

Chapter 25

ULTRASONIC TREATMENT IN THE PRODUCTION OF CLASSICAL COMPOSITES AND CARBON NANOCOMPOSITES

**Aleksandr Evhenovych Kolosov¹, Elena Petryvna Kolosova¹,
Volodymyr Volodymyrovych Vanin¹ and Anish Khan²**

¹Aleksandr Evhenovych Kolosov
Chemical, Polymeric and Silicate Machine Building Department of Chemical Engineering Faculty
National Technical University of Ukraine «Igor Sikorsky Kyiv Polytechnic Institute», Kyiv, Ukraine
19 Build., 37 Prospect Peremohy, 03056, Kyiv, Ukraine
e-mail: a-kolosov@ukr.net, www.kolosov.ua

¹Elena Petryvna Kolosova
Faculty of Physics and Mathematics
National Technical University of Ukraine «Igor Sikorsky Kyiv Polytechnic Institute», Kyiv, Ukraine
7 Build., 37 Prospect Peremohy, 03056, Kyiv, Ukraine
e-mail: mrselkolosova@gmail.com

¹Volodymyr Volodymyrovych Vanin
Faculty of Physics and Mathematics
National Technical University of Ukraine «Igor Sikorsky Kyiv Polytechnic Institute», Kyiv, Ukraine
7 Build., 37 Prospect Peremohy, 03056, Kyiv, Ukraine
e-mail: fmf@kpi.ua

²Anish Khan
Chemistry Department Faculty of Science
Center of Excellence for Advanced Materials Research
King Abdulaziz University, Jeddah 21589, P.O. Box 80203, Saudi Arabia
email: akrkhan@kau.edu.sa

Abstract

Low-frequency ultrasonic treatment in the production of classical composites and carbon nanocomposites is one of the dominant methods used in the preparation of such kinds of composites. For example, ultrasonic is a basic method of physical modification of liquid epoxy media, as well as to a method that intensifies the processes of sonification of liquid epoxy media, capillary impregnation, "wet" winding and dosed application. This method also promotes the production of defect-free and monolithic structures of such reinforced composites. The influence of ultrasonic treatment regimes on the technological and operational properties of epoxy polymers is investigated, as well as the strengthening of reinforced composites based on them. Technical means of ultrasonic cavitation treatment for classical epoxy binders and polymer composite materials based on them are investigated. Ultrasonic dispersing of nanoparticles in solutions and liquid polymeric media is studied. Aspects of preparation of nanosuspensions for the production of polymeric nanocomposites with ultrasonic treatment as also the production of nanomodified epoxy compositions and prepregs based on them are described. Ultrasonic treatment in the production of graphene and graphene aerogels is analyzed. Epoxy composites based on graphene aerogels with exceptional operational properties are studied.

Keywords: Thermoset, Epoxy, Composite, Classical, Nanomodified, Carbon, Graphene, Aerogel, Technology, Ultrasonic, Treatment

1. Introduction – Prerequisites for the Application of Ultrasonic Treatment in the Producing of Classical Composites and Carbon Nanocomposites

At present, in addition to the widespread use of classical polymer composite materials (PCMs) based on well-studied epoxy matrices [1], innovative world technologies for the creation of a new generation of PCMs are intensively developing. These studies reach the nanoscale molecular level

of research into the components that make up PCMs [2]. In connection with this, one of the promising directions in modern science is the production of composites based on polymers filled with carbon nanomaterials, such as carbon nanotubes (CNTs), fibrils, graphene, graphene aerogels (GA), nanoplates, nanofibers, etc. [3].

It is therefore not surprising that many economically developed countries, as well as developing countries, in the next 20 years, associate the further economic growth of their countries with the orientation of industrial sectors to the production and use of precisely nano-structured materials and nanocomposites [4].

Recently, studies related to the use of nanosystems are developing quite actively. To date, three main branches have been outlined in this area of research: 1) from nano- to macrocomposites (obtaining binders, materials from melts, etc. by means of substance synthesis); 2) from macro- to nanosubstances (due to disintegration), and, 3) by using various nanomaterials (nanotubes, fullerenes, etc.) as microadditives for compositional binders [5].

For example, nanotechnology is increasingly used in various technological processes in the chemical and other industries, in particular, in chemical and oil and gas engineering. Nanocoatings have broad prospects for increasing the efficiency of the oil and gas complex. The latter is used for hydrophobizing surfaces of a wide range of structures and equipment. For example, these are antistatic coatings for fuel pipelines, as well as for various structural metal products. This operation is carried out in order to provide these structural elements with chemical resistance, water repelling and antifriction properties for a long time of operation.

Another area of application of nanotechnology in chemical engineering is the preparation of polymer compositions that strengthen stress concentration zones (in the form of holes, cutouts, fillets, thickness differences, etc.) in various power structures. No less effective is the use of such nanomodified polymer compositions for "healing" defects, microcracks and other damages that occur during the manufacture and operation of such structures, as well as to eliminate and seal gaps in the holes and joints of bolted and riveted joints.

In addition to the manufacture of multifunctional nano-structured coatings and compositions, nanotechnologies have the prospect of using in improving the manufacturing technology of various high-strength and corrosion-resistant body and structural elements based on reinforced thermoplastic PCMs, in particular, carbon plastics [6]. Moreover, an increasing number of researchers come to the conclusion that the most promising method for improving the properties of reactoplastic PCMs is their modification by carbon nanostructured components, including CNTs. The latter possesses a number of unique properties that allow solving the problems arising in the production of nanomodified (NM) reactoplastic PCMs, that is, NM PCMs.

Therefore, in contrast to classical PCMs, the physico-chemical properties and technological aspects of the preparation of which have been studied comparatively comprehensively, NM PCMs are increasingly being considered as their real alternative. This is mainly due to the increased rigidity and strength, as well as the electrical conductivity of NM PCMs with a small bulk content of NM fillers. Fullerenes, CNTs, diamond-like and fullerene-like structures have unique and substantially different physico-chemical properties. This allows them to be used as modifiers of polymer binders and to obtain on their basis NM PCMs with wide ranges of performance values [7].

