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Review Article

Non-Organic Dielectric Layers for Graphene and Flexible Electronics

Abstract

Future electronics technology is expected to develop from rigid to flexible devices, which requires breakthroughs in materials' properties, especially flexibility, in combination with desirable electrical insulating, semiconducting and metallic properties. Recently emerging 2D materials such as graphene are promising for an active conductive layer in a wide spectrum of flexible electronic devices. Developing optimized dielectrics for the graphene active layer is critical for graphene applications. The advances and limitations of qualitatively different traditional dielectric metal oxide layers (high-k dielectrics Al_2O_3, HfO_2, and ZrO_2) used as a gate in graphene field effect transistors on flexible substrates are considered in the first part of the present review. Its second part analyzes properties of novel dielectric materials (h-BN, Y_2O_3, graphene oxide, fluorinated graphene, composite dielectrics, ion gels) used for graphene transistors. Dielectric layers fabricated from fluorinated graphene or in combination with graphene oxide are the most promising graphene based flexible and transparent electronics.

Introduction

Future electronics technology will evolve from rigid devices to bendable, rollable, foldable, stretchable or transparent ones that are wearable like clothes or accessories [1-3]. The first breakthrough in this direction occurred in the form of flexible electronics for a wide spectrum of applications (bio- and medical items, sensors and gadget displays on the textile or clothing electronics and so on). These flexible devices are expected to excel the rigid ones in durability, weight, and comfort. However their development necessitates breakthroughs in materials since in combination with desirable electrical insulating, semiconducting and metallic properties they need flexibility. Recently emerged 2D materials such as graphene, graphene derivates (graphene oxide, fluorinated graphene), hexagonal boron nitride (h-BN), and transition metal dichalcogenides are attractive because of their outstanding electrical and optical properties. Mechanical properties of these materials are different, and only part of them meets the requirements of flexible or stretchable electronics.

Graphene based printed electronics is also a recently emerged and a fast grown field that has attracted large scientific and technological interest for the past few years. Graphene presents great promise as an active layer in wide spectrum of devices of flexible electronics and, first of all, in field effect transistors. Recent reports demonstrate successful realization of graphene field effect transistors (FETs) on flexible or even on stretchable substrates [4-8]. To achieve such applications the development of optimized dielectrics for the graphene active layer is critical (gate and interlayer dielectrics or/and substrate for graphene). The carrier transport in graphene films takes place at the interfaces with the dielectric or the semiconductor; therefore, the quality of such interface and the interaction with nearby dielectric layers (charge carrier scattering) determine the device performance. Nevertheless, the development of dielectric materials that can achieve high-performance device operation, good mechanical properties, and low-temperature fabrication is not well established because the graphene thin film is very sensitive to surface conditions of dielectric layers [9].

In present review, the main materials applied nowadays as dielectric films for graphene based devices fabricated on the flexible substrate using traditional or printed technologies are discussed. The further opportunities for utilizing the graphene derivates such as graphene oxide and fluorinated graphene are also demonstrated.

Traditional materials for gate insulators in graphene FET on flexible substrates

Presently, an indium tin oxide (ITO) is widely used as a transparent conductor for optoelectronic devices. However, ITO has poor mechanical properties; it tends to crack easily or shows defects when strained [3]. For these reasons, the use of graphene has been widely investigated in recent years as a transparent conductor for optoelectronic and photonic applications because of its combination of electrical, mechanical, and optical properties. The conductive graphene films or reduced oxide graphene layers are generally considered as a material for transistor channels or electrodes [10].

