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Review

The use of carbon-containing compounds to prepare functional and structural composite materials. A review.

Elena G. Zemtsova *, Andrey Yu. Arbenin, Yuri V. Sidorov, Nikita F. Morozov, Petr M. Korusenko, Boris N. Semenov and Vladimir M. Smirnov

Institute of Chemistry, St. Petersburg State University, 7/9 Universitetskaya nab., 199034 Saint Petersburg, Russia; aua47@yandex.ru (A.Y.A.); si_dorov@mail.ru (Y.V.S.); n.morozov@spbu.ru (N.F.M.); korusenko_petr@mail.ru (P.M.K.); semenov@bs1892.spb.edu (B.N.S.); vms11@yandex.ru; (V.M.S.)

* Correspondence: ezimtsova@yandex.ru (E.G.Z.)

Abstract: The review presents data on the synthesis, properties of carbon-containing compounds, their application in the technology of obtaining functional and structural composites. Such materials are widely used in recent years due to their good physical and mechanical properties. The review discusses in detail the influence of the chemical composition of various carbon-containing substances, and the size of the dispersed phase on the mechanical and physical properties of produced metal matrix composite. The review also concerns methods for the modern synthesis of graphene, carbon nanotubes, and metallic functional and structural composites with reinforcing carbon-containing compounds. Also presented are the results of metal matrix composite modeling. They show that the improved mechanical properties of CNT/Al composites can be attributed to three factors: CNT hardening, matrix grain refinement, and layered architecture.

Keywords: composite materials; functional properties; structural properties; carbon-containing substances; graphene; carbon nanotubes; aluminum

1. Introduction

The development of modern technology requires a qualitative improvement in technical characteristics, which can be achieved only if fundamentally new structural and functional materials are created and integrated. Operating conditions put forward extremely stringent requirements for new structural materials, the main of which is to ensure maximum strength and rigidity of structures with minimum weight characteristics. Replacing monolithic traditional materials with composites improves the reliability and weight efficiency of structures [1-5].

Composites based on light metal alloys reinforced with carbon compounds, due to their high anti-friction characteristics in combination with high wear resistance, bearing capacity, damping properties, low specific gravity, high operating temperatures, are very promising materials for friction pairs of ship structures, helicopters, and oil-producing equipment, rolling mills, textile machines.

One of the directions of the modern industry development is the transition to new functional and structural composites. Progress in the nuclear, aerospace, automotive, energy, semiconductor, transport and many other technology fields is largely based on the development of methods for obtaining new composites with a nanodispersed phase and improved functional properties [4-12].

For structural materials, operating conditions put forward extremely stringent requirements, the main of which are to ensure maximum strength and rigidity of structures with minimum weight characteristics, maximum impact strength in a wide temperature range, high wear resistance and load-bearing capacity, necessary tribological properties, high fatigue strength, reliability and long service life when exposed to significant loads.

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At the same time, it is important at the present stage to increase the competitive advantages of new structural products that replace traditional metallic materials. These requirements are met by aluminum matrix composites (AMC) with a carbon-containing based dispersed phase, in which the targeted regulation of the composition and improvement of the methods for manufacturing composites allows reaching a fundamentally new level of performance properties and low cost [4-7].

With a high-volume fraction of reinforcing particles for composites, the Al-SiC, Al-C systems with carbon of various structures, Al-TiC, Al-B_4C exhibit high contact strength, due to which materials of various fields of technology can be made ~~from~~ them [8-10, 13-18].

There is a significant reserve for further improvement of the properties of particulate filled composites through the development of nanotechnologies and the implementation of the principles of transformation hardening, which are especially effective for heterophase systems, which include composites: the development of methods for introducing reinforcing components of various nature, volume content and size into a metal matrix, including mechanically activated modifying nanophases; creation of hybrid composite due to poly-reinforcement, functional reinforcement; regulation of the matrix alloys composition; the use of ultrafast hardening in order to obtain an amorphous state and the formation of submicrocrystalline structures during subsequent heat treatments [4, 5].

An effective way to further improve the service properties of composites - high-temperature strength, rigidity, bearing capacity, wear resistance, as well as reduce the cost and improve the manufacturability of composites can be the implementation of the reinforcing components synthesis directly in the manufacturing process of composites. Thus, the improvement of methods for the synthesis of dispersion-filled composites in order to achieve the desired performance properties is an urgent task. Carbon-containing substances (carbon, carbides, etc.) are actively used to obtain such composites [13-18]. Especially actively carbon-containing substances began to be used with the development of research in the field of nanostructures and nanostructured materials [10,13-16]. Since the 1990s, works have been published showing that the introduction of carbon-containing nanostructures into a metal matrix in order to strengthen it can significantly improve the properties of composites and expand the scope of their application [17-18].

From the analysis of the scientific literature, we can conclude that at present, the existing methods for the synthesis of structural composites do not allow achieving a uniform distribution of the dispersed phase in the bulk metal matrix [3-6]. The lack of a uniform distribution of the dispersed phase prevents the achievement of the maximum effect in the mechanical properties of the resulting structural composite [1-5].

The review analyzes the current state of research in the field of synthesis of carbon-containing materials, as well as composites with a carbon-containing dispersed phase, and their practical application.

2. Carbon-containing compounds, their synthesis and application in the technology of obtaining composite functional and structural materials

To date, various forms of carbon nanostructures are known, including fullerenes, graphene [19,20], carbon nanofibers (CNFs), carbon filaments and nanotubes (CNTs) [21-24], as well as various carbides [6-10] (Fig.1).

Initially, we will consider publications on graphene-based carbon-containing substances, which reflect the production of functional composites, and then we will move to consideration of the production of structural composites based on other carbon-containing substances and a metal matrix.