Another trend of modern development and application of nanomaterials and derivatives based on them is the use of aerogels. The view of the huge variety of aerogels, the most interesting for further study are aerogels based on carbon nanomaterials. The latter is a class of ultralight substances in which the liquid phase (at the lattice sites) is completely replaced by a gaseous phase. That is why GA was named modern ultra-light material. He thus outstripped the air-graphite record, which for a long time retained the “palm tree” in this category.

Aerogels are characterized by a whole complex of unique properties. First of all, it is low density, high specific surface area, and high hydrophobicity index. No less important indicators of aerogels are low thermal conductivity, high elasticity (the ability to restore the shape after multiple compression and stretching), as well as the ability to sorb organic liquids. Moreover, depending on

the purposes of the application, aerogels based on carbon nanomaterials can exhibit magnetic and electrically conductive properties, while retaining the flexibility of their 3D-structure.

Therefore, it is not surprising that the impressive properties of new aerogels based on graphene are of great interest to scientists around the world. At the same time, it is urgent to find the most effective application of GA in various fields. Among the latter – environmental protection, medicine, electronics, military, biology, chemistry and many others. For example, the ability of GA to sorb organic liquids can be used to eliminate oil spills in aqueous media.

When obtaining NM PCMs, one of the main problems is to ensure high-quality wetting and uniform distribution of the nanofiller in the liquid polymer matrix. On the one hand, nanosuspension is incorporated into the composition of the polymer binder of CNTs for the subsequent fabrication of a nanocomposite on its basis, which substantially increases the strength properties of the finished (polymerized) products. Moreover, the optimal concentration and uniform distribution of CNTs in the binder play a decisive role in the final strengthening of NM PCMs.

On the other hand, due to the peculiarities of the nanoparticles used (their propensity to mutual attraction and agglomeration), problematic situations in the production of NM PCMs are de-agglomeration and further dispersion of the used nanoparticles in liquid polymer media. It is obvious that the incorporation of CNTs into the structure of the polymer composite affects not only the structure and properties of the liquid polymer binder but also the NM PCM as a whole.

For example, the dimensions of the agglomerate in nanofluids can significantly affect the thermal conductivity and viscosity of nanofluids and lead to different heat transfer characteristics. Analysis of the literature data on the elastic, strength, rheological, electrical properties of composites filled with CNTs shows that neglecting the quality of dispersion generates, in the end, a large dispersion of the service (operational) properties of nanomodified PCMs [8]. However, we have to state that at the moment there are no unambiguously and clearly formulated industrial-technological principles concerning the incorporation, distribution, and stabilization of CNTs dispersions in NM PCMs.

It is known that low-frequency ultrasonic (US) is one of the most common methods of decomposition of CNTs agglomerates [9]. In addition, its use facilitates the dispersion of nanoparticles in base liquids in the preparation of nano-based liquids, both on the basis of organic solutions and on the basis of liquid polymers. It is also known that the synthesis of aerogels based on graphene is promising the formation of a three-dimensional structure under the influence of low-frequency US. Numerous studies have found that the use of US contributes to the intensification of the basic operations of the technological process of producing classical PCMs [10].

This, in turn, leads to an improvement in the operational characteristics of the sonicated classical PCMs and NM PCMs.

Among them – sonication (sounding) of the polymeric binders (PBs), impregnation, winding, dosed application in the preparation of fibrous prepregs. Also, positive results of using the optimal modes of US treatment are: reduction of the time of hardening of composites and obtaining of defect-free PCM structures.

The above confirms once again that the study of effective methods and the determination of the degree of their influence on the qualitative parameters of the final polymer product, as well as the development of hardware-technological schemes for obtaining classical PCMs and NM PCMs is an urgent and priority area of modern research. In turn, the new technological methods developed as a result of the research will undoubtedly find wide application in the production of structural and functional both classical PCMs and NM PCMs. For such materials, reducing the mass of the product, due to the improvement of their physico-mechanical and operational characteristics, is an urgent task for resource and energy saving on an industrial scale [11].

Thus, the aforementioned brief analysis of the different aspects of molding of US treatment in the production of classical PCMs and NM PCMs brings out the actual directions of investigations. The following directions can be identified:

- technical means of US cavitation treatment for classical epoxy oligomers (EOs), epoxy compositions (ECs), epoxy binders (EBs) and PCMs based on them;

- US dispersing of nanoparticles in solutions and liquid polymeric media;
- preparation of nanosuspensions for the production of polymeric nanocomposites with US treatment;
- US treatment in the production of graphene and graphene oxide (GO);
- epoxy composites based on GA with exceptional operational properties;
- production of NM polymer compositions and preregs based on them.

The above aspects are shortly described in this chapter.

Conclusions

The survey material in the present chapter confirms that US technology in the production of classical PCMs and carbon NM PCMs is one of the dominant methods for synthesizing new and physical modifications of existing polymer composites to improve their operational (functional) properties. And the main physical phenomenon associated with US, which has to do with the synthesis of new materials, is low-frequency acoustic cavitation. It manifests itself in the formation, growth and implosive collapse of bubbles in the liquid. This phenomenon creates extreme conditions inside the collapsing bubble and serves as the source of most sonochemical phenomena in liquids or in liquid solutions with fillers.

The main problematic situations in the production of NM PCMs is the need for dispersing (deagglomerating) nanofillers in a liquid matrix. In many cases, US is practically a non-alternative method for solving the above-mentioned problem situations. Besides, use of intense US allows for the tailoring of unique materials from sol-gel processes. This makes high-power US a powerful tool for chemistry and materials' research and development.