Traditional gate insulators SiO_2, Al_2O_3, and HfO_2 have several limitations for use in graphene transistors on flexible substrates, including low-facture strains less than 1%, poor mechanical strength, high growth temperatures, and poor interface between graphene and dielectric layers [3,11,12]. Nevertheless, oxides based high-k dielectric materials, such as Al_2O_3, HfO_2, and ZrO_2, are the most widely used in graphene FETs [13,14]. For example, Lu et al. [15], have demonstrated high-mobility and low-voltage graphene FETs, fabricated on a polyethylene terephthalate (PET) substrate with a high-capacitance natural aluminum oxide as the gate dielectric (evaporation of AL with its oxidation) in a self-aligned device configuration. The high capacitance of the native aluminum oxide and the self-alignment, which minimizes access resistance, yield a high current on/off ratio and an operation voltage below 3 V, along with high electron and hole mobility of 230 and 300 cm^2/Vs, respectively. Moreover, the native aluminum oxide is resistant to mechanical bending and exhibits self-healing upon electrical breakdown.

The use of atomic layer deposition (ALD) with special precursors for high-k dielectrics allows decreasing the growth temperature and partially overcoming the limitation mentioned above. Petrone et al. [16], fabricate FETs on polyethylene naphthalate (PEN) substrate from graphene, grown by chemical vapor deposition (CVD) with a 6-nm gate dielectric of HfO_2 conformally grown by ALD at 150 °C yielding a dielectric constant of $k \approx 13$. Figure 1 demonstrates graphene FET schema and characteristics with and without strain. The source-togate current, I_{sg}, is measured to remain below 0.5 pA over the entire strain range during device characterization, indicating negligible leakage current through the dielectric even at high strain; carrier mobility μ for these flexible FETs is ~1500 cm²/Vs and does not practically change with strain up to 1.75%.

Although mobility remains relatively constant with strain, the position of the Dirac point in Figure 1d with respect to gate voltage V_{gs} is observed to shift with increasing strain. The authors attribute this shift to changes in device electrostatics, related to mobile trapped charges in the gate oxide and at the graphene–oxide interface, as the substrate is flexed. I–V characteristics (Figure 1e) are for the plotted increasing levels of strain, ranging from 0% to 1.75%. Changes in I_d with increasing strain are most likely connected with HfO_2 degradation under applied strain.

FETs fabricated on smooth spin-coated polyimide films fabricated from CVD graphene with gate dielectric from 20-nm thick Al_2O_3 deposited by ALD process exhibit field-effect mobility up to 4930 cm²/Vs and 1130 cm²/Vs for electrons and holes, respectively [17]. Liquid polyimide was spin-coated on a 50-nm thick plasma-enhanced chemical vapor deposited Si_3N_4 sacrificial layer on silicon. Temperature dependent measurements indicate that carrier transport is not limited by intrinsic mechanisms but by charged impurities, surface roughness, and defects, suggesting that further increases in mobility are possible. Thus further improvement of charge transport is achievable with better fabrication processes. The main advantage is the use of additional rigid substrate during the fabrication process. It was demonstrated that the performance of graphene field effect transistors fabricated on flexible substrates is easily degraded by deformation, delamination and shrinkage during the device fabrication [18]. Multiple thermal annealing on graphene devices could be performed without damages to the flexible substrate using a rigid supporting substrate (Figure 2). As a result, a very high performance including electron mobility about 13000 and hole mobility 9200 cm²/Vs could be achieved for graphene FET with the use of Al_2O_3 gate oxide [18]. Results of mechanical tests of the FET given in Figure 2b,c demonstrate some degradation of the carrier mobility.

Additional technological improvements or special design are suggested to enhance the FET operation with Al_2O_3 gate oxide. Multi-finger electrodes are implemented on flexible substrates to strengthen its current drive for FET with 15-nm thick Al_2O_3 [19]. Bendability of these FETs is tested with the bending radius of down to 1.3 mm (strain ~ 4.6%), the devices remain fully functional with less than 8.7 % reduction and no reduction in the electron and hole mobility after repeated bending tests, respectively. Silicon-nitride passivation offers efficient chemical protection over diverse liquids and robust mechanical protection against impacts.

The wrinkled Al_2O_3 layer containing effective built-in air gaps with a small gate leakage current of 10^{-13} A was used for fabrication of the top gated nanotube FET [20]. The resulting devices with a geometrically wrinkled Al_2O_3 dielectric layer exhibit an excellent on/off ratio of ~10^5, a mobility of ~40 cm²/Vs and a low operating voltage less than 1V. The transistors retained performance under strains as high as 20% without appreciable leakage current increases or physical degradation.