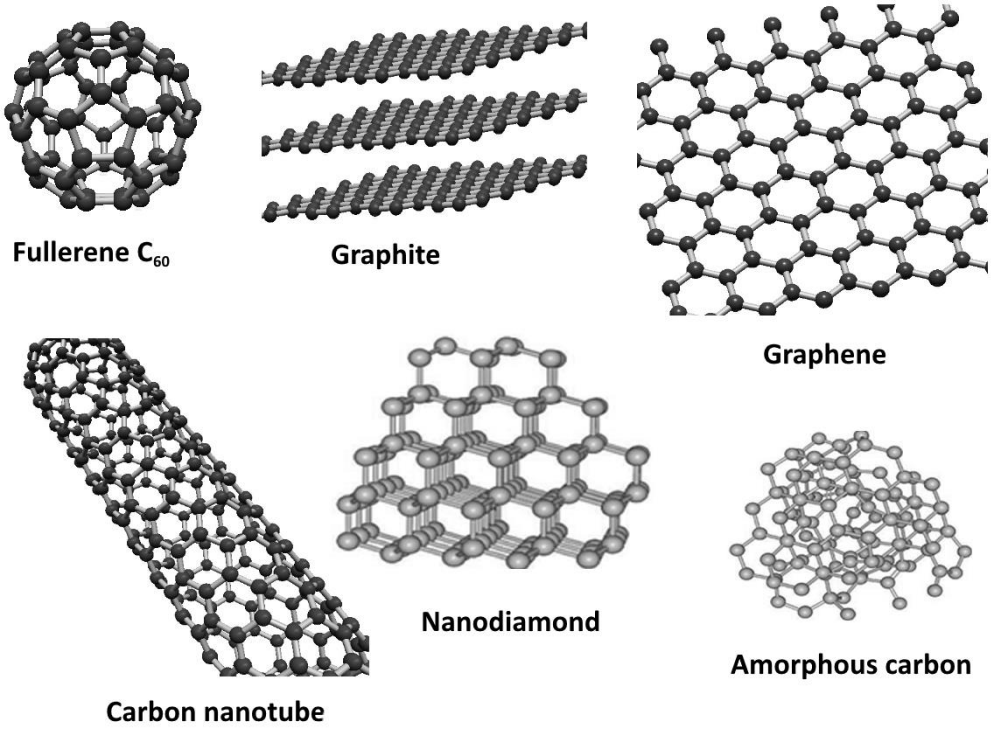


Figure 1. Different forms of nanocarbon.

2.1. Carbon-containing functional composites based on graphene

The active interest in graphene is due to the search for new functional film (2D) materials and technologies based on them, aimed at cost-effective, large-scale production of electronic components and flexible electronics devices [25-30].

Graphene is a single layer of carbon atoms tightly packed in a benzene ring structure. The graphene model is widely used to describe the properties of many carbon-based materials, including graphite, fullerenes, nanotubes, and others [30-33]. An overview of graphene synthesis techniques is shown in the flow chart in Figure 2.

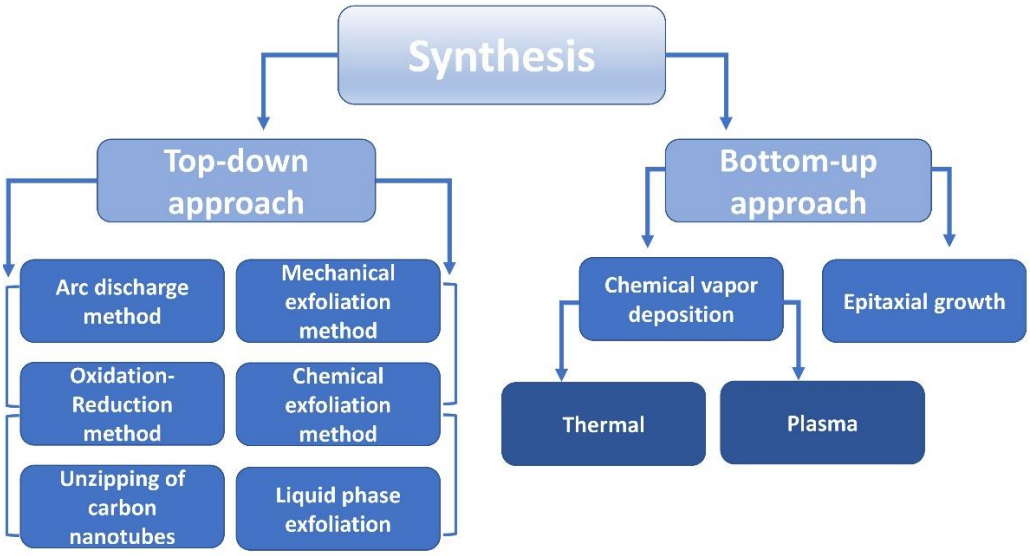


Figure 2. A process flow chart of Graphene synthesis [34].

It was noted [29] that the structure of graphene is responsible for its high specific surface area of ~2630 m^2/g and good electrical conductivity.

The mobility of carriers in graphene films at room temperature, which is due to scattering by impurities or nanowaviness of the crystal, has a huge potential for growth and, according to experimental estimates, is limited by the value of 200000 $cm^2/W \cdot s$, due to phonon scattering [30-35].

Young's modulus or modulus of elasticity of monolayer graphene membranes is 340 ± 50 N/m or E = 1.0 ± 0.1 TPa, and the tensile strength of defect-free graphene is determined as σ ~ 42 N/m. Despite its incredible strength, graphene is flexible and can be subjected to 20% deformation without the crystal lattice breaking [36].

For a suspended monolayer graphene "particle" obtained using mechanical separation, the thermal conductivity was about 5000 1/W K [37, 38].

For graphene, a constant transparency (~97.7%) in the visible range and a transmittance that linearly decreases with the number of layers for n-layer graphene were experimentally found [39]. Opacity increases with graphene thickness, so that each graphene layer adds 2.3% [40-43]. In [44], the authors were able to solve the problem of the instability of two-dimensional crystals and isolation of graphene. In this work, the authors experimentally obtained graphene by mechanical splitting of graphite and investigated its unique properties. The production of highly oriented pyrolytic graphite (HOPG) requires temperatures about 3300 K, while the splitting onto a substrate is carried out at a temperature an order of magnitude lower than the growth temperature; therefore, two-dimensional graphene films are obtained stable [32,45].

In addition, the large specific surface area of 2D materials motivates their use in surface-active applications such as catalysis and sensing. The atomic thickness and high anisotropy provide 2D materials with excellent mechanical flexibility and optical transparency, which provide great opportunities for the development of 2D materials-based optoelectronic devices and wearable flexible devices [46,47]. It should be noted that during the synthesis of 2D materials, the formation of van der Waals heterostructures is assumed, in which different 2D layers with a strong covalent bond in the plane are transferred to each other, and in the vertical direction the layers are held by rather weak van der Waals forces [39].

Nonlinear characteristics at room temperature make these structures interesting for practical applications. Since 2013 works began to appear on heterostructures using layers of transition metal dichalcogenides ($NbSe_2$, MoS_2, WS_2, WSe_2, TaS_2, etc.) [20, 29, 48, 49] (Fig. 3). At present, several types of such heterostructures have been created and studied, and they have already shown a huge potential for the development of this direction [29]. A new version of the lateral transistor was proposed using a tunnel-transparent barrier built into a planar graphene channel [48, 50,51].

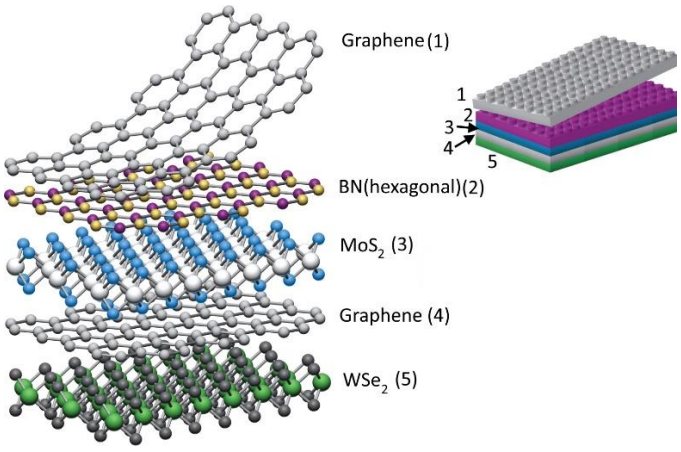


Figure 3. Layer-by-layer formation of a heterostructure based on graphene and other two-dimensional materials [20].

Graphene can be considered as a molecule that can be chemically modified to obtain a new material with a different chemical composition and interesting properties [52, 53].

Depending on the environment, the electronic configuration of the carbon atom (which has four valence electrons) can change significantly, allowing it to bond with other atoms. The combination of carbon atoms with oxygen forms graphene oxide (GO), which can be characterized as a single monolayer of graphite with randomly distributed sp^2 and sp^3 regions containing hydroxyl (–OH), epoxy (=O), carbonyl (–C=O) and carboxyl (–COOH) functional groups. The chemical composition and heterogeneity of GO are still widely discussed [52, 54] (Fig. 4).

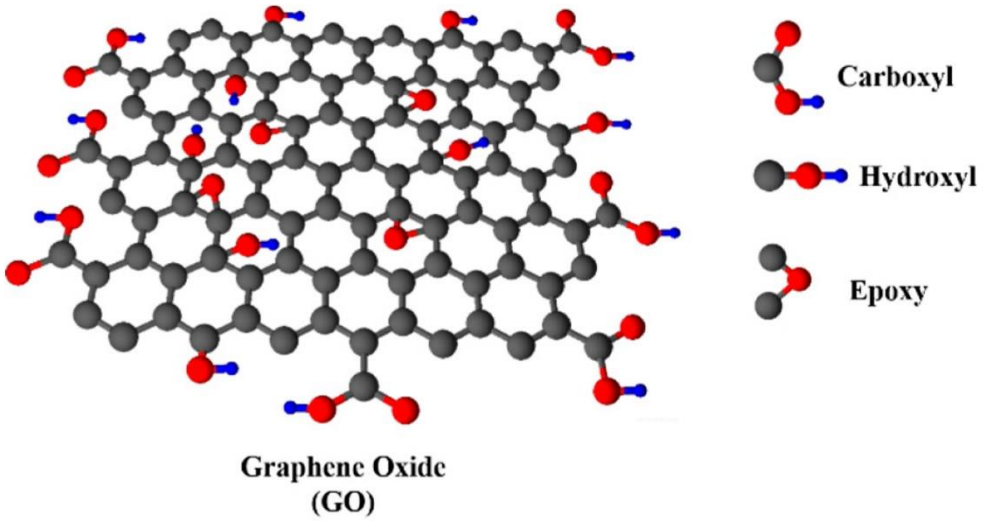


Figure 4. Structure of GO [54].

The optical, electrical, and mechanical properties of graphene are ideal for creating micro- and nanomechanical systems, transparent and conductive electrodes, and photonics. The chemical modification of graphene leads to the materials with a wide range of functional properties, which depend on the type of modification and the degree of functionalization [55]. The synthesis of graphene and chemically modified graphene with various degrees of functionalization makes it possible to create a wide range of new materials that can be used in the rapidly developing field of printed electronics [23].

Next, we consider the features of using carbon compounds to obtain structural composite materials, without considering graphene, which is practically not used in the production of structural materials.

2.2. Carbon-containing substances are promising solids for the preparation of structural composite materials with a metal matrix

Currently, CNFs and CNTs are attracting a lot of attention not only because of their structural features and unique physical and mechanical properties, but also because of their potential technological applications. The use of CNFs and CNTs can be divided into two groups: (i) the first group includes materials using ensembles of carbon nanostructures, such as composite materials (where CNTs and CNFs are used as reinforcing elements), materials for chemical power sources, adsorbents, catalysts, etc. Note that composites containing CNT/CNF have better mechanical properties [6]; (ii) the second group includes materials using individual CNTs due to their high mechanical and electrical properties [7].

CNTs and CNFs can be used to produce both structural and electrically conductive composites [55]. Due to their unique physical and chemical properties, even small additions of CNTs and CNFs to various materials can significantly improve their mechanical and electrical characteristics. Since the 1990s, works have been published suggesting the

introduction of carbon nanostructures into a metal matrix in order to strengthen it, which has significantly improved the properties of composite materials and expanded their scope [56-64]. Theoretical and experimental studies of the mechanical properties of single-walled carbon nanotubes (SWCNTs) are described in many works [5, 62, 65, 66]. The value of the SWCNT and double walled CNTs (DWCNTs) elasticity modulus has a widespread, from 80 to 140 and 140 to 190 GPa, respectively (Fig. 5) [67]. Subsequently, the mechanical properties of CNTs were repeatedly confirmed [68]. Destruction experiments were carried out on SWCNT bundles [65]. The average tensile strength was 30 GPa with a range of values from 13 to 50 GPa. It was found that the load led to bundle separation, while the inner tubes of the bundle did not collapse, but slipped relative to the outer tubes, which were subjected to destruction. In the case of MWCNTs, it was found that the elastic modulus average value of ~1200 GPa [69]. The strength parameters varied from 20 to 90 GPa [70].

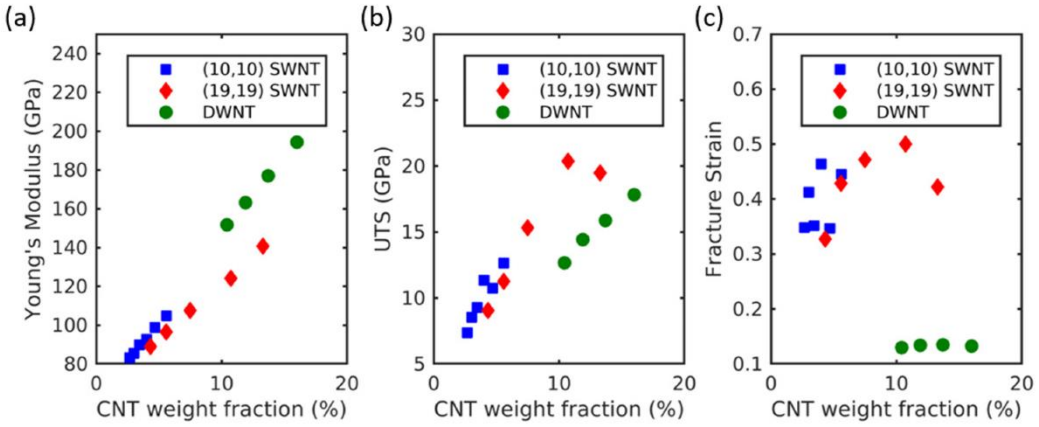


Figure 5. Improvement in mechanical properties of CNT-Al composites as a function of CNT weight fraction: (a) Young's modulus, (b) ultimate tensile strength, and (c) fracture strain [67].

From the point of view of practical use, low density values are extremely important: ~1.4 g/cm^3 for SWCNTs, ~1.8 g/cm^3 for MWCNTs. High heating rates (up to 1000 K/min) reduce the sintering duration, which makes it possible to limit grain growth, and in the case of metal-MWCNT systems, it becomes possible to limit the formation of an undesirable carbide phase during consolidation [71, 72].

The value of the specific strength of CNTs is much higher than that of all known materials [22]. As a result, the thermal conductivity of some SWCNTs is very high [73]. The thermal conductivity coefficient obtained by calculation at 400 K is 2.7 kW/(m·K). This value approaches the thermal conductivity of nanodiamond film (2.2 kW/(m·K) at 310 K) [74]. At ambient temperature, the thermal conductivity of nanotubes is below these values, but nevertheless it is greater than the thermal conductivity of diamond and is equal to 6.6 kW/(m·K). The thermal conductivity of MWCNT measured at room temperature exceeded 3 kW/(m·K), i.e., it has an order of magnitude close to that of diamond, but only in the direction corresponding to the tube axis [75, 76]. The experimentally obtained values of the thermal conductivity of CNTs are noticeably lower than the calculated values: at normal ambient temperature, along the axis of parallel SWCNTs, they exceed 200 W/(m K), which is approximately equal to the thermal conductivity of metals. For materials with randomly arranged CNTs, this coefficient is an order of magnitude lower [22].

The current density of SWCNTs can theoretically reach 10^9 A/cm^2 (according to some data, even 10^{13}) A/cm^2, which is orders of magnitude higher than that of known conductors and superconductors. And the experimentally obtained values of the current density of MWCNTs without defects - 10^7 A/cm^2, when using contacts made of Au and Ga - $2 \cdot 10^8$ A/cm^2. The electrical conductivity of MWCNTs depends mainly on the properties of their outer layer [77].

2.3. Basic methods for the synthesis of carbon nanotubes

The main modern methods for the synthesis of CNTs are the discharge arc method and methods of deposition from liquid or gaseous media using organometallic catalysts [78,79]. In the discharge-arc method, carbon particles are removed from the graphite electrode in an inert gas flow in an arc discharge plasma and deposited in the form of honeycomb structures on the cathode surface. In this case, mainly MWCNTs are obtained. The method of deposition of CNTs from liquid media (for example, acetone, alcohol) is the decomposition of a mixture of a carbon-containing compound and a catalyst (iron acetates, cobalt acetates) in an argon flow at temperatures of 600–800 °C. It is possible to obtain SWCNTs in the resulting soot with a low yield. When obtaining nanotubes from the vapor phase (CVD method), a hydrocarbon (methane, ethylene, acetylene) is passed over a metal-containing catalyst at temperatures of 500–800 °C. The method is widely used, but has several drawbacks, including the need to maintain high temperatures, as well as the low yield of nanotubes. One of the most promising methods for the formation of structures with a developed surface morphology is the metalorganic chemical vapor deposition (MOCVD) using volatile organometallic compounds (metallocenes) and hydrocarbon precursors [80,81]. The use of this method makes it possible to avoid catalyst passivation and brings the pyrolysis process as almost continuous. In the world, among the variety of methods for synthesizing CNTs, the MOCVD technology is the most widespread [82,83].

An important stage in the control of chemical properties is the CNTs functionalization. In composites and other systems, an important role is played by the uniform distribution of nanotubes over the matrix bulk. The high tendency of nanotubes to form clusters, so-called agglomerations, containing up to several hundred CNTs, creates significant difficulties in their dispersion into the material matrix. CNT agglomerations can take the form of a bundle, a bundle, a coiled ball, and so on [22]. Most of the works on the preparation of composite materials with polymer [84, 85], metal [86–91], and ceramic [92, 93] matrices contain results confirming the expediency of the decision to apply additional processing (functionalization) of CNTs for the purpose of chemical grafting of polar groups to the surface. This treatment makes it possible to obtain a more stable dispersion medium in polar solvents, to increase the chemical activity of the surface, and to increase the bond with the matrix [21, 94]. The most common are oxidative functionalization methods with the formation of oxygen-containing groups covalently bonded to the CNT surface – carboxyl, hydroxyl, aldehyde, ether, ketone, anhydride, lactone (–OH, –C=O, –COOH) [21, 86, 94-99]. At the same time, functionalization by oxidation often leads to a decrease in some of the tubes: a decrease in their diameter and length, opening of the ends, and to the appearance of carboxylated fragments on the side surfaces. Thus, high physical and mechanical characteristics, low density and nanosized particles make CNTs a promising reinforcing component.

3. Advantages of composites over traditional materials

The main advantages of dispersion-strengthened composites over traditional materials include the optimal combination of strength and ductility, high wear resistance, and satisfactory manufacturability [100,101]. The materials combine valuable mechanical properties with the required operational properties, have a density 2.5 and 3 times lower than steel and bronze. As a result, the formation of a set of properties that is as close as possible to the required one is ensured, the possibility of manufacturing both structurally homogeneous isotropic parts, and functionally reinforced and layered compositions. The use of composite materials ensures the reliability of the structure during operation; weight loss; a significant reduction in the cost of products. In modern technology, the simultaneous development of fundamental sciences, technical disciplines and tribology has made it possible in practice to create friction units of the highest quality, operating not only in transport and technological machines, but also in extreme conditions, high and low temperatures, in vacuum, with radiation, in chemically aggressive environments on earth and

in space. However, even today there are problems of creating friction units that are optimal in terms of characteristics, and scientific achievements in this area are far from perfect. The issue of reducing energy consumption for friction, increasing the durability and reliability of tribocouples is very acute [102-104]. In this connection, work on the creation of fundamentally new antifriction materials with a heterophase structure, including metal composites, in which highly plastic metal matrices and refractory high-strength high-modulus fillers are artificially combined, is becoming important. The advantages of aluminum alloys as composite matrices are high thermal conductivity, heat capacity, high technological properties and lightness. Aluminum based alloys are three times lighter than copper-based alloys. The addition of particles does not significantly increase the mass, strength, fracture toughness and load-bearing capacity of the material. The particles of carbon nanotubes and silicon carbide are chemically stable in the aluminum matrix and effectively inhibit the movement of dislocations and thereby increase the strength of the alloys and slightly change its specific gravity. Composite systems "aluminum alloys-ceramic particles" have 2-3 times lower density than bronze, have an optimal combination of strength and ductility and have sufficient corrosion resistance. They combine high wear resistance with the required operational and mechanical properties [102, 103, 105-107]. Reinforcing fillers with a hardness that differs sharply from the matrix not only increase the wear resistance of the composite, but acting as supporting supports, reduce the friction coefficients in a wide range of loading parameters [105,106]. The combination of a plastic matrix and high-strength fillers suggests a significant increase in the bearing capacity of bearing materials and an extension of the temperature ranges of work [106]. Good casting properties, cutting capabilities and weldability characterize the manufacturability of dispersion-hardened composites based on aluminum alloys. The attractiveness of the material is increased by the possibility of varying mechanical properties by choosing alloying systems and modes of subsequent thermal and thermomechanical treatments. One of the distinguishing features is the possibility of creating composite surfaces on substrates of different materials. The most important advantage of composites is the ability to create structures with predetermined properties that most fully meet the nature and conditions of work.

3.1. Interfacial interaction of composite components

The interfacial interaction of the composite components sets its physical and mechanical properties: strength, hardness, fracture toughness, heat resistance, and others. When controlling the process of interfacial interaction for composites, one should consider the conditions for the compatibility of components and the stability of the interface between phases [103, 105, 108, 109]. When obtaining composites, where fibers are used as a hardener, the interfacial interaction must be limited, since intense interaction leads to a deterioration in the final physical and mechanical properties. At high temperatures, chemical reactions can occur at the composite interface. Under the condition that a chemical reaction occurs on the surface, the bond between the reinforcing component and the matrix becomes stronger. But if the layer is thicker, the reaction products can, on the contrary, weaken the bond, up to the destruction of the fiber. Chemical interaction can occur both during the composites' preparation and during their high-temperature treatment [63, 104, 106]. From the point of view of boundary processes, there are three classes of fibrous composite materials [4, 5], when the matrix and fiber: (i) are not mutually reactive and soluble, that is, they do not form solid solutions and chemical compounds; (ii) are not mutually reactive, but soluble, then they form solid solutions, but do not form chemical compounds; (iii) react with the formation of a chemical compound at the interface. Changes in the chemical composition occur at the phase boundary. The main task of the interface is to ensure the connection between the matrix and the hardener. So, the mechanical properties of the boundary region are very important, in particular, stability under various loading methods. In this area, the following types of bonding are distinguished [6]:

1) Mechanical bonding due to friction forces. Such materials have low strength in transverse tension and longitudinal compression (composites of the first class).

2) Bond due to surface tension forces. It is typical for materials formed during wetting and dissolution without a chemical reaction. When wetting, a slight dissolution of the components occurs when the fibers are impregnated with a molten matrix (composites of the first and second classes).

3) Reactive bond during a chemical reaction at the interface, which results in the formation of other chemical compounds (composites of the third class).

4) Exchange-reaction bond - a kind of the reaction bond, in which a chemical reaction does not occur simultaneously, but in several stages.

5) Oxide bond - a kind of the reactive bond, characteristic of composites reinforced with fibers or oxide particles. In composite materials with metal matrices, bonding is achieved through the formation of spinels at interphase boundaries or through an oxide film through which bonding occurs. The interaction between the metal and SWCNTs, MWCNTs with open ends can occur in this way [109, 110].

Contact interaction at the open ends of CNTs occurs due to the strong σ-bonds formation, which can lead to a reaction with the formation of metal carbide. The possibility of chemical interaction of CNT and fullerene with metal surfaces was experimentally shown in [108-111]. The calculations showed that the binding energy of an Al atom with an ideal surface of an unmodified SWCNT did not exceed 0.4 kJ/mol or $4 \cdot 10^{-3}$ eV due to weak π-bonds. The presence of oxygen-containing functional groups on the CNT surface has a significant effect on the interaction with some metal atoms [108, 109]. Firstly, metals create a covalent bond with oxygen, and secondly, groups containing oxygen create conditions for the convergence of the energies of the valence orbitals of metal and CNT atoms, contributing to their greater overlap. The interaction of an Al atom with a SWCNT is due to the formation of a covalent bond with the carboxyl functional group and directly with the SWCNT surface [112]. In practice, the surface of nanotubes is not ideal, therefore, depending on the degree of purity of CNTs, the number of defects and grafted functional groups on their surface, the formation of metal carbide is also possible, which occurs due to the substitution of carbon atoms. Variation of the Gibbs free energy of formation of various carbides (per mole of graphite) depending on temperature. According to the experimental data [113-115], aluminum is practically insoluble in carbon, and the solubility of carbon in aluminum does not exceed 0.02–0.04 wt. %. The main reaction that determines the interaction of carbon structures with Al is the formation of Al_4C_3 carbide. The interactions of amorphous (glassy carbon), quasi-crystalline (carbon fibers) and crystalline (graphite, nanotubes) carbon structure with Al [116, 117] showed that aluminum carbide is formed at 970–1370 K at the time of isothermal contact of carbon materials with liquid aluminum for three or more minutes. When obtaining composites with dispersed or fiber hardeners, the main tasks are to achieve a uniform distribution of the hardener in the matrix and the formation of a strong contact at the interface between the phases of the components. At present, there are many methods for obtaining composites based on a metal matrix with an arming component in the form of MWCNTs: melt processing (casting, melt infiltration), gasothermal spraying (plasma spraying, cold spraying), electrochemical, (electro- and chemical deposition) new methods (powders mixing at the molecular level, nanoscale dispersion, sandwich processing, laser deposition), etc (Fig. 6). However, the most accessible and widely used methods for obtaining metal matrix composites reinforced with MWCNTs are solid-phase methods of powder metallurgy (PM) [88, 104, 118, 119]. A typical technological process of PM method consists of mixing of components and their consolidation. Mixing is usually carried out using mechanical alloying in mills of various types or ultrasonic processing of components in liquid media [61]. Consolidation methods based on the sintering of metal powders can be divided as follows: (i) - conventional sintering, (ii) - hot pressing. The use of this technological scheme, as a rule, leads to a chaotic arrangement of MWCNTs.

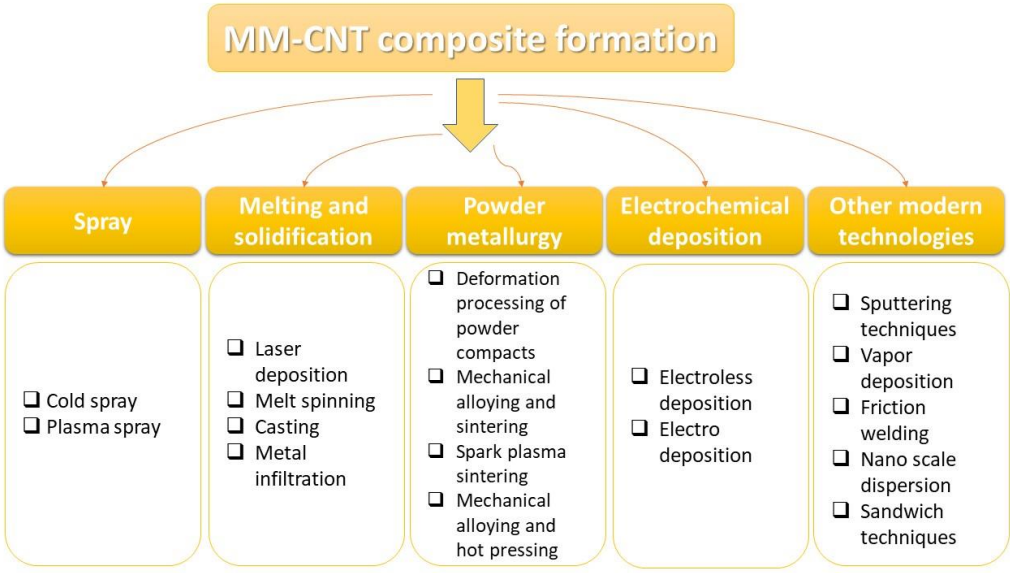


Figure 6. List of different processing routes for MM-CNT composites [5].

In [120, 121], MWCNTs were grown on Al powder (Ni catalyst). The resulting powder was pressed at 600 MPa, sintered at 640°C for 2 hours, and additional pressing at 2 GPa was carried out to achieve the required density and properties. Microhardness increased from 0.15 GPa to 0.65 GPa, tensile strength from 140 MPa to 398 MPa. It is also reported that the high increase in strength is associated with a strong interfacial bond in the form of a thin layer of Al_4C_3 between the components (Fig. 7). Ag-based composites reinforced with MWCNTs were prepared by mixing powders in a mortar and compaction at 320 MPa, followed by sintering at 700°C for 1 h [122]. In order to increase the adhesive bond with the matrix, the MWCNTs were treated with acid to roughen the surface. Due to insufficient density after sintering, the composites were pressed again at 400 MPa for further compaction. The authors stated that the uniform dispersion of MWCNTs without the formation of any clusters was achieved up to 8 vol. % MWCNTs in the composite, after sintering at 700°C [122]. Most studies on sintered metal MWCNT composites indicate insufficient density, which can be increased by reducing the particle size of the matrix powder, improving the uniformity of the distribution of nanotubes, or by using additional deformation treatments [89]. In hot pressing, powder mixtures or cold pressed compacts are processed at high temperature in a mold in a protective environment. Under the combined influence of temperature and pressure, the material is easily deformed, which contributes to the achievement of high density. Al-MWCNT composites were prepared by hot pressing powder mixtures at 520°C and 25 MPa [118] with a holding time of more than 30 min. The authors of [123, 124] obtained Al-MWCNT composites by ball milling of raw materials for different times. The Al particle size was ~13 μm, that of MWCNTs was l ~ 5 μm, and d ~ 10–20 nm. Subsequently, the mixture was subjected to hot pressing at 560°C. It is shown that with an increase in the milling time, the degree of damage to MWCNTs increased, which was noted by an increase in the intensity of the Al_4C_3 phase. Tensile strength and yield strength for the composite Al-0.5 wt. % CNTs (6 h grinding), compared with pure Al, amounted to 42.3% and 18.4%, respectively. Hot pressing has the advantage of producing high-density (>95%) metal-MWCNT composites. The hot-pressing time to achieve the optimal density in most studies was ~1 h. Long-term temperature exposure can lead to the metal carbide formation, depending on the quality of the MWCNTs used. Some of these studies have found a reaction between MWCNTs and metal [124, 125], while others have followed the stability of MWCNTs. At present, an effective method of consolidation is spark plasma sintering (SPS), the basic principle of

which is to heat the powder material by passing a pulsed current with simultaneous application of pressure [126-128].

Another paper [129] provides a general view of the strengthening mechanisms of Al-CNT nanocomposites fabricated using 1 vol. % CNT using the powder metallurgy method using ultrasonic treatment and its combination with a ball mill (Fig. 7). It is shown that this approach results in the formation of a homogeneous dispersion of CNTs with minimal damage. In addition, Al_4C_3 were found, formed during the preparation of composites, which play an important role in transferring the load from the matrix to the reinforcement. As a result, the authors showed that the presence of Al_4C_3, CNTs, and a high density of dislocations near the latter is the key to improving the mechanical properties of nanocomposites.

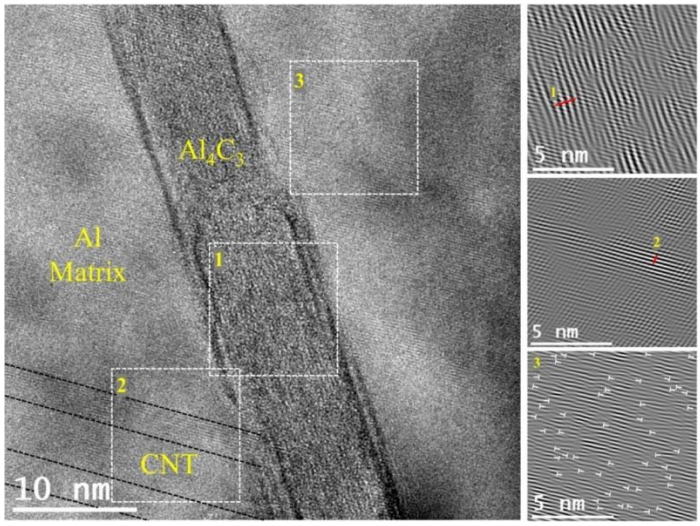


Figure 7. HRTEM image and inverse FFT showing the Al_4C_3 and CNT embedded in Al matrix [129].

Carbon nanofibers and nanotubes are attracting a lot of attention not only because of their structural features and unique physical and mechanical properties, but also because of their potential technological applications. In general, the use of CNFs and CNTs can be divided into two groups. The first group includes materials using ensembles of carbon nanostructures, such as composite materials (where CNTs and CNFs are used as reinforcing elements), materials for chemical power sources, adsorbents, catalysts, etc. Note that composites containing CNT/CNF have the best strength-to-weight properties among other materials [84, 130]. The second group includes the use of individual CNTs due to their high mechanical and electrical properties [84, 130].

Table 1. Mechanical properties of carbon materials [84, 130].

Characteristic	Graphite	Carbon fibers	MWCNTs	SWCNTs	Steel
Tensile strength, GPa	100	3-7	300-600	300-1500	0.4
Modulus of elasticity, GPa	1000	200-800	500-1000	1000-5000	2000
Maximum tensile elongation, %	10	1-3	20-40	20-40	26

Table 1 shows that the properties of MWCNTs are slightly lower than those of SWCNTs, since destruction will begin with the outer, defective, layer.

There are much fewer studies aimed at reinforcing the ceramic matrix of CNTs, and metal-CNT composites have been little studied.

3.2. Modeling of the hardening of an aluminum metal-matrix composite reinforced with CNTs

The observed hardening of metal matrix composites is the result of the interaction of several hardening mechanisms:

1) due to the effective transfer of the load between the matrix and the reinforcing elements;

2) due to the internal thermal stresses caused by a mismatch in the coefficients of thermal expansion between the matrix and carbide particles;

3) by grinding the grain of the matrix (Hall-Petch law);

4) due to a mismatch between the elastic moduli of the matrix materials and the inclusion;

5) hardening caused by the dispersed phase according to Orowan;

6) hardening caused by the appearance of an interfacial layer between the reinforcing particle and the matrix.

The mechanisms that are implemented in each particular case, are determined both by the geometry, size and concentration of particles, and by the conditions for introducing reinforcing particles into the matrix.

The existing methods for modeling the hardening of metal-matrix composites can be conditionally divided into 2 groups

- micromechanical modeling based on empirical or semi-empirical models of hardening mechanisms.
- computer modeling, including modeling by molecular dynamics methods, finite element modeling, multiscale modeling, representing some combination of molecular dynamics and finite element modeling.

The main problems affecting the MMC hardening are the uneven particles distribution in the bulk, the aggregation of reinforcing particles, and the formation of defects at the particle-matrix interface. One of the important tasks is to prevent these problems [103, 131, 132].

Unlike macroscale descriptions of hardening mechanisms such as the Hall-Petch or Orowan ratios, where the differential increase in strength associated with a single hardening mechanism is added to the base strength of the material, the microscale considers dislocations to interact directly with various obstacles independently and uses the absolute critical allowed shear stress for each type of obstacle.

Casati R. et al. [62] reviewed the theoretical and experimental foundations related to bulk MMCs and the main results achieved in this area. The structural properties and mechanical characteristics caused by the addition of nanoparticles and nanotubes to base metals are presented, and modern methods for the synthesis of MMCs are described.

Kun X. et al. [133] the mechanisms of MMCs hardening with CNTs-reinforced Al matrix, including load transfer, dispersion strengthening, dislocation formation caused by thermal mismatch, fine-grained hardening, etc., have been studied.

Xu R. et al. [134] developed a microstructure-based work hardening model by modifying and combining the Cox-Mecking approach to dislocation density evolution, reverse stress models for grain boundaries, and reverse stress models for second phases. The expected strain hardening characteristics of the composites were successfully matched with the experimental results. It has been shown that the effects of CNT reinforcement and grain refinement can significantly increase the strength of composites. Authors in [135, 136] discussed the formation of coated CNTs (there is an amorphous interfacial surface between the reinforcement and the metal matrix), which have a great influence on the force acting on dislocations. This article proposes a model of a three-phase compound cylinder with new boundary conditions. This model studies the interaction of CNT edge dislocations with a coating containing the interface effect.

Liu Z.Y. et al. [137] using Tsai-Halpin' equation showed that with an increase in the concentration of CNTs in MMC, the elastic modulus of composites increases. The tensile strength of composites is significantly increased compared to the matrix alloy, which is explained by the mechanisms of load transfer, grain refinement, and misfit dislocation due to the addition of CNTs. Modifications are proposed to consider the mechanisms of

hardening according to Orovan, due to the redistribution of the load and the refinement of the grain of the matrix material, as well as the discrepancy between the thermal expansion coefficients. The simulation results are in good agreement with the experimental ones.

Dong S. et al. [138] proposed a dislocation-based model to investigate the synergistic effect arising from the grain size of the matrix and the size of the reinforcement on the strengthening mechanisms of nano/ultrafine-grained CNT-reinforced MMCs. The model based on the three-phase composite material approach is suitable for studying the elasto-plastic stress-strain relationships of MMCs. The overall flow strength of nano/ultrafine grain MMS depends on the grain size of the matrix or CNT, as well as the volume fraction and microstructure of the reinforcement. It is assumed that dislocations formed by a mismatch in the thermal expansion coefficient are redistributed only in a strongly dislocated volume that surrounds a separate inclusion. This zone as a matrix phase in the calculation model has a higher strength than the rest of the metal matrix. Therefore, the size of the zone is an additional scale of the microstructure and requires more attention. All these results are explained by the strengthening effect that is due to thermal expansion mismatch weakening as the grain size of the matrix decreases, which leads to the less pronounced role of reinforcement in the overall response of MMC. The model can provide some insight into the strengthening mechanisms of CNT-reinforced nano/ultrafine-grained MMCs.

Chen B. et al. [139] studied the influence of the length-to-diameter ratio of CNTs for hardening aluminum MMCs. To this end, aluminum samples reinforced with CNTs with different aspect ratios were fabricated by three different powder metallurgy methods. The microstructural study showed that CNTs were uniformly dispersed in materials ranging in aspect ratios from 6.5 to 55. Tensile results showed that CNTs exhibited a strong strengthening effect in composites regardless of their aspect ratios. Post-load studies and quantitative analysis demonstrated a transient hardening mechanism for CNTs that is closely related to the aspect ratio or length of the CNTs. The origin of such a transition has been studied from the point of view of the interaction of a dislocation with a CNT under load. These data may provide new insights into the enhancement behavior of CNT-reinforced MMCs [140].

Zhou M.Y. et al. [141] studied the properties of the matrix composite AZ61 reinforced with microscale SiC and CNTs. Ultra-high strength and good ductility (tensile strength: 412 MPa, yield strength: 345 MPa and elongation: 8.0%) were achieved in the AZ61-5SiC-0.5CNTs composite. The greatly improved composites strength is mainly due to the synergistic effects of grain refinement, load transfer, and increased dislocation density in the matrix due to thermal mismatch, while good ductility can mostly be attributed to a weakened base texture.

It is noted that the formation of clusters and damage to CNTs during processing can reduce the strengthening effect of CNTs. The distribution of a part of the CNTs along the grain boundaries leads to a weakening of the strengthening effect of the Orowan mechanisms. The weakening of the base texture due to the addition of hybrid reinforcement favorably affected the ductility, however, the weakening of the base texture also compensated to some extent the effects of hardening [131].

In [142] Choi B. et al. carried out molecular dynamics (MD) modeling study of the mechanical behavior of an aluminum composite reinforced with CNTs with different diameters. Stress-strain curves under tensile loading were obtained, and variations in the curves were analyzed using MD images, which allow a detailed analysis of the behavior of atoms. It is shown that Young's modulus and impact strength increased significantly as the reinforcement of CNTs increased.

In [132] Zhou X. et al. the mechanical behavior of MMCs reinforced with Ni-coated CNTs has been studied by MD simulation. The results show that the Young's modulus of Ni-coated SWCNT/Mg is obviously larger than that of uncoated SWCNT/Mg. The results also show that the interfacial bonding of a SWCNT/Mg composite can be dramatically

increased by adding a Ni coating to improve the wettability of the nanotube surface and the Mg matrix. In addition, the effect of the amount of Ni coating on the interfacial bonding characteristics of SWCNT/Mg composites was studied. In [143] Meguid S.A. by MD using the LAMMPS software package, CNT pulling tests from a matrix are simulated to determine the effect of various parameters that affect the strength on the interfacial shear of metal matrices reinforced with CNTs. The study focuses on the influence of cell size, cell geometry, and potential features assumed in MD simulations.

In [144] Zhang J. F. et al. the influence of the choice of a representative element, the division of a representative element into finite elements, and the interface layer near inclusions on the deformation curves of the metal composite is estimated. It is shown that simplified models can be used in estimation calculations (the calculation error in this case is no more than 2%).

In the work [145] the experimental data for CNT-Al composites were compared with the data of MD and modified rule of mixtures (see Figure 8). Due to dispersion problems observed in experiments, experimental data are given for CNT volume fractions up to 10%. Experimental data showed a large scatter, as the manufacturing process and distribution of CNTs affected the mechanical properties. However, the MD data are generally consistent with experimental data.

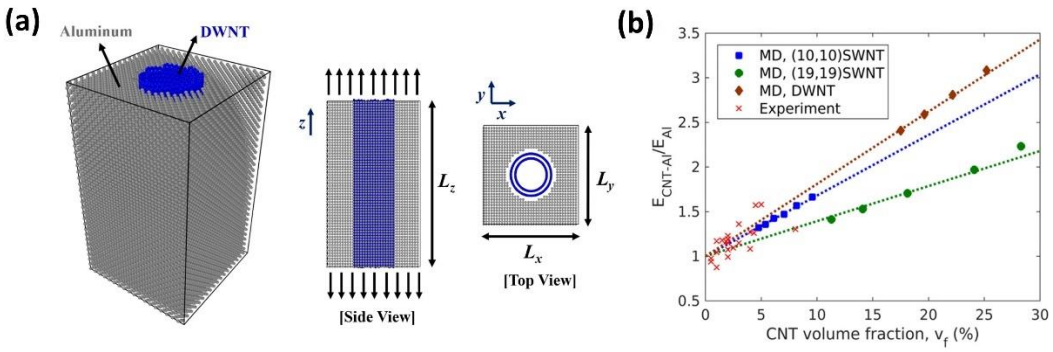


Figure 8. (a) CNT-Al composite in representative volume elements for the MD simulations. Lz =124.44 Å and Lz=97.25 Å for DWNT-Al and SWNT-Al, respectively. Lx and Ly were adjusted from 44.5 to 89.1 Å according to the CNT volume fraction and CNT size; (b) The variation of the Young's moduli of CNT-Al composites with CNT volume fractions [145].

In [103] Song S. et al. the effect of Ni-plated carbon tubes on the strength and deformation properties of a metal composite with an aluminum matrix has been studied. It is shown that the main mechanisms of hardening are Orowan dispersion hardening, which is due to the small diameters of CNTs (8 and 15 nm), hardening generated by a mismatch in the coefficients of linear expansion, as well as hardening due to redistribution of the load between the matrix and reinforcing elements. It has been established that the aspect ratio of CNTs plays the most important role in the strengthening of composites, ratio of length to transverse dimension. Hardening analysis was based both on a micromechanical model, and by the finite element method.

The review by Jagannatham M. et al. [146] provides an analysis of publications devoted to modeling the properties of metal composites reinforced with CNTs. The sensitivity of the simulated properties to several factors is expected, such as the curvature and orientation of CNTs, the presence of reaction products at the interface, changes in the length of CNTs, etc. To compare the calculated properties with experimental ones, a three-dimensional microstructure of the composite is required, which is impossible due to the small size of CNTs. display using available tomography methods. It is shown that the presence of aluminum carbide Al_4C_3 slightly increases the Young's modulus of the composite.

Ahmadi M et al. in [147] and Pérez L. et al. in [148] were to investigate the effect of the Al_4C_3 interfacial surface on the elastic modulus of aluminum matrix nanocomposites reinforced with randomly distributed CNTs using a numerical micromechanical model based on the finite element method (FEM). In addition, the influence of the volume fraction and diameter of CNTs on the elastic modulus of nanocomposites with an aluminum matrix was studied. The results show that the Al_4C_3 interfacial surface significantly affects the elastic modulus of nanocomposites with an aluminum matrix, especially with a decrease in the CNT diameter. With an increase in the thickness of the interfacial boundary, the modulus of elasticity of the nanocomposite increases. The applicability of the presented model for predicting the elastic modulus of CNT-reinforced aluminum-matrix nanocomposites is investigated by comparing the results of the FEM with experimental data available in the literature.

In [149] Dong S. et al., the modulus of elasticity and the distribution of local stresses in MMS with a nonrectilinear shape of CNTs are studied using the FEM and a micromechanical model. It is shown that the results of modeling by the finite element method are close to the results obtained based on the micromechanical model and experimental data.

It is concluded that the effect of CNT waviness on the elastic properties of MMCs reinforced with randomly distributed CNTs is small.

The paper by Su Y. et al. [150] attempts to experimentally and numerically investigate the relationship between microstructure and mechanical behavior of CNT/Al composites. Three-dimensional (3D) computer structural modeling of CNT/Al composites is performed in which the size, morphology, orientation, location, and volume fraction of CNTs are reproduced to be similar to the microstructures of real microstructures of CNT/Al composites.

Based on the developed models of CNT/Al composites, the strengthening of the mechanical properties of the constituent materials of CNT/Al composites, as well as reasonable loads and boundary conditions, is studied. The tensile mechanical behavior of CNT/Al composites has been numerically evaluated and experimentally confirmed. MMC simulation results show that the improved mechanical properties of CNT/Al composites can be attributed to three factors: CNT hardening, matrix grain refinement, and layered architecture. Using the microscopic structural modeling techniques presented here, the influence of model size, interfacial behavior, volume fraction of CNTs, and layered structures on the mechanical behavior of CNT/Al composites can be reproduced in order to understand the mechanisms of hardening and deformation of CNT/Al composites.

4. Conclusions

It should be noted that in the traditional synthesis of a composite material, the carbon material used as a dispersed phase must first be synthesized, purified, functionalized, and only then introduced into the matrix. However, even this procedure may not lead to the expected increase in the mechanical properties of the composite due to poor bonding between the carbon substance and the metal. It should be noted that the development of new structural metal reinforced with high-strength dispersed fillers (nanoparticles, nanofibers, tubes) occupies a significant place in the work of researchers. Replacing monolithic traditional materials with composite materials makes it possible to increase the reliability and weight efficiency of structures. Composite materials based on light aluminum alloys reinforced with particles, due to their high antifriction characteristics in combination with a low specific gravity, high operating temperatures (up to 0.8-0.9 of the melting temperature of the matrices), are very promising materials.

In this regard, at present, along with the traditional direction of the search for new solid substances and materials (the synthesis of solid substances with an equilibrium crystal structure), the synthesis of solid chemical compounds and materials based on their metastable states is attracting more and more attention. From the point of view of physical mesomechanics, the design of nanostructured materials will make it possible to obtain

new objects with a multilevel organization, which is necessary for a more correct approach to the study of structural transformation at the nanoscale level.

Using the microscopic structural modeling techniques presented here, the influence of model size, interfacial behavior, volume fraction of CNTs, and layered structures on the mechanical behavior of CNT/Al composites can be reproduced in order to understand the mechanisms of hardening and deformation of CNT/Al composites. MMC simulation results show that the improved mechanical properties of CNT/Al composites can be attributed to three factors: CNT hardening, matrix grain refinement, and layered architecture.

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