At the present stage, CNTs, graphene, GO and GAs are the most important carbon nanofillers for constructing functional NM PCMs. At the same time, the use of each of the above-mentioned nano-fillers in the polymer composite has its own characteristics. For example, it was found that the addition of graphene to epoxy composites leads to an increase in the rigidity and strength of the material compared to composites containing only CNTs. Graphene is better combined with an epoxy polymer, more effectively penetrating (incorporating) in the structure of the composite. Therefore, according to its unique structure and properties, graphene became a universal nano-dimensional building block material for the self-assembly of new materials with new properties and functions.

It is noted that the method of freeze-drying is more promising and quite versatile so that it can be used for the dispersion of graphene in a wide range of other composite precursors. Another

promising type of nanofillers for polymer composites, namely GAs, not only maintain the unique structural advantages of graphene sheets, but also explore outstanding properties, including lower density, excellent electrical conductivity and mechanical strength, and unusual adsorption properties.

Especially it is necessary to note the most perspective directions of application of carbon nanocomposites as functional materials. Namely – as substrates for catalysts, artificial muscles, electrodes for supercapacitors, light conductors and insulating materials, sensor batteries, sorbents (environmental protection) and gas sensors, materials for bulletproof vests, medicine and others. Also, graphene-based nanocomposites can be used in the manufacture of aircraft components, which must remain light and resistant to physical impact.

Finally, for the sake of fairness, it should nevertheless be noted that in spite of the successes achieved in the synthesis and modification of both existing classical and new NM PCMs, the basic successes in this direction so far fall to the stage of laboratory research. So when going to the industrial scale of production of such materials, among other things, it will also be necessary to take into account the so-called scale effect. That is, when the automatic transfer of the results of laboratory research to the production level, there will certainly be obstacles and "undercurrents" that can not be simulated in "ideal" laboratory conditions. Obviously, given the pace of development of modern science and technology, this is a matter of the near future.

References

1. Kuleznev VN, Gusev VK. *Fundamentals of plastics processing technology*. Moscow: Khimiya; 1995.
2. Harris PJF. Carbon nanotube composites. *Int. Mater. Rev.* 2004; 49 (1): 31–43.
3. Ray SS. *Polymer Nanocomposites and Their Applications*. California: Stevenson Ranch, American Scientific Publishers; 2006: 68–187.
4. Kablov EN, Kondrashov SV, Yurkov G Yu. Perspectives of use of carbon-containing nanoparticles in binding for polymeric composite materials. *Russian Nanotechnologies*. 2013; 8 (3–4): 24–42.
5. Kolosov AE. Preparation of Nano-Modified Reactoplast Polymer Composites. Part 1. Features of used nanotechnologies and potential application areas of nanocomposites (a review). *Chem. and Petrol. Eng.* 2015; 51 (7–8): 569–573. doi: [10.1007/s10556-015-0088-y](https://doi.org/10.1007/s10556-015-0088-y).
6. Kolosov AE. Preparation of Reactoplastic Nano-Modified Polymer Composites. Part 5. Advantages of using nano-modified structural carbon-fiber composites (a review). *Chem. and Petrol. Eng.* 2017; 52 (9–10): 721–725. doi: [10.1007/s10556-017-0259-0](https://doi.org/10.1007/s10556-017-0259-0).
7. Kolosov AE. Preparation of Reactoplastic Nanomodified Polymer Composites. Part 4. Effectiveness of modifying epoxide oligomers with carbon nanotubes (review). *Chem. and Petrol. Eng.* 2016; 52 (7–8): 573–577. doi: [10.1007/s10556-016-0235-0](https://doi.org/10.1007/s10556-016-0235-0).
8. Kolosov OE. *Preparation of Traditional and Nanomodified Reactoplastic Polymer Composite Materials* [in Ukrainian]. Kiev: VPI VPK Politehnika; 2015.
9. Kolosov AE. Preparation of Reactoplastic Nanomodified Polymer Composites. Part 3. Methods for dispersing carbon nanotubes in organic solvents and liquid polymeric media (review). *Chem. and Petrol. Eng.* 2016; 52 (1–2): 71–76. doi: [10.1007/s10556-016-0151-3](https://doi.org/10.1007/s10556-016-0151-3).
10. Kolosov AE, Sakharov AS, Sivetskii VI, Sidorov DE, Sokolskii AL. Method of selecting efficient design and operating parameters for equipment used for the ultrasonic modification of

liquid-polymer composites and fibrous fillers. *Chem. and Petrol. Eng.* 2012; 48 (7–8): 459–466. doi: [10.1007/s10556-012-9640-1](https://doi.org/10.1007/s10556-012-9640-1).

11. Kolosov AE, Kolosova EP. Functional Materials for Construction Application Based on Classical and Nano Composites: Production and Properties. In: Rita Khanna, Romina Cayumil, eds. *Recent Developments in the field of Carbon Fibers*. InTechOpen; 2018. ISBN: 978-953-51-6055-7. (in print).

12. Juan A. Gallego-Juarez, Karl F. Graff. *Power Ultrasonics: Applications of High-intensity Ultrasound*. Elsevier; 2014. 1166 p.

13. Suslick KS. Sonochemistry. In: *Kirk-Othmer Encyclopedia of Chemical Technology*. 4th ed. New York: J. Wiley & Sons; 1998: Vol. 26: 516-541.

14. Fedotkin IM, Gulyi IS. *Cavitation, Cavitation Engineering and Technology, Their Use in Industry. Part II* [in Russian]. Kiev: OKO; 2000.

15. Leighton TG. *The Acoustic Bubble*. London: Academic Press; 1994. 613 p.

16. <http://www.hielscher.com/sonochem>

17. Bang JH, Suslick KS. Applications of Ultrasound to the Synthesis of Nanostructured Materials. *Adv. Mater.* 2010; 22: 1039-1059. <https://doi.org/10.1002/adma.200904093>.

18. Kolosov AE. Low-Frequency Ultrasonic Treatment as an Effective Method for Modifying Liquid Reactoplastic Media. *Chem. and Petrol. Eng.* 2014; 50 (1–2): 79–83. doi: [10.1007/s10556-014-9859-0](https://doi.org/10.1007/s10556-014-9859-0).

19. Kolosov AE. Effect of low-frequency ultrasonic treatment regimes on reactoplastic polymer composite material operating properties. *Chem. and Petrol. Eng.* 2014; 50 (3–4): 150–155. doi: [10.1007/s10556-014-9871-4](https://doi.org/10.1007/s10556-014-9871-4).

20. Kolosov AE. Efficiency of liquid reactoplastic composite heterofrequency ultrasonic treatment. *Chem. and Petrol. Eng.* 2014; 50 (3–4): 268–272. doi: [10.1007/s10556-014-9893-y](https://doi.org/10.1007/s10556-014-9893-y).

21. Kolosov AE. Low-Frequency Ultrasonic Treatment of Liquid Reactoplastic Media with Pressure Variation. *Chem. and Petrol. Eng.* 2014; 50 (5–6): 339–342. doi: [10.1007/s10556-014-9904-z](https://doi.org/10.1007/s10556-014-9904-z).
22. Kolosov AE. Prerequisites for using ultrasonic treatment for intensifying production of polymer composite materials. *Chem. and Petrol. Eng.* 2014; 50 (1–2): 11–17. doi: [10.1007/s10556-014-9846-5](https://doi.org/10.1007/s10556-014-9846-5).
23. Kolosov AE, Sakharov AS, Sivetskii VI, Sidorov DE, Sokolskii AL. Substantiation of the efficiency of using ultrasonic modification as a basis of a production cycle for preparing reinforced objects of epoxy polymer composition. *Chem. and Petrol. Eng.* 2012; 48 (5–6): 391–397. doi: [10.1007/s10556-012-9629-9](https://doi.org/10.1007/s10556-012-9629-9).
24. Kolosov AE, Karimov AA, Khozin VG, Klyavlin VV. Impregnation of fibrous fillers with polymer binders. 3. Ultrasonic intensification of impregnation. *Mech. of Compos. Mater.* 1989; 24 (4): 494–502. doi: [10.1007/BF00608132](https://doi.org/10.1007/BF00608132).
25. Kolosov AE, Virchenko GA, Kolosova EP, Virchenko GI. Structural and technological design of ways for preparing reactoplastic composite fiber materials based on structural parametric modeling. *Chem. and Petrol. Eng.* 2015; 51 (7–8): 493–500. doi: [10.1007/s10556-015-0075-3](https://doi.org/10.1007/s10556-015-0075-3).
26. Karimov AA, Kolosov AE, Khozin VG, Klyavlin VV. Impregnation of fibrous fillers with polymer binders. 4. Effect of the parameters of ultrasound treatment on the strength characteristics of epoxy binders. *Mech. of Compos. Mater.* 1989; 25 (1): 82–88. doi: [10.1007/BF00608456](https://doi.org/10.1007/BF00608456).
27. Kolosov AE, Karimov AA, Repelis IA, Khozin VG, Klyavlin VV. Impregnation of fibrous fillers with polymeric binders 6. Effect of parameters of ultrasound treatment on strength properties of wound fibrous composites. *Mech. of Compos. Mater.* 1990; 25 (4): 548–555. doi: [10.1007/BF00610711](https://doi.org/10.1007/BF00610711).
28. Kolosov AE, Sivetskii VI, Kolosova EP, Lugovskaya EA. Procedure for analysis of ultrasonic cavitator with radiative plate. *Chem. and Petrol. Eng.* 2013; 48 (11–12): 662–672. <https://doi.org/10.1007/s10556-013-9677-9>.

29. Luzgarev AS, Tkachenko TB, Moroz AA, Luzgarev SV. Preparation of carbon material dispersions in polydimethylsiloxane rubber solutions. *Vestn. KemGU. Ser. Khim.* 2013; 3 (55): pp. 88–90.
30. Choi SUS, Eastman JA. Enhancing thermal conductivity of fluids with nanoparticles. In: *Developments and Applications of Non-Newtonian flows*. New York: ASME; 1995. <https://www.osti.gov/biblio/196525/>.
31. Luzgarev SV, Denisov VYa. Effective approach to modification of polysiloxane structure and properties. *Sovr. Naukoem. Tekhnol.* 2005; 8: 34–35.
32. Sapronov OO. Study Of The Nature Of Chemical And Physical Links Of Epoxy Nanocomposites by IR, EPR Spectral Analysis and Optical Microscopy. *Intercollegiate digest "Scientific Notes"*. Lutsk, 2013; 43: 187-198.
33. Green AA, Hersam MC. Emerging Methods for Producing Monodisperse Graphene Dispersions. *Journal of Phys. Chem. Let.* 2010; 544-549. doi: 10.1021/jz900235f.
34. Hielscher K. Ultrasound for the Preparation of Graphene. In: *Catalogue of Hielscher Ultrasonics*. 2018. <http://pdf.directindustry.com/pdf/hielscher/ultrasound-preparation-graphene-hielscher-ultrasonics/24814-379925.html>.
35. Vakhrushev AV, Fedotov AYu, Vakhrushev AA, Suetin MV. Patent RU 2301771, IPC B82B3/00. Method and device for mixing nano-particles. Appl. No. 2005138015/28. Subm. Dec. 6, 2005. Publ. Jun. 27, 2007, Byull. No. 18.
36. Tarasov VA, Stepanishchev NA, Stepanishchev AN. et al. Patent RU 2500695, IPC C08J3/205. Method of preparing nanosuspension for producing polymer nanocomposite. Appl. No 2012124228/04. Subm. Jun. 13, 2012. Publ. Dec. 10, 2013, Byull. No. 34.
37. Ageev OA, Syurik YuV. Patent RU 2400462, IPC C07C1/00, B82B1/00 (2006.01). Method of preparing polymer/carbon nanotubes composite on substrate. Appl. No. 2009113378/04. Subm. Apr. 9, 2009. Publ. Sept. 27, 2010, Byull. No. 27.

38. Bingle Ruan, Anthony M. Jacobi. Ultrasonication effects on thermal and rheological properties of carbon nanotube suspensions. *Nanoscale Research Letters*. 2012; 7: 127. <https://doi.org/10.1186/1556-276X-7-127>.
39. Geim AK. Graphene: Status and Prospects. *Science*. 2009; 324: 1530-1534. <http://arxiv.org/ftp/arxiv/papers/0906/0906.3799.pdf>.
40. Xu H, Suslick KS. Sonochemical Preparation of Functionalized Graphenes. *J. of Amer. Chem. Soc.* 2011; 133: 9148-9151. doi: 10.1021/ja200883z.
41. Eletsii AV, Iskandarova IM, Knizhnik AA, Krasikov DN. Graphene: fabrication methods and thermophysical properties. *Phys. Usp.* 2011; 54: 227–258. doi: [10.3367/UFNe.0181.201103a.0233](https://doi.org/10.3367/UFNe.0181.201103a.0233)
42. Vanmaekelbergh D. Self-assembly of colloidal nanocrystals as route to novel classes of nanostructured materials. *Nano Today*. 2011; 6: 419-437. <http://dx.doi.org/10.1016/j.nantod.2011.06.005>.
43. Park G, Lee KG, Lee SJ; Park TJ., Wi R, Kim DH. Synthesis of Graphene-Gold Nanocomposites via Sonochemical Reduction. *J. of Nanosc. and Nanotechn.* 2011; 11 (7): 6095-6101. <https://doi.org/10.1166/jnn.2011.4446>.
44. An X, Simmons T, Shah R, Wolfe C, Lewis KM, Washington M, Nayak SK, Talapatra S, Kar S. Stable Aqueous Dispersions of Noncovalently Functionalized Graphene from Graphite and their Multifunctional High-Performance Applications. *Nano Letters*, 2010; 10: 4295-4301. doi: 10.1021/nl903557p.
45. Stankovich S, Dikin DA, Piner RD, Kohlhaas KA, Kleinhammes A, Jia Y, Wu Y, Nguyen ST, Ruoff RS. Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide. *Carbon*. 2007; 45: 1558-1565. <https://doi.org/10.1016/j.carbon.2007.02.034>.
46. Stengl V, Popelkova D, Vlácil P. TiO₂-Graphene Nanocomposite as High Performance Photocatalysts. *J. of Phys. Chem. C*. 2011; 115: 25209-25218. doi: 10.1021/jp207515z.
47. Jiao L, Zhang L; Wang X, Diankov G, Dai H. Narrow graphene nanoribbons from carbon nanotubes. *Nature*. 2009; 458: 877-880. doi: [10.1038/nature07919](https://doi.org/10.1038/nature07919).

48. Savoskin MV, Mochalin VN, Yaroshenko AP, Lazareva NI, Konstantinova TE, Baruskov IV, Prokofiev IG. Carbon nanoscrolls produced from acceptor-type graphite intercalation compounds. *Carbon*. 2007; 45: 2797-2800. <https://doi.org/10.1016/j.carbon.2007.09.031>.
49. Viculis LM, Mack JJ, Kaner RB. A Chemical Route To Carbon Nanoscrolls. *Science*. 2003; 299 (5611): 1361. <https://doi.org/10.1126/science.1078842>.
50. <https://en.wikipedia.org/wiki/Gel>
51. Wencai Ren, Hui-Ming Cheng. When two is better than one. *Nature*. 2013; 497: 448–449. doi: 10.1038/497448a <https://www.nature.com/articles/497448a>
52. http://www.physics.by/e107_files/mono/monograf_4fed_pdf/4fed_gl5.pdf
53. Qiu L, Liu JZ, Chang SLY, Wu Y, Li D. Biomimetic superelastic graphene-based cellular monoliths. *Nature Commun*. 2012; 3: 1241. doi: 10.1038/ncomms2251.
54. Chen ZP, Ren WC, Gao LB, Liu BL, Pei SF, Cheng HM. Three-dimensional flexible and conductive interconnected graphene networks grown by chemical vapour deposition. *Nature Materials*. 2011; 10: 424–428. <http://dx.doi.org/10.1038/nmat3001>.
55. Sultanov FR, Mansurov ZA. About aerogels based on carbon nanomaterials. *Chem. Bull. of Kazakh Nat. Univ*. 2014; 4: 67-82. http://dx.doi.org/10.15328/chemb_2014_467-82.
56. Hu H, Zhao Z, Wan W, Gogotsi Yu, Qiu J. Ultralight and Highly Compressible Graphene Aerogels. *Advanced Materials*. 2013; 25: 2219-2223. <http://dx.doi.org/10.1002/adma.201204530>.
57. Yehong Cheng, Shanbao Zhou, Ping Hu, Guangdong Zhao, Yongxia Li, Xinghong Zhang, Wenbo Han. Enhanced mechanical, thermal, and electric properties of graphene aerogels via supercritical ethanol drying and high-temperature thermal reduction. *Scientific Reports*. 2017; 7: 1439. doi: [10.1038/s41598-017-01601-x](https://doi.org/10.1038/s41598-017-01601-x).
58. Lin Y, Ehlert GJ, Bukowsky C, Sodano HA. Superhydrophobic Functionalized Graphene Aerogels. *ACS Applied Materials & Interfaces*. 2011; 3: 2200-2203. doi: 10.1021/am200527j.

59. Jihao L, Jingye L, Hu M, Siyuan X, Bowu Z, Linfan L, Hongjuan M, Jianyong Z, Ming Y. Ultra-light, compressible and fire-resistant graphene aerogel as a highly efficient and recyclable absorbent for organic liquids. *J. Mater. Chem. A*, 2014; 2: 2934-2941. doi: 10.1039/C3TA14725H.
60. Han Hu, Zongbin Zhao, Yury Gogotsi, Jieshan Qiu. Compressible Carbon Nanotube–Graphene Hybrid Aerogels with Superhydrophobicity and Superoleophilicity for Oil Sorption. *Environmental Science & Technology Letters*. 2014; 1(3): 214–220. <http://dx.doi.org/10.1021/ez500021w>.
61. Haiyan S, Zhen X, Chao G. Multifunctional, Ultra-Flyweight, Synergistically Assembled Carbon Aerogels. *Advanced Materials*. 2013; 25: 2554–2560. <https://doi.org/10.1002/adma.201204576>.
62. Dan Li, Richard B. Kaner. Graphene-Based Materials. *Science*. 2008; 320 (5880): 1170-1171. doi: 10.1126/science.1158180.
63. Sun HY, Xu Z, Gao C. Multifunctional, Ultra-Flyweight, Synergistically Assembled Carbon Aerogels. *Adv. Mater.* 2013; 25: 2554-2560. <http://dx.doi.org/10.1002/adma.201204576>.
64. Li D, Muller MB, Gilje S, Kaner RB, Wallace GG. Processable aqueous dispersions of graphene nanosheets. *Nat. Nanotechnol.* 2008; 3: 101-105. doi: [10.1038/nnano.2007.451](https://doi.org/10.1038/nnano.2007.451).
65. Wufeng C, Lifeng Y. In situ self-assembly of mild chemical reduction graphene for three-dimensional architectures. *Nanoscale*. 2011; 3: 3132-3137. doi:10.1039/C1NR10355E.
66. Compton OC, An Z, Putz KW, Hong BJ, Hauser BG, Brinson LC, Nguyen SBT. Additive-free hydrogelation of graphene oxide by ultrasonication. *Carbon*. 2012; 50: 3399-3406. <https://doi.org/10.1016/j.carbon.2012.01.061>.
67. Zongping Chen, Wencai Ren, Libo Gao, Bilu Liu, Songfeng Pei, Hui-Ming Cheng. Three-dimensional flexible and conductive interconnected graphene networks grown by chemical vapour deposition. *Nature Mater.* 2011; 10: 424–428. doi: 10.1038/nmat3001.
68. Li N, Chen Z, Ren W, Li F, Cheng HM. Flexible graphene-based lithium ion batteries with ultrafast charge and discharge rates. In: *Proc. Natl. Acad. Sci. USA*. USA. 2012; 109 (43): 17360-17365. doi: 10.1073/pnas.1210072109.

69. Cao X, Xiehong Cao, Yumeng Shi, Wenhui Shi, Gang Lu, Xiao Huang, Qingyu Yan, Qichun Zhang, Hua Zhang. Preparation of Novel 3D Graphene Networks for Supercapacitor Applications. *Small*. 2011; 7: 3163–3168. doi: 10.1002/sml.201100990.
70. Chen Z, Xu C, Ma C, Ren W, Cheng H-M. Lightweight and flexible graphene foam composites for high-performance electromagnetic interference shielding. *Adv. Mater.* 2013; 25 (9): 1296–1300. <https://doi.org/10.1002/adma.201204196>.
71. Zineb Benzait, Levent Trabzon. A review of recent research on materials used in polymer–matrix composites for body armor application. *Journal of Composite Materials*. Publ. March 14, 2018. <https://doi.org/10.1177/0021998318764002>
72. Zhang J, Xionga Z, Zhao XS. Graphene–metal–oxide composites for the degradation of dyes under visible light irradiation. *J. Mater. Chem.* 2011; 21: 3634–3640. <http://dx.doi.org/10.1039/c0jm03827j>.
73. Hu H, Zhao Z, Zhou Q, Zhou Y, Qiu J. Direct polymer infiltration of graphene aerogels for the production of conductive nanocomposite. In: *Proceedings of International Conference “Carbon-2013”*. Rio de Janiero, Brazil; 2013: 152-155.
74. Ne Myo Han, Zhenyu Wang, Xi Shen, Ying Wu, Xu Liu, Qingbin Zheng, Tae-Hyung Kim, Jinglei Yang, Jang-Kyo Kim. Graphene Size-Dependent Multifunctional Properties of Unidirectional Graphene Aerogel/Epoxy Nanocomposites. *ACS Appl. Mater. Interfaces*. 2018; 10 (7): 6580–6592. doi: 10.1021/acsami.7b19069.
75. Pak Seong Yeol. A Study on Thermal Behavior of Lightweight Carbon/Polymer Composites. <http://hdl.handle.net/10371/136760>.
76. An F, Li X, Min P, Li H, Dai Z, Yu ZZ. Highly anisotropic graphene/boron nitride hybrid aerogels with long-range ordered architecture and moderate density for highly thermally conductive composites. *Carbon*. 2018; 126: 119-127. <https://doi.org/10.1016/j.carbon.2017.10.011>.
77. Guan FL, An F, J Yang, X Li, XH Li, ZZ Yu. Fiber-reinforced three-dimensional graphene aerogels for electrically conductive epoxy composites with enhanced mechanical properties.

Chinese Journal of Polymer Science. 2017; 35 (11): 1381–1390. <http://dx.doi.org/10.1007/s10118-017-1972-z>.

78. Kim Jang-Kyo. Highly-aligned 2D and cellular interconnected 3D graphene/polymer composites with exceptional multi-functional properties. In: *21st International Conference on Composite Materials*. Xi'an, 20-25 August, 2017. <http://www.iccm21.org/uploadfile/2017/0405/20170405045406689.pdf>

79. Jia J, Sun X, Lin X, Shen X, Mai YW, Kim JK. Exceptional Electrical Conductivity and Fracture Resistance of 3D Interconnected Graphene Foam/Epoxy Composites. *ACS Nano*. 2014; 8: 5774–5783. doi: 10.1021/nn500590g.

80. Wang Z, Shen X, Han NM, Liu X, Wu Y, Ye W, Kim JK. Ultralow Electrical Percolation in Graphene Aerogel/Epoxy Composites. *Chem. Mater.* 2016; 28: 6731–6741. doi: 10.1021/acs.chemmater.6b03206.

81. Ye S, Feng J, Wu P. Highly elastic graphene oxide–epoxy composite aerogels via simple freeze-drying and subsequent routine curing. *Journal of Materials Chemistry A*, 2013; 10. <http://pubs.rsc.org/-/content/articlelanding/2013/ta/c2ta01142e/unauth#!divAbstract>

82. Wan YJ, Gong LX, Tang LC, Wu LB, Jiang JX. Mechanical properties of epoxy composites filled with silane-functionalized graphene oxide. *Composites Part A: Applied Science and Manufacturing*. 2014; 64: 79-89. <https://doi.org/10.1016/j.compositesa.2014.04.023>

83. Ahmed S Wajid, HS Tanvir Ahmed, Sriya Das, Fahmida Irin, Alan F Jankowski, Micah J Green. High-Performance Pristine Graphene/Epoxy Composites With Enhanced Mechanical and Electrical Properties. *Macromolecular Materials and Engineering*. Wiley Online Library. First publ. 17 July 2012. <https://doi.org/10.1002/mame.201200043>

84. Sidorov DE, Kolosova EP, Kolosov AE, Shabliy TA. Analysis of blown process for producing polymer products by extrusion blow molding. *Eastern-European Journal of Enterprise Technologies*. 2018; 2/1 (92): 14–21. doi: 10.15587/1729-4061.2018.126015.

85. Sidorov DE, Kolosov AE, Pogorelyi OV, Gur'eva AA. Engineering approach to the determination of the radiation field of a Polyethyleneterephthalate (PET) medium under radiant heating. *Journal of Engineering Physics and Thermophysics*. 2015; 88 (6): 1409–1415. doi: [10.1007/s10891-015-1325-0](https://doi.org/10.1007/s10891-015-1325-0).
86. Sakharov AS, Kolosov AE, Sokolskii AL, Sivetskii VI. Modeling the mixing of polymeric composites in an extrusion drum mixer. *Chem. and Petrol. Eng.* 2012; 47 (11–12): 799–805. doi: [10.1007/s10556-012-9553-z](https://doi.org/10.1007/s10556-012-9553-z).
87. Sakharov AS, Kolosov AE, Sivetskii VI, Sokolskii AL. Modeling of Polymer Melting Processes in Screw Extruder Channels. *Chem. and Petrol. Eng.* 2013; 49 (5–6): 357–363. doi: [10.1007/s10556-013-9755-z](https://doi.org/10.1007/s10556-013-9755-z).
88. Kovalenko KG, Kolosov AE, Sivetskii VI, Sokolskii AL. Modeling Polymer Melt Flow at the Outlet from an Extruder Molding Tool. *Chem. and Petrol. Eng.* 2014; 49 (11): 792–797. doi: [10.1007/s10556-014-9837-6](https://doi.org/10.1007/s10556-014-9837-6).
89. Sidorov DE, Sivetskii VI, Kolosov AE, Sakharov AS. Shaping of corrugation profiles during production of corrugated tubular articles. *Chem. and Petrol. Eng.* 2012; 48 (5–6): 384–390. doi: [10.1007/s10556-012-9628-x](https://doi.org/10.1007/s10556-012-9628-x).
90. Kolosov AE, Sakharov AS, Sidorov DE, Sivetskii VI. Aspects of profile shaping of corrugated tubular components. Part 1. Modeling of parameters of different profiles of corrugations, and also their shaping equipment. *Chem. and Petrol. Eng.* 2012; 48 (1–2): 60–67. doi: [10.1007/s10556-012-9575-6](https://doi.org/10.1007/s10556-012-9575-6).
91. Kolosov AE, Sakharov AS, Sidorov DE, Sivetskii VI. Manufacturing Technology: Aspects of profile shaping of corrugated tubular components. Part 2. Modeling the extrusion welding of layers of corrugated tubular articles. *Chem. and Petrol. Eng.* 2012; 48 (1–2): 131–138. doi: [10.1007/s10556-012-9588-1](https://doi.org/10.1007/s10556-012-9588-1).

92. Kolosov AE, Sakharov AS, Sidorov DE, Sivetskii VI. Aspects of profile shaping of corrugated tubular components. Part 3. Extrusion shaping of tubular polymeric blanks for manufacture of corrugated pipes. *Chem. and Petrol. Eng.* 2012; 48 (3–4): 199–206. doi: [10.1007/s10556-012-9598-z](https://doi.org/10.1007/s10556-012-9598-z).
93. Kolosov AE, Sakharov OS, Sivetskii VI, Sidorov DE, Pristailov SO. Effective hardware for connection and repair of polyethylene pipelines using ultrasonic modification and heat shrinkage. Part 1. Aspects of connection and restoration of polymeric pipelines for gas transport. *Chem. and Petrol. Eng.* 2011; 47: 204-209. doi: [10.1007/s10556-011-9447-5](https://doi.org/10.1007/s10556-011-9447-5).
94. Kolosov AE, Sakharov OS, Sivetskii VI, Sidorov DE, Pristailov SO. Effective hardware for connection and repair of polyethylene pipelines using ultrasonic modification and heat shrinkage. Part 2. Production bases for molding of epoxy repair couplings with shape memory. *Chem. and Petrol. Eng.* 2011; 47: 210. doi: [10.1007/s10556-011-9448-4](https://doi.org/10.1007/s10556-011-9448-4).
95. Kolosov AE, Sakharov OS, Sivetskii VI, Sidorov DE, Pristailov SO. Effective hardware for connection and repair of polyethylene pipelines using ultrasonic modification and heat shrinkage. Part 3. Analysis of surface-treatment methods for polyethylene pipes connected by banding. *Chem. and Petrol. Eng.* 2011; 47: 216. doi: [10.1007/s10556-011-9449-3](https://doi.org/10.1007/s10556-011-9449-3).
96. Kolosov AE, Sakharov OS, Sivetskii VI, Sidorov DE, Pristailov SO. Effective hardware for connection and repair of polyethylene pipelines using ultrasonic modification and heat shrinkage. Part 4. Characteristics of practical implementation of production bases developed using epoxy-glue compositions and banding. *Chem. and Petrol. Eng.* 2011; 47: 280. doi: [10.1007/s10556-011-9460-8](https://doi.org/10.1007/s10556-011-9460-8).
97. Kolosov AE, Sakharov OS, Sivetskii VI, Sidorov DE, Pristailov SO. Effective hardware for connection and repair of polyethylene pipelines using ultrasound modification and heat shrinking. Part 5. Aspects of thermistor couplings and components used in gas-pipeline repair. *Chem. and Petrol. Eng.* 2011; 47: 285. doi: [10.1007/s10556-011-9461-7](https://doi.org/10.1007/s10556-011-9461-7).
98. Yeromin AV, Kolosov AE. Modeling of energy effective solutions regarding the heating system and facade heat insulation during implementation of thermomodernization. *Eastern-*

European Journal of Enterprise Technologies. 2018; 8 (91): 49–58. doi: [10.15587/1729-4061.2018.123021](https://doi.org/10.15587/1729-4061.2018.123021).

99. Kolosov AE, Klyavlin VV. Determination of the parameters of a geometric model of the structure of directionally reinforced fiber composites. *Mech. of Compos. Mater.* 1988; 23 (6): 699–706. doi: [10.1007/BF00616790](https://doi.org/10.1007/BF00616790).

100. Kolosov AE, Klyavlin VV. Several aspects of determination of the adequate model of the structure of oriented fiber-reinforced composites. *Mech. of Compos. Mater.* 1989; 24 (6): 751–757. doi: [10.1007/BF00610779](https://doi.org/10.1007/BF00610779).

101. Kichigin AF, Kolosov AE, Klyavlin VV, Sidyachenko VG. Probabilistic-geometric model of structurally inhomogeneous materials. *Soviet Mining Science*. 1988; 24 (2): 87–94. doi: [10.1007/BF02497828](https://doi.org/10.1007/BF02497828).

102. Kolosov AE. Impregnation of fibrous fillers with polymer binders. 1. Kinetic equations of longitudinal and transverse impregnation. *Mech. of Compos. Mater.* 1988; 23 (5): 625–633. doi: [10.1007/BF00605688](https://doi.org/10.1007/BF00605688).

103. Kolosov AE, Repelis IA, Khozin VG, Klyavlin VV. Impregnation of fibrous fillers with polymer binders. 2. Effect of the impregnation regimes on the strength of the impregnated fillers. *Mech. of Compos. Mater.* 1988; 24 (3): 373–380. doi: [10.1007/BF00606611](https://doi.org/10.1007/BF00606611).

104. Kolosov AE, Repelis IA. Saturation of fibrous fillers with polymer binders 5. Optimization of parameters of the winding conditions. *Mech. of Compos. Mater.* 1989; 25 (3): 407–415. doi: [10.1007/BF00614811](https://doi.org/10.1007/BF00614811).

105. Carlos A. Ávila-Orta, Zoe V. Quiñones-Jurado, Miguel A. Waldo-Mendoza, Erika A. Rivera-Paz, Víctor J. Cruz-Delgado, José M. Mata-Padilla, Pablo González-Morones, Ronald F. Ziolo. Ultrasound-Assist Extrusion Methods for the Fabrication of Polymer Nanocomposites Based on Polypropylene/Multi-Wall Carbon Nanotubes. *Materials*. 2015; 8(11)Ж 7900-7912. doi:[10.3390/ma8115431](https://doi.org/10.3390/ma8115431).

106. Carlos Ávila-Orta, Carlos Espinoza-González, Guillermo Martínez-Colunga, Darío Bueno-Baqués, Alfonso Maffezzoli, Francesca Lionetto. An Overview of Progress and Current Challenges in Ultrasonic Treatment of Polymer Melts. *Adv. in Polym. Techn.* 2013; 32 (S1): E582–E602. <https://doi.org/10.1002/adv.21303>.
107. Yanan Zhao, Eusebio Cabrera, Tairong Kuang, Dan Zhang, Willie Yang, Jose M. Castro. Ultrasonic Processing of Epoxy/CNT Nanopaper Composites. In: *SPE ANTEC*. Anaheim. 2017: 798–803. <http://leaders.4spe.org/spe/conferences/ANTEC2017/papers/390.pdf>.
108. Yablokova MYu, Serbin VV, Avdeev VV et al. Patent RU 2415884. IPC C08J3/20, C08J5/04, B32B27/18 (2006.01). Method of preparing nano-modified binder, binder and prepreg based on said binder. Appl. No. 2008147219/05. Subm. Dec. 1, 2008, publ. Apr. 10, 2011, Byull. No. 10.
109. Kablov EN, Gunyaev GM, Il'chenko SI et al. Patent RU 2278028F. IPC B32B27/38, C08J5/24, C08L63/00, C09J163/00, C08K3/04, C07C211/01 (2006.01). Prepreg and article made of the same. Appl. No. 2005110370/04. Subm. Apr. 11, 2005, publ. Jun. 20, 2006, Byull. No. 17.
110. Kolosov OE, Sivetskii VI, Kolosova OP. *Preparation of Fiber-Filled Reactoplastic Polymer Composite Materials with Ultrasonic Treatment* [in Ukrainian]. Kiev: VPK Politekhnik; 2015.
111. Kolosov AE. Preparation of Reactoplastic Nanomodified Polymer Composites. Part 2. Analysis of means of forming nanocomposites (patent review). *Chem. and Petrol. Eng.* 2016; 51 (9–10): 640–645. <https://doi.org/10.1007/s10556-016-0100-1>.