In this case zirconium oxide (ZrO_2) is an excellent high-*k* dielectric material with multiple desirable characteristics, including a high dielectric constant (~23), a wide band gap (5.1 – 7.8 eV) and good thermal stability [21]. In our case it was fabricated at high temperatures (~ 1000°C). Although ZrO_2 has higher dielectric constant than the more popular materials (Al_2O_3, HfO_2), materials created at relatively low temperatures (200-300°C) exhibit poor thermal stability [22]. However, as flexible device technology does not support high temperatures, ZrO_2 may be used as gate dielectric.

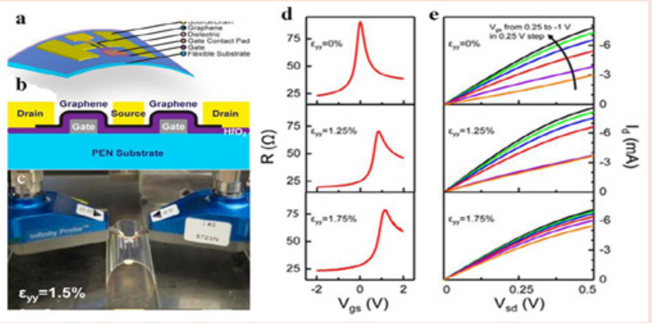

Figure 1: (a) Schematic of graphene FET fabricated on PEN, a flexible and transparent substrate. (b) Cross-sectional schematic of flexible FET device. (c) Photograph of electronic measurement approach for FET under 1.5% strain. (d,e) Low-field transport characteristics of a flexible FET with a device channel width of 30 μm. (d) Device resistance, R, is plotted against gate-to-source voltage, V_{gs}, at a fixed source-to-drain bias of Vsd = 10 mV. (e) Current−voltage (I−V) characteristics plotting drain current, I_d, as a function of V_{sd}. I−V curves are taken at fixed Vgs decreasing from 0.25 V (orange) to −1 V (black) in 0.25 V steps. Data are presented for increasing values of strain of ε_{yy} = 0%, ε_{yy} = 1.25%, and ε_{yy} = 1.75%. Reprinted with permission from Ref. 16. Copyright (2012) American Chemical Society.

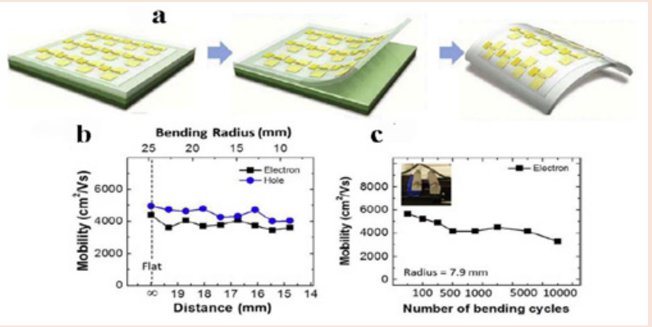

Figure 2: (a) Fabrication process for flexible FET with use of additional rigid substrate. (b) Mobility as a function of bending radius and distance. (c) Mobility as a function of bending cycles. Bending radius was 7.9 mm. Inset shows the automated bending machine with device. Reprinted with permission from Ref. 18.

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The deposition of high-*k* dielectrics is usually achieved using ALD with utilization of reactive surface groups [23]. Functionalization of graphene surface for ALD either introduces undesired impurities or breaks the chemical bonds in the graphene lattice, inevitably leading to a significant degradation in carrier mobilities. Synthesizing ZrO_2 at high temperature, and then transferring them onto graphene through a dry transfer process at room temperature preserves the integrity of the graphene lattice. As a result the top-gated graphene FET with transconductance of 2.0 mS/μm, and carrier mobility of ~1300 cm^2/Vs is obtained [23].

Few attempts have been made to produce ZrO_2 in a solution [24]. But the electrical properties of the film annealed at a low temperature were not encouraging because residual organic particles remained at the dielectric. Hasan et al. [22], demonstrated the use of plasma annealing at a reasonably low temperature and achieved improved dielectric properties such as lower leakage current, higher dielectric constant, and better reliability; and such ZrO_2 layers could be applied in all-printed electronic devices in the near future.

One more very important item for gate dielectrics is a built-in and interface charges. The lower values of these charges the more promising dielectric we have. The standard values of built-in or interface charge density in the widely used Al_2O_3 films are in the range 10^{12}-10^{13} cm^{-2} [25], in HfO_2 they vary within 10^{11}-10^{12} cm^{-2} [26,27], and in the Si/SiO_2Graphene/ ZrO_2 structures they range within (1-15)x10^{11} cm^{-2}’eV^{-1} [22]. Thus, relatively high charge values are observed for Al_2O_3 and HfO_2 films. From this point of view only h-BN discussed in more details below has demonstrated low values of charges. According to the measurements using the atomic force microscopy probe the charge values are ~2-3x10^{10} e/cm^2 [28].

The effect of various dielectrics (SiO_2, Al_2O_3, HfO_2, and ZrO_2,) on charge mobility in single-layer graphene is theoretically investigated by Konar [29]. It is found that though high-k dielectrics can strongly reduce Coulombic scattering by dielectric screening (Figure 3), scattering from surface phonon modes arising from them wash out this advantage (arrays in Figure 3).

Calculations show that the available choice of dielectrics offers not many advantages for improving carrier mobility in actual FET devices at room temperatures.

Promising insulating materials for graphene-based flexible electronics

Hexagonal boron nitride (h-BN) have been used in a top or bottom gate configurations as they can provide an atomically smooth surface that can protect the surface charge traps and the rippling of transferred graphene films [30]. The difficulty in synthesizing high-quality and large area h-BN films, and mechanical properties of h-BN, however, limit its applications on flexible substrates. Perton et al. [31], fabricated flexible graphene field-effect transistors with graphene channels fully encapsulated in hexagonal boron nitride (h-BN). For FETs with channel lengths of 2 μm the authors managed to demonstrate an exceptional room-temperature carrier mobility up to 10 000 cm^2/Vs and a lower mobility for lower channel lengths (for instance, mobility of 2200 cm^2/Vs for channel length of 375 nm). Study of mechanical flexibility has revealed strain limits of 1%. Strain-induced fluctuations in mobility result in less than 3% degradation at ε = 0.5% and less than 13% degradation at ε = 1%. Conductivity exhibits less than 20% degradation from its unstrained value over the entire measured strain range (ε = 0-1%). Mechanical failure of flexible FETs occurs at strains greater than ε = 1%. Failure of flexible FETs may be connected not only with gate dielectric but with metal contacts. Metal electrodes are uniformly stretched to approach their maximum allowable strain without degradation of over 4%, which corresponds to the bending radius of ~1.5 mm [32]. This fact is in good agreement with the results for flexible MoS_2 FETs implementing h-BN dielectrics [33], properties were stable within 13% up to 1.5 % strain. Elastic properties and intrinsic strength of ultrathin (2-5 monolayers) hexagonal BN film grown with use of chemical vapor deposition was examined by nanoindentation and show 2D elastic modulus in the range of 200-500 N/m, and a strain limit of h-BN films greater than 3.0%. [34]. These properties look very promising for applications in the flexible electronics, but structure and properties of CVD grown h-BN are strongly distinguished from h-BN exfoliated from crystals, and only the last one provides high mobility in graphene.

Yttrium oxide (Y_2O_3) is an attractive gate dielectric material for electronic devices due to its high dielectric constant (a relative dielectric constant of κ=10 on graphene) [35]. Since high quality ultrathin Y_2O_3 layers (~ 5 nm) provide very large capacitance on top of graphene surfaces which is comparable to the quantum capacitance of graphene, the top-gated Y_2O_3/graphene devices are considered as ideal structures for exploring the density of states of pristine and disordered graphene [36]. Similarly to other kinds of oxide layers, the electron–hole mobility asymmetry is observed. After Y_2O_3 deposition, however, the graphene samples maintain a high mobility value (~20 000 cm/Vs at cryogenic temperature), which is much higher than those previously reported 1200 cm/Vs for room temperature [37]. Thus the ultrathin Y_2O_3 layers deposited on graphene hardly introduce any interface phonon scattering or resonant scattering centers, which is different from usual behavior of other kinds of oxide layers [36]. In the case of successful fabrication process of

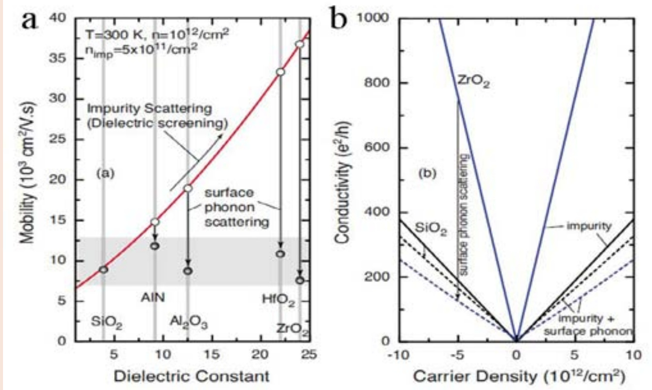

Figure 3: Color online (a) Electron mobility in graphene as a function of the gate dielectric constant. High-k dielectrics reduce Coulombic impurity scattering but strong SO phonon scattering reduces the RT carrier mobility to ~10^4 cm^2 /Vs. (b) Electron and hole conductivity as a function of carrier concentration for graphene on SiO_2 and ZrO_2 Reprinted with permission from Ref. 29.

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ultrathin Y_2O_3 layers, which avoids the growth of precursors normally occurring in the atomic-layer deposition process, the damaging effect caused by plasma in sputtering or by the multi-step lithography and lift-off processes.

The ion gel (for instance, poly(styrene-methyl methacrylate-styrene)triblock copolymer and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide) can be used by means of the solution-based process and exhibit high capacitance of the gate dielectric ~ 5 μF/cm^2 [38]. The ion gel–gated graphene transistors provided both high on/off current and low-voltage operation. Lee et al. [39] presented a stretchable and transparent all-graphene transistor array on a stretchable rubber substrate with using an ion gel dielectric in a low-temperature printing process. Such monolithic graphene devices had hole and electron mobilities about 1190 and 420 cm^2/Vs, respectively, with stable operation of more than 10^3 stretching cycles. Kim et al. [40], reported transparent, flexible graphene transistors and inverters in a coplanar-gate configuration made by a printing process; such devices consisted of only two materials: graphene and an ion gel gate dielectric. These devices exhibited excellent performance, including low-voltage operation with a high transistor on current and mobility, excellent mechanical flexibility, environmental stability, and reasonable inverting behavior upon connecting two transistors. Sire et al. [41], demonstrated graphene transistors created from suspension and operated at gigahertz frequencies. Some parameters of these transistors are as follows: current gain cutoff frequencies achieve 2.2 GHz, power gain cutoff frequencies are 550 MHz, a field-effect hole mobility is 102±19 cm^2/Vs and maintains high stability under bending. It is the highest reported mobility for printed graphene or carbon nanotube inks (~90 cm^2/Vs) [42,43]. Dielectric films used in this study as a gate insulator is yttrium oxide.

Recently, as an alternative, graphene oxide (GO), which can be produced by the oxidation process of graphene in the same way as native oxide of Si, SiO_2, has been exploited as a gate dielectric for graphene-based FETs [44]. This material with good mechanical and optical properties offers a unique advantage for high performance flexible and transparent electronic devices because it can be formed on a graphene channel by solution-based or direct oxidation process at room temperatures [45,46]. In particular, Jeong [47], reported a GO insulator in an electronic device (memory elements) that can be operated with good environmental stability.

High-performance, flexible all-graphene-based thin film transistor was fabricated on plastic substrates using a graphene active layer, graphene oxide dielectrics, and graphene electrodes (Figure 4) [48]. The GO dielectrics exhibit a dielectric constant of 3 -5 at different temperatures, leakage current of 17 mA/cm^2 for 100 nm thick, and breakdown bias was 1.5x10^6 V/cm. Graphene-based EFTs showed a hole and electron mobility of 300 and 250 cm^2/(V·s), respectively. Flexibility of these transistors was tested by bending the supporting PET substrate. The bending properties of the devices are also very good, as expected, due to the excellent mechanical properties of both graphene and GO (Figure 4). Typical transfer characteristics were quite stable under the operation of tensile strains from 0 to 3.5% (corresponding to bending radii calculated using models for this geometry of 4.13 mm) and showed complete recovery after the strain was relaxed [48]. The normalized hole and electron mobility had a distribution of less than 10%

Standley et al. [49], have fabricated transistors comprising single or bilayer graphene channels, graphite oxide gate insulators, and metal top-gates. The graphite oxide layers show relatively high leakage current at room and cryogen temperature (~20 A/cm^2 for 4 nm GO). It is one of the main problems of GO as dielectric layer. Increase in GO thickness leads to weak decrease in leakage current (compared with data for [48]). The breakdown electric field of graphite oxide was found to be comparable to SiO_2, typically~(1–3)x10^6 V/cm, while its dielectric constant is slightly higher, κ ≈ 4.3. The carrier mobility in a FET was found to be equal to ~ 700 cm^2/Vs. Another flexible graphene field effect transistor on polyimide substrate using graphene oxide as top-gate dielectric was fabricated by Jewel et al., [50]. Good current saturation and peak hole and electron mobilities about 500 cm^2/Vs and 160 cm^2/Vs are observed. A maximum transconductance of 0.42 mS and the intrinsic cutoff frequency of 117 GHz are achieved when the gate length is reduced up to 0.25 μm.

Generally, GO have combined an excellent flexibility with relatively large leakage current and strong limitation on enhanced temperature (even under current flow). This limitation on enhanced temperature is connected with reducing GO: for instance, annealing at 100°C typically leads to a decrease in GO layer resistivity by 4-5 orders of magnitude [51,52].

The most stable graphene derivative with dielectric properties is fluorinated graphene (FG). Fluorinated graphene is low k material with k = 1.2 [53]. Recently, new simple approach for graphene fluorination (treatment in aqueous solution of hydrofluoric acid) was suggested [54,55]. In the case of graphene suspension such treatment leads not only to fluorination of the flakes but also to additional

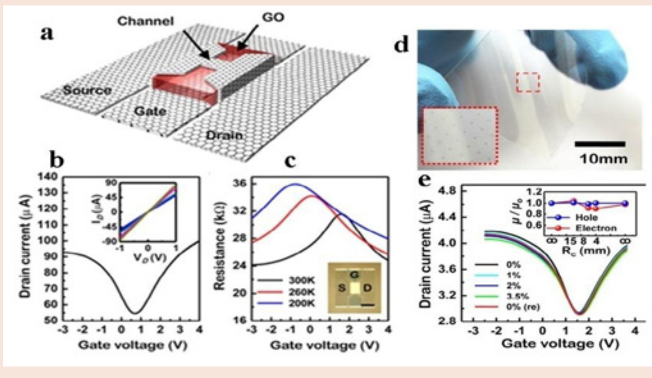

Figure 4: (a) Schematic illustration of bottom-gated graphene/GO transistor from Ref. [48 Lee 2012]. The graphene channel which was monolithically patterned with source and drain electrodes is above the GO dielectric. (b) Typical transfer characteristic of graphene/GO transistors, indicating the hole and electron mobilities are 300 and 250 cm^2/Vs at V_{DS} =−0.1 V, respectively. (c) Resistance versus applied gate voltage at different temperatures. Inset shows the microscope image of the real device that has a channel length of 10 μm and width of 25 μm, respectively (scale bar: 100 μm). (d) Optical image of the all-graphene-based transistor formed on plastic substrate. (e) Transfer curves measured as a function of tensile strains (from 0% to 3.5%). Inset shows effective mobility as a function of the bending radius. Reprinted with permission from Ref. 48. Copyright (2012) American Chemical Society.

flake fragmentation and exfoliation (Figure 5) [56,57]. As a result a considerable decrease in thickness and lateral sizes of the graphene flakes (up to 1-5 monolayer in thickness and 20-30 nm in diameter) is found to be accompanied by simultaneous transition of the flakes from conducting to insulating state. Smooth and uniform insulating films with roughness ~ 2 nm and thicknesses down to 10 -20 nm were deposited from the suspension on silicon. The electrical and structural properties of the films suggest their use as insulating elements in thin-film nano- and microelectronics device structures. In particular, it was found that the films prepared from the fluorinated suspension display rather high breakdown voltages (field strength of (1-3)x10^6 V/cm), ultra-low densities of charges in the film and at the interface with silicon substrate in metal-insulator-semiconductor structures (~ (1-5)x10^{10} cm^{-2}). Such excellent characteristics of the dielectric film can be compared only with well-developed SiO_2 layers. The films from the fluorinated suspension are cheap, practically feasible and easy to produce.

Combination of the oxidized and fluorinated graphene suspensions for creation of the insulated films gives the most outstanding results for the decrease of leakage current in the film [58]. Two-layer GO-FG films (thin film of fluorographene on graphene oxide) exhibit good insulating properties: the leakage currents in GO-FG film (~ 10^{-4} A/cm^2) was by 3-5 orders of magnitude lower than that in the graphene oxide or fluorographene films (Figure 6). Moreover, a significant increase in thermal stability of GO-FG films was revealed: annealing of the films up to temperatures 350°C leads to a decrease in film resistance of about one order of magnitude. These effects are connected with good affinity of the materials when application of thin fluorographene films (a few nanometers) from suspension with much smaller flakes (by an order of magnitude) decorates and eliminates structural defects in the graphene oxide films, and blocking conductivity in graphene oxide. Relatively low charges in the film and at the interface with silicon (3x10^{10} – 1.4x$10^{11}$$cm^{-2}$) were obtained in two-layer GO-FG films. The built-in charge density in the composite film of 10-20% fluorographene suspension in the graphene oxide suspension is much less than that in the two-layer film (< 1x$10^{10}$$cm^{-2}$). The effective permittivity of two-layer and composite films varies from 1.1 to 4.3 depending on composition, which is important for applications. The created two-layer and composite films may be practically applied in 2D printed and flexible electronics as insulating films (gate dielectrics, substrates for graphene, protected coatings, etc.).

Transparent dielectrics can be fabricated from graphene oxide or fluorinated graphene dispersions [51,59-61]. The film thickness which determines film transparency can be controlled by the concentration of the graphene oxide suspension and oxidation (fluorination) degree. For instance, the optical transmittance for GO film with 9 nm thickness was found to be equal to ~ 95% [60], and the optical transmittance values at a wavelength of 550 nm were 87% - 96% for the films with thickness ~16 and 3 nm made from 1.5 and 0.5 mg/ml suspensions, respectively [61].

FETs fabricated using a poly(methylmethacrylate) (PMMA) and lithium fluoride (LiF) composite dielectric is presented by Kumar et al., [62]. Increasing the concentration of LiF in the composite dielectric reduces the operating gate voltages significantly, from 10 V to 1 V, due to a decrease in resistance. Electron and hole mobility of 350 and 310 cm^2/Vs at V_D = -5 V are obtained for graphene FETs with 10 % LiF concentration in the composite. Using composite dielectric also enabled excellent performance on flexible substrates without any significant change in mobility and resistance. Flexible FETs with only 5 % and 12 % variation in mobility for 30^0 and 75^0 bending are obtained. LiF is a high k-dielectric and PMMA a low k dielectric with the values of dielectric constants 9.0 and 2.6, respectively.

Summary and outlook

Among traditional high-k materials (Al_2O_3, HfO_2, and ZrO_2) used for gate insulators in graphene FETs on flexible substrates fabricated by means of atomic layer deposition the maximal value of strain without strong degradation of the device characteristics are obtained for Al_2O_3 (strain 4.6%). Typical values of carrier mobilities observed in FET with different high–k dielectrics are ranged from 500 – 2000 cm^2/Vs. Moreover in the case when FET was fabricated without

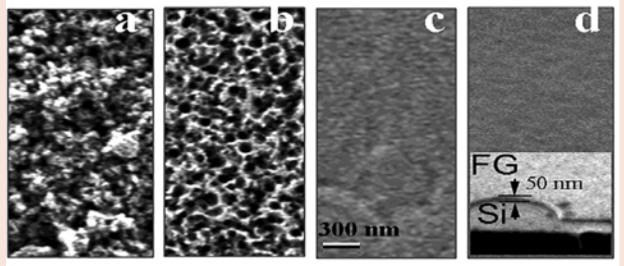

Figure 5: SEM images of the surface of films fluorinated in the aqueous solution of HF during different times. The scales in the images are identical. a – pristine (non-fluorinated) film; b, c, and d – films fluorinated respectively during 2, 10, and 40 days. The inset in Fig. 5 d shows an image of an edge of the film taken at the angle 45° to the surface; the film thickness indicated in the figure was evaluated with allowance for measurement geometry Reprinted with permission from Ref. 56.

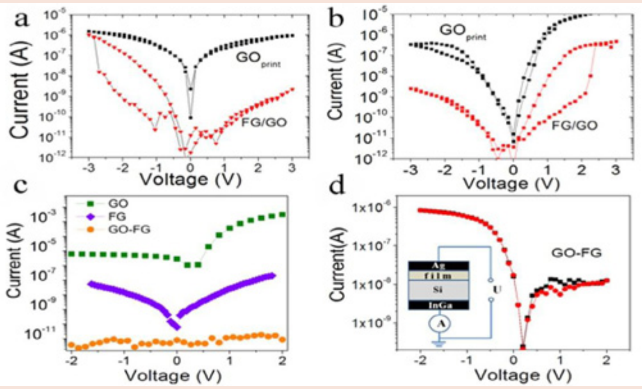

Figure 6: Current – voltage characteristics of two-layer and composite films: (a) for the lateral and (b) the vertical FG/GO structure with thickness of the printed graphene oxide layers of 35 nm, and thickness of fluorinated grapheme of 2-3 nm. (c) Comparison of the current values through the GO, FG and FG/GO films deposited by droplets. (d) Current – voltage characteristic for the vertical structure of the composite GO-FG film. Contact squares in all cases were 0.3-0.5 mm^2. [58].

damages to the flexible substrate due to using a rigid supporting substrate very high device performance including carrier mobilities of about 9000 - 13000 cm²/Vs were obtained. Nevertheless, Al_2O_3 demonstrates relatively high leakage current and charges in the layers and at the interface.

Fluorinated graphene or combination of FG with graphene oxide are very promising alternative variants. They have the same strain limitation as the graphene. Two-layer FG – GO films or composite FG: GO layers have low values of the leakage current and the charges in the layer and at the interface. Dielectric constant of any variants of FG – GO compositions are varied in the range of 4.3 – 1.2. Carrier mobilities in the FET with GO gate layers are found to be ~300700 cm²/Vs. Moreover, both FG and GO are high transparent layers. Generally, dielectric layers fabricated from fluorinated graphene or in combination with graphene oxide are the most promising graphene based derivative flexible and transparent electronics.

Mechanical properties of h-BN limit its applications for devices on flexible substrates. Mechanical properties of Y_2O_3, ion gels and other new gate dielectric materials for flexible electronics are not studied yet. Further development and study of these materials will clarify their perspectives for applications.

Acknowledgements

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