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学位論文題目	Development of CNT-based functional nanocomposites and their applications (カーボンナノチューブを用いた機能性ナノコンポジットの創製とその応用に関する研究)
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論文内容の要旨

The advancement of nanotechnology is progressing rapidly and the exploitation of them has been geared toward usage in various fields and applications such as nanocomposites, electronic devices, smart materials, and so on. Manipulation of size, morphology, structure, and purity of the nanomaterials has allowed the innovation of advanced materials and improvement to the traditional materials. Furthermore, nanomaterials also have generated a great attention from the researchers and commercial product developers as they can show novel and interesting properties than those of the corresponding bulk materials. In this dissertation, the focus has been forwarded to the fabrication of nanomaterials and the characterization of nanocomposites as electromagnetic (EM) wave absorbers and electroactive actuators. Carbon nanotubes (CNTs) have been utilized mainly in the research to fabricate CNT-based nanocomposites. Moreover, nanoparticles such as barium titanate (BTