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STUDY OF STRENGTH AND STRAINS OF COMPOSITE MATERIALS BASED ON FILLED POLYMERS

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Abstract. The article aims to study compressive and bending strength of polymer materials and their strains with various amounts of filler to determine the optimal value. The polymer composite material consists of polymethyl methacrylate as a binder and quartz sand as a filler with the fractions at 0.14 mm, 0.315 mm, 0.63 mm. The filler amounts were changed from 100 to 500 weight parts. The strength of polymer filled material was determined by testing cube samples under compressive and beam samples under bending. The strains were determined by testing prism samples under tensile. The study results show the significant impact of the fraction type on the strength and deformation characteristics of composite materials based on polymethyl methacrylate. The samples with the finest fraction of filler 0.14 mm and 150 weight part demonstrate the biggest strength – 90 MPa under compressive and 40 MPa under bending. The increase both particles size and their amount led to the decrease the strength of filled composite. The analysis of the dependencies between stresses and strains shows than deformations drop as far as the amount of mineral filler declines. In proportion as the quantity of filler goes up, the creep deformations occur under less amount of tensile stress. It is linked the less interaction on the border "filler-polymer" and, as consequences, the bigger possibilities of polymer to plastic deformations. Based on obtained dependencies, the patterns of defect development under loading have been proposed that involves taking into account interphase interaction on the borders "filler-polymer" and "filler-filler". The less the particles size, the bigger their specific surface and structure-forming effect. The increase of sand particles size and their amount leads to growth of porosity, goes down the strength of composite material and drops the capability of elastic recoverable strains. The practical importance of this study is the possibility the usage of the filled polymer composites for recovering and renovation works where traditional cost cement fillers are replaced with cheap and ecofriendly fine sand.

Key words: filled polymer material, strength, stress, strain, polymethyl methacrylate, filler.

Introduction. Filled polymers are an important class of materials due to their stiffness, strength, low density, abrasion resistance, significant electrical properties, and ease of manufacture. They are widely used in civil engineering as a reliable method of mitigating the vibration and dynamic impacts on structures such as bridges and buildings. The application of protective coatings based on various polymers is the most effective method of corrosion protection to isolate the material surface from direct influence of aggressive environments due to the high strength, wear resistance and resistance to aggressive environments of polymer composite materials. However, each application depends on a unique set of operating conditions, including cyclic loads, impact conditions, tensile conditions, and strain rates. The study of the behaviour of these types of composite materials under different conditions is crucial for design, modelling, and ensuring durability. Time-dependent behaviour, such as creep and stress relaxation, remains a serious problem for predicting the service life of filled polymer composites.

The widespread use of composite materials is hindered by a lack of understanding of their long-term characteristics under operating conditions. Understanding the mechanical failure behaviour, such as fatigue and creep, is crucial for determining the expected service life of structures and estimating the potential maintenance costs. Due to the limited knowledge of the long-term behaviour of polymer filled materials, many applications in civil infrastructure are limited by a conservative consideration of creep. More accurate predictions of creep behaviour are needed to improve cost-effectiveness when replacing standard fillers with waste materials or by-products. Thus, studying the deformation properties and predicting the influence of structural formation features on the ultimate strength of filled polymer composites is a topical task.

Analysis of recent research and publications. The inclusion of fillers affects the entire range of properties of filled polymeric materials starting from their manufacture. An increase in the amount of filler leads to a significant rise in the viscosity of polymer mixtures that causes technological difficulties when coatings are applied to the surface [1]. To overcome this negative effect, the authors proposed to introduce highly dispersed amorphous silicon dioxide in addition to aluminosilicate spheres. One of the main disadvantages of polymer materials is a significant amount of shrinkage during application and subsequent curing. As shown by the authors [2], to reduce shrinkage deformations, the most suitable filler is quartz sand with a particle size of 0.14 mm in the amount of 150 weight parts of acrylic polymer. Shrinkage deformations are 5 times less than in the unfilled composition and 10 % less than when filled with Portland cement. In addition, the shrinkage deformations are the smallest when the optimal thickness of the coating is 5 mm. This is explained by the action of the filler surface which orientates the layers of polymer molecules. In turn, this causes strengthening of the nearest polymer layers, but the strength decreases with distance from the surface. As the coating thickness increases, the cohesive forces are less than the tensile stress. It leads to cracking of thicker layers.

Filled polymer compositions are of great importance in the creation of anticorrosion protective coatings [3-5]. The authors of these studies pay attention to the targeted control of the interaction processes between the polymer and fillers to create coatings with high performance characteristics. In paper [3], it is shown that localised areas of the filler particle surface are activators of physical and chemical interfacial bonds during the structure formation of anticorrosion coatings. The optimal combination of a modifier, iron-carbide charge and antioxidant ensures the maximum increase in the degree of gelation of the epoxy matrix and the creation of a barrier to the penetration of an aggressive environment. It is confirmed by the results of experimental studies in [3]. Fillers such as graphite, mica, and aluminium oxide, which differ in chemical nature and particle size, were used in [4] to study the water absorption, chemical resistance, and microhardness of epoxy compositions. Mica has the greatest impact on water absorption and chemical resistance to acids. In the opinion of the authors [4], it is associated with the high concentration of surface hydroxyl groups that act as ‘traps’ for water molecules. When the amount of mica increases, water absorption and the ability to absorb acids decreases, due to the neutralisation of its surface hydroxyl groups.

To establish the ultimate content of dispersed particles of furnace soot and crystalline boron in an epoxy binder for the formation of protective coatings with the required characteristics, it was proved that when furnace soot particles were introduced in the amount of 15 wt. %, composites with a bending tensile stress of 50.53 MPa and a bending elastic modulus of 3.48 GPa were formed [5]. For the creation of composites with a destructive bending stress of 73.66 MPa and a bending elastic modulus of 4.70 GPa, the critical content of fine-dispersed crystalline boron particles should be 30...40 wt.% per 100 wt.% of polymer. In the study [6], fillers of different chemical nature were used: barium sulfate, calcium carbonate, zinc oxide, metakaolin, microsilica, wollastonite, talc, fly ash, and blast furnace granulated slag. This allowed us to clarify the principles of using each of these types and determine the most appropriate application. For example, the use of fillers in the form of industrial waste reduces the corrosion resistance of coatings. In contrast, the use of silicates is effective to increase the corrosion resistance of powder coatings.

The deformation mechanisms of carbon black filled rubbers subjected to single and repeated cyclic tensile tests were discussed it the article [7]. For a low filler content, the destruction mainly occurs in the elastic rubber network with an irreversible break in the bonding of the molecular chains. For high filler content, the damage initially occurs in the elastic rubber network, and then the filler network reorganises and carries the load due to the increasing of strain giving way to localised damage near the fillers. As a result, rubber releases both bound at the filler boundary and trapped in the filler aggregates. This partially reversible release includes the loss of weak molecular interactions (chain slippage, physical bonding) and prevents further development of irreversible cavities in the elastically active phase of the rubber during testing.

The properties of filled composite materials are affected not only by the degree of dispersion but also by the shape of the particles. In the work [8], the degree of reinforcement provided by silicon dioxide as a spherical filler with a low aspect ratio and high interaction between fillers was compared with mica as a plate-like filler with a higher aspect ratio and less interaction between fillers. The mechanical characteristics of composites based on epoxy resin reinforced with transverse carbon fibre can be improved by the inclusion of hybrid nano- and microparticles [9]. Fillers in the form of graphene nanoplates in carbon plastic promote interfacial interaction, create a chemical bonding network with the epoxy resin, as well as cause a mechanical blocking effect that increases flexural strength.

The main limitation of filled polymers is their brittle nature mainly due to the low stiffness and low fracture toughness of the resin. It leads to a deterioration of matrix-predominated composite properties including impact viscosity, compressive strength, in-plane shear, fracture toughness and interlaminar strength. However, a wide variety of fillers makes it possible to control the elastic-strength, deformation, and other properties of polymer-based composite materials in a wide range. In the study [10], dispersed fillers such as microcalcite, graphite, and molybdenum disulfide were used to obtain a composite material based on an epoxy binder. The volume content of the filler was 5, 10, 20, 30, or 40 percent of the total volume of the composition. The authors have established the dependence of the composite deformation on the filler content: with an increase in the filler content, the deformation decreases. The epoxy resin samples filled with molybdenum disulfide were less deformed than samples filled with microcalcite and graphite. With an increase in the filler content, the reversible deformation slowed down, and the value of the relative total deformation decreased. In addition, for samples of the polymer composite material containing more than 20 % graphite, the reversible deformation occurs more slowly than for the polymer composite material with microcalcite and molybdenum disulfide at the same volume filler content.

The study [11] was dedicated to investigating the influence of nanofillers added to epoxy composite materials on their mechanical properties. The fillers can react with macromolecules in the epoxy matrix and contribute to a significant improvement in performance. The mechanical properties of these samples were investigated under tensile, compressive, bending and impact loading. The maximum tensile strength was found for the composite material modified with 0.5 wt. % nanosilica. For all types of samples, the tensile strength decreased with the addition of nanoparticles due to their agglomeration. It was found that the highest rising of compressive strength was at 89 % for the sample modified with 0.5 wt. % iron nanoxide. The impact strength was also increased with the addition of nanoparticles. The largest increasing was at 127 % for the nano-iron oxide modified sample. As for bending properties, the highest strength was found for the samples with 1.0 wt. % of nanosilica. The various functionality of the filler surface significantly affects the properties of the composite [12]. It has been shown that the deformation of the system decreases with the strengthening of polymer-filler interaction and with increasing filler proportion. For a system with a weak polymer-filler interaction, it was found that the strain increases with increasing the volume fraction of the filler and was even greater than that of the pure polymer system.

The analysis of the above-mentioned studies shows that there is no strictly unambiguous dependence between the strength and deformability of filled polymer compositions and the quantity and type of fillers. In each specific case, these characteristics depend on the chemical nature of the filler, the size and shape of its particles, and the intensity of surface reactions. Since the cheapest filler is sand, a significant amount of which is formed during the recycling of destroyed concrete structures [13], this study aims to investigate the properties of polymer compositions filled with sand of different fractions.

Purpose and tasks. The purpose of this of this study is to investigate the dependence of the strength and deformation of polymeric materials on the amount of filler. This implies the following research tasks: to investigate the compressive and bending strength of a polymer with fillers of different fractional composition; to determine the optimal quantity and type of filler; to analyse the effect of the selected composite material on the development of deformations.

Materials and research methods. The composition of the polymer composite material includes polymethyl methacrylate as a binder and quartz sand of 0.14 mm, 0.315 mm, and 0.63 mm fractions as a filler. Samples of the composite material were prepared by mixing polymethyl methacrylate with the hardener and filler. In this case, the amount of the 0.14 mm sand fraction ranged from 120 to 200, and the other fractions – from 200 to 500 mass parts by weight of the polymer part. Such ranges of fillers quantities were substantiated in previous studies [2], where it was found that the smallest shrinkage deformations were observed for filled polymer composites with a fraction of up to 200 wt.%. As for the fillers of larger fractions, their small amount increases the magnitude of shrinkage deformations, so it is advisable to use them in the amount of 200 to 500 wt.%. The strength of the filled polymer material was determined by testing the specimens with short-term static loading under compression and bending. For the axial compression test, cubes with an edge size of 40 mm were used (Fig. 1). For the static bending test, beams with a design span of 65 mm and a cross section of 10 mm ×15 mm were used. Deformations were determined on prisms 40 mm × 40 mm × 160 mm. The measurements were performed using the method of electrical resistance strain gauges on a press with a force gauge scale of 250 kN. Four strain gauges were glued to each edge of the sample along and perpendicular to the line of load action, connected in series, which made it possible to obtain averaged data on the value of longitudinal and transverse deformations. The transverse deformations were measured using a strain gauge station complete with a multipoint switch. After centring the specimens along the physical axis, the load was applied to the specimens in steps of 0.1 R until they were destroyed. The value of longitudinal and transverse deformations was recorded at each loading stage.

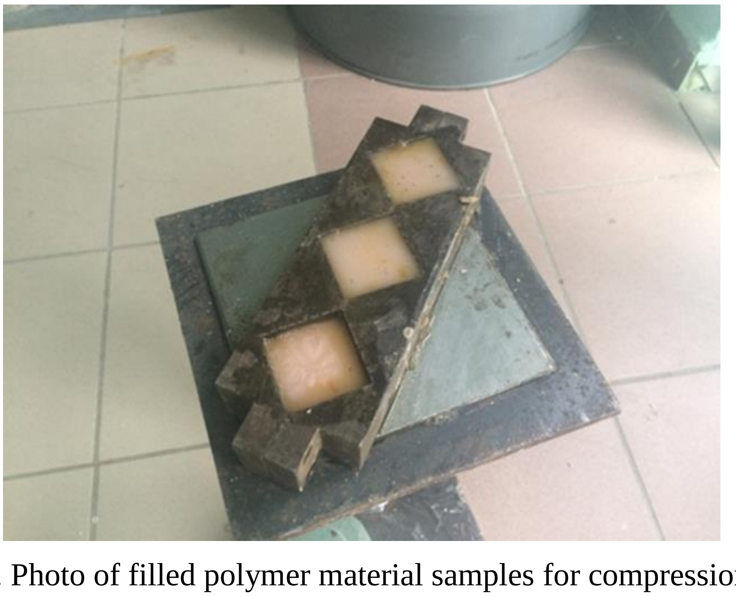

Fig. 1. Photo of filled polymer material samples for compression test

Research results. It was found that the strength of the filled polymer material is influenced by the proportion of quartz sand and its fraction (Fig. 2).

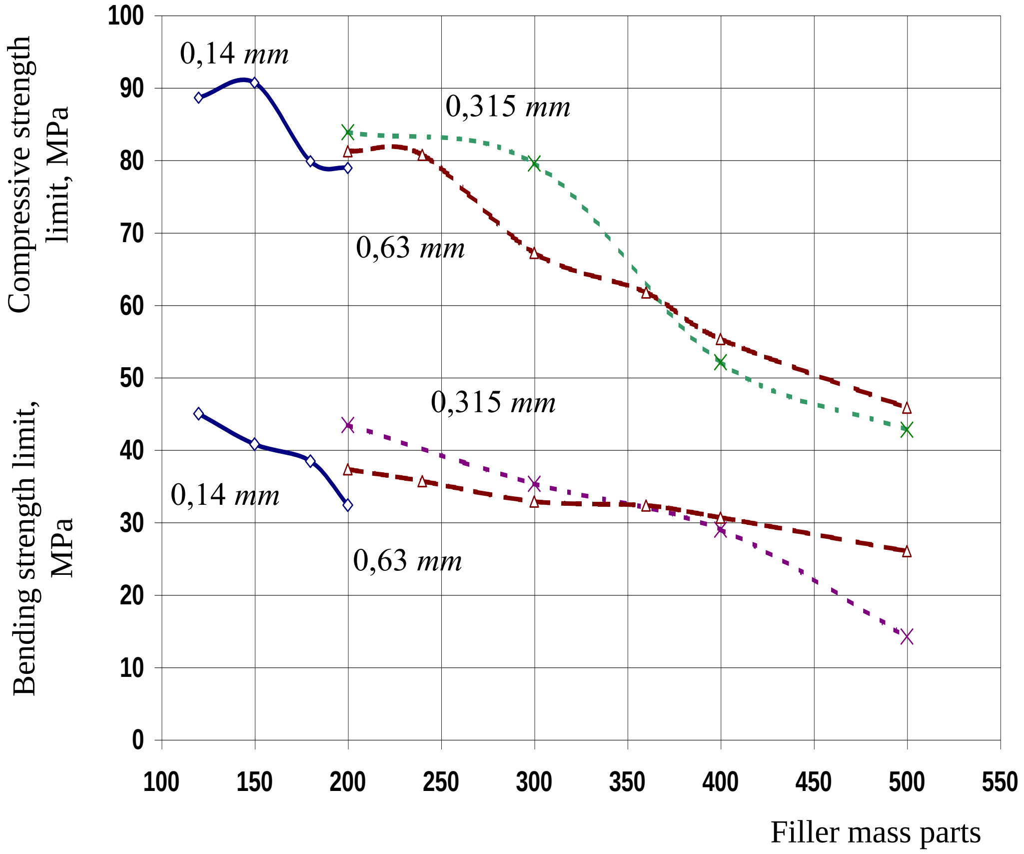

Fig. 2. Dependence of acrylic polymer mortar strength on the amount of filler

Filling the composition with sand with a grain fraction of 0.14 mm from 120 to 200 wt. % changes the compressive strength from 90.7 to 78.9 MPa and bending strength from 44.97 to 32.4 MPa. Filling the composition from 200 to 500 wt. % with sand with a grain fraction of 0.315 mm changes the compressive strength from 83.8 to 42.8 MPa and the bending strength from 43.4 to 24.2 MPa. A similar effect is observed for compositions with sand with a grain fraction of 0.63 mm. The finer the sand, the less it is needed to obtain strong compositions. When fine sands with a fraction of 0.14 mm are used, a decrease in strength of up to 40 % was observed after the introduction of 180 wt.% of sand, while for compositions with sand fraction of 0.315 mm, a decrease in strength was observed after the introduction of 300 wt.%, for compositions with sand fraction of 0.63 mm – 450 wt.%. For acrylic polymer mortar filled with sand grain fraction of 0.315 and 0.63 mm in the amount of 500 wt.%, the strength decreased by 51.5% and 43.8% under compression and 74.9% and 35.1% under bending, respectively.

The test results indicate the structuring role of the filler. Inorganic fillers can initiate the polymerisation process due to the existence of active centres on their surface which act as polymerisation centres and contribute to the formation of a continuous spatial polymer framework. The optimum ratio of filler to polymer is such that the thickness of the acrylic polymer film is sufficient to wet the entire surface of the particles but does not lead to an increase in creep deformation. A significant decrease of strength is explained by exceeding filler concentration that leads to a deficit in polymer volume and, as a result, to a decrease in wettability, disruption of the structure continuity. An increase in the proportion of sand in the composition from 400 to 500 wt. % leads to a decrease of the compressive and bending strength of the polymer mortar by 20%. Thus, with an increase in the proportion of polymer and a decrease in the fraction of quartz sand grains, the strength of the composition increases. It is in good agreement with the general principles of structure formation of composite materials. To study the dependence of the deformations of filled polymeric materials on the amount of filler, the composition with the highest values of compressive and bending strengths, i.e., samples with a fraction of 0.14 mm, was chosen. The obtained dependences for four filler values are shown in Fig. 3.

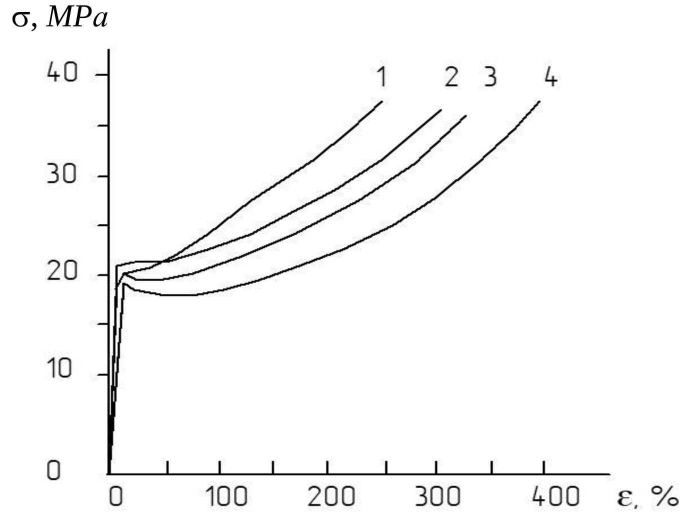

Fig. 3. Stress-strain curves for acrylic polymer mortar with different mass fractions of filler:
1 – 120 wt. %; 2 – 150 wt.%; 3 – 175 wt.%; 4 – 200 wt.%

The analysis of the longitudinal strain curves (Fig. 3) showed that the deformation decreases when the amount of mineral filler decreases, since an increase in the number of contacts between the polymer matrix and the filler naturally leads to a strengthening of the structure of the filled polymer and decreasing in its internal deformability. An increase in the filler content leads to a noticeable change in the slope of the strain curve associated with strain hardening. This is caused by the formation of micropores, which facilitates the plastic flow of the material. When the content of 0.14 *mm* sand increases, the deformation pattern changes from homogeneous to heterogeneous, and the yield peak appears (Fig. 3, curve 2). At the same time, the values of stress and strain at the yield strength gradually decrease: σ_y = 21.5 *MPa*, ε = 12 for a sand content of 150 wt.%; σ_y = 19.2 MPa, ε = 6 for a sand content of 200 wt.%. When the mass fraction of the filler increases, the change in creep strain occurs according to patterns that are largely related to the structure of the filled polymer. In the case of a system with a low filling level, there is no formation of a rigid structure of particles in the polymer, and the attraction and interaction of the filler with the polymer chains significantly reduces the load on the composite material. In a highly filled system, the filler particles begin to form aggregates with each other, and the repulsion between similarly charged sand particles promotes the sliding of molecular chains which leads to a continuous increase in strain with an increase in the filling proportion.

Conclusions. The results of the study indicate a significant effect of the sand fractional wakeup on the strength and deformation properties of the composite material based on polymethyl methacrylate. The highest strength (90 *MPa* under compression and 40 *MPa* under bending) was demonstrated by samples with the smallest fraction of 0.14 *mm* at an amount of 150 mass parts. An increase in both the particle size and the number of particles leads to a decrease in the strength of the filled composition, which is explained by the disruption of the optimal ratio of the filler particle surface to the amount of polymer that can be adsorbed. Also, for a low filler content, damage mainly occurs in the elastic polymer network, while at a high filler content, deformation occurs with damage initially localised in the elastic polymer network, but which is subsequently localised near the filler-filler interface. This mechanism has been confirmed by tensile deformation studies: at a higher degree of filling, a yield peak appears, and the overall magnitude of deformation increases.

The practical significance of the research is the possibility of using filled polymer composites for restorative repair work, in which traditional expensive cement fillers are replaced with cheaper and more environmentally friendly fine sand. Further research will be aimed at studying the adhesion of the developed composite materials to the surfaces intended for repair or restoration, as well as the durability of the applied coatings under the influence of various aggressive environments.

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ДОСЛІДЖЕННЯ МІЦНОСТІ І ДЕФОРМАЦІЙ КОМПОЗИЦІЙНИХ МАТЕРІАЛІВ НА ОСНОВІ НАПОВНЕНИХ ПОЛІМЕРІВ

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Анотація. Метою даної роботи є вивчення залежності міцності при стиску та згині і деформацій полімерних матеріалів від кількості наповнювача для визначення його оптимальної кількості. Полімерний композиційний матеріал складається із зв'язуючого поліметилметакрилата і наповнювача – піску кварцового фракцій 0,14 мм, 0,315 мм, 0,63 мм. Кількість наповнювачів кожної фракції змінювалася від 100 до 500 масових частин. Міцність полімерного наповненого матеріалу визначали випробуванням зразків-кубів на стиск і зразків-балочок при згині. Деформації визначали на зразках-призмах при їх розтягуванні. Результати дослідження свідчать про значний вплив фракційного складу піску на міцнісні і деформативні властивості композиційного матеріалу на основі поліметилметакрилату. Найбільшу міцність (90 МПа при стиску і 40 МПа при згині) продемонстрували зразки із найменшою фракцією наповнювача 0,14 мм при кількості 150 масових частин. Збільшення як розміру частинок так і їх кількості призводить до зменшення міцності наповненої композиції. Аналіз залежностей напруження-деформації показав, що деформації знижуються зі зменшенням кількості мінерального наповнювача. При збільшенні кількості наповнювача деформації текучості з'являються при менших значеннях напруження розтягу, що пов'язано із меншою взаємодією на границі наповнювач-полімер і, за рахунок цього, більшої здатності полімеру до пластичних деформацій. На основі отриманих залежностей запропоновано механізм розвитку пошкоджень структури при навантаженні, який полягає у врахуванні міжфазної взаємодії на границях розподілу полімер-наповнювач і наповнювач-наповнювач. Менший розмір частинок наповнювача призводить до збільшення їх питомої поверхні і більшої структуруючої ролі поверхні. Збільшення розмірів частинок піску і їх кількості збільшує пористість матеріалу, що зменшує міцність композиційного матеріалу і здатність до пружних оборотних деформацій. Практичне значення проведених досліджень полягає у можливості застосування наповнених полімерних композитів для відновлювальних ремонтних робіт, в яких традиційні дороговартісні цементні наповнювачі замінено більш дешевим і екологічним дрібним піском.

Ключові слова: наповнений полімерний матеріал, міцність, напруження, деформації, поліметилметакрилат, наповнювач.

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