докл. науч.-техн. конф. — Киев, 1985. — С. 66, 67.

14 Dierk Raabe. Computational Materials Science: The Simulation of Materials Microstructures and Properties. — N.Y.: WileyVCH, 1998. — 380 p.

15 Stukhlyak P. Investigation of the phenomena revealed on phase interface in epoxide-composites / P. Stukhlyak, I. Dobrotvor, M. Mytnyk, M. Mykytyshyn // Przetworstwo tworzyw. Polymer processing. — 2017 (styczen-luty). — No. 1(175)/23. — P. 53–63.

16 Freund L. Thin Film Materials. Stress, Defect, Formation / L. Freund, S. Suresh. — N.Y.: Cambridge University Press, 2003. — 800 p.

17 Stukhlyak P. Epoxycomposite materials modified with an ultraviolet irradiation / P. Stukhlyak, A. Buketov. — Ternopil: Zbruch, 2009. —237 p.

18 Buketov A.V. A study of creep of epoxy composites with continuous fibers and modified fine filler in aggressive media / A. Buketov, P. Stukhlyak, V. Levyts'kyi, I. Dobrotvor, N. Dolgov // Strength of Materials. — Vol. 43. — No. 3. — May, 2011. — P. 338–350.

И.Г. Добротвор, Д.П. Стухляк, А.В. Букетов,
А.Г. Микитишин, Р.З. Золотий, О.В. Тотоско

# Дисперсті толтырылған эпоксикомпозиттердің жылу-физикалық сипаттамаларын зерттеуді автоматизациялау

Автоматтандырылған режимде берілген температуралардың диапазонында зерттеулер жүргізуге мүмкіндік беретін, әзірленген құрылғы көмегімен толтырылған эпоксикомпозиттердің жылу-физикалық сипаттамалары зерттелген. Дисперсті толтырылған композиттер үшін (T, q) = $\Delta 1/1$ салыстырмалы сызықтық ұзарту үлгісінің 1,5-2 К/мин қызу және салқындату жылдамдықтарымен 2,5-3 К/мин тікелей және кері әрекетінің тәуелділігі қарастырылған. Лаплас операторы мен $eu$ және $ed$ скаляр өрісін қолдану арқылы макромолекулалық кернеулердің релаксациялық процестерінің стационарлы немесе квазистационарлық аймақтарын бөлуге болады. Зерттеу автоматтандыру кернеудің минималды мәндеріне сәйкес келетін температураның және толтырғыштың параметрлерін анықтауға мүкіндік береді. Бірінші жуықтауда бұл жолақтар $30 < q < 50$, $385 < T < 395$ K және $60 < q < 90$,

$340 < T < 380$ К. Релаксация процестері осы көрсеткіштерде композитте қарқынды жүреді. Мұндай композиттер төмен қалдықты кернеулі болады.

*Кілм сөздер:* композит, эпоксидті матрица, сызықтық кеңейтудің термалды коэффициенті, зерттеуді автоматизациялау.

И.Г. Добротвор, Д.П. Стухляк, А.В. Букетов,
А.Г. Микитишин, Р.З. Золотий, О.В. Тотоско

# Автоматизация исследований теплофизических характеристик дисперсно-наполненных эпоксикомпозитов

Исследованы теплофизические характеристики наполненных эпоксикомпозитов с помощью разработанного устройства, которое позволяет в автоматизированном режиме проводить исследования в заданном диапазоне температур. Изучены зависимости прямого и обратного хода показателя относительного линейного удлинения образца $\epsilon$ (T, q) = $\Delta l$ / l со скоростью нагрева 1,5–2 К/мин и скоростью охлаждения 2,5–3 К/мин для композитов с дисперсным наполнителем. Использование оператора Лапласа и скалярного поля *eu* и *ed* позволило выделить плоские области стационарности или квазистационарности процессов релаксации макромолекулярных напряжений. Автоматизация исследований позволит определить параметры температур и содержания наполнителя, соответствующие минимальным значениям напряжения. В первом приближении это зоны 30 <q <50, 385 <T <395 К и 60 <q <90, 340 <T < 380 К. Релаксационные процессы при данных параметрах проходят в композите наиболее интенсивно. Такие композиты имеют низкие остаточные напряжения.

*Ключевые слова:* композит, эпоксидная матрица, термический коэффициент линейного расширения, автоматизация исследований.

## References

1 Stukhlyak, P. (1994). *Epoksidnye kompozity dlia zashchitnykh pokrytii [Epoxy composites for protective coatings]*. Ternopil: Zbruch [in Russian].

2 Katz, G.S., & Milevsky, D.V. (1981). *Napolniteli dlia polimernykh kompozitsionnnykh materialov [Fillers for polymeric composite materials]*. Moscow: Khimiia [in Russian].

3 Dobrotvor, I.G., Stukhlyak, P.D., & Buketov, A.V. (2009). Investigation of the formation of external surface layers in epoxy composites. *Materials Science, Vol. 45,* 4, 582–588.

4 Teraoka, I. (2002). Polymer solutions. An Introduction to Physical Properties. N.Y.: Wliey – Interscience.

5 Buketov, A., Maruschak, P., Sapronov, O., Brailo, M., Leshchenko, O., Bencheikh, L., & Menou, A. (2016). Investigation of thermophysical properties of epoxy nanocomposites. *Molecular Crystals and Liquid Crystals, Vol. 628,* 167–179.

6 Stukhlyak, P., Buketov, A., Dobrotvor, I., & Dolgov, N. (2010). Influence of ultraviolet radiation on the properties of epoxy composites. 9th International symposium of Croatian metallurgical society Material and metallurgy, Summaries of lecture, Sibenic, Croatia, June, 20-24, 237.

7 Rohwer, K. (1992). Application of higher order theories to the bending analysis of layered composite shells. *Iut. J. Solids and Struct, Vol. 28,* 1, 105–119.

8 Stukhlyak, P.D., Dobrotvor, I.Г., Buketov, A.V., & Zolotiy, R.Z. (2006). Doslidzhennia vplivu prirodi napovniuvachiv i tovshchini pokrittiv na zminu hradienta koloriv na vnutrishni napruzhennia v epoksikompozitakh [Investigating the Influence of the Nature of Fillers and the thickness of the coatings to change the gradient of colors on the internal stresses in epoxy composites]. *Visnik Kiivskoho natsionalnoho universiteta tekhnolohii ta dizaina – Bulletin of the Kiyv National University of Technology and Design, 5(31),* 82–87 [in Ukrainian].

9 Kunze, Kh.-I. (1989). Metody fizicheskikh izmerenii [Methods of physical measurements]. Moscow: Mir [in Russian].

10 Sachenko, A.A., Dobrotvor, I.G., Karachka, A.F., & Chirka, M.I. (1987). Spetsializirovannaia informatsionno-izmeritelnaia sistema na bazise mikro-EVM [Specialized information-measuring system based on a micro-computer]. *Elektronnaia tekhnika. Seriia 8, Upravlenie kachestvom, standartizatsiia, metrolohiia, ispytaniia – Electronic equipment. Ser. 8, Quality management, standardization, metrology, tests, Issue 2(125),* 127–132 [in Russian].

11 Menmon, Dzh., & Sperling, L. (1979). *Polimernye smesi i kompozity [Polymer mixtures and composites].* Moscow: Khimiia [in Russian].

12 Handbook of composites. G. Lubin van Nostrand Reinhold Company inc. N.Y. (1982).

13 Sachenko, A.A., & Dobrotvor, I.G. (1985). Vydelenie kvazistatsionarnykh intervalov nestatsionarnykh vremennykh protsessov [Isolation of quasistationary intervals of nonstationary time processes]. Application of computer technology and mathematics in scientific research: *tezisy doklady nauchno-tekhnicheskoi konferentsii – theses reports of the scientific and technical conference.* (pp. 66, 67). Kiev [in Russian].

14 Dierk Raabe. (1998). Computational Materials Science: The Simulation of Materials Microstructures and Properties. N.Y.: WileyVCH.

15 Stukhlyak, P., Dobrotvor, I., Mytnyk, M., & Mykytyshyn, M. (2017). Investigation of the phenomena revealed on phase interface in epoxide-composites. *Przetworstwo tworzyw. Polymer processing, (styczen-luty), 1(175)/23,* 53–63.

16 Freund, L., & Suresh, S. (2003). *Thin Film Materials. Stress, Defect, Formation.* N.Y.: Cambridge University Press.

17 Stukhlyak, P., & Buketov, A. (2009). *Epoxycomposite materials modified with an ultraviolet irradiation.* Ternopil: Zbruch.

18 Buketov, A.V., Stukhlyak, V., Levyts'kyi, I., & Dobrotvor, N. (2011). Dolgov A study of creep of epoxy composites with continuous fibers and modified fine filler in aggressive media. S*trength of Materials, Vol. 43,* 3, May, 338–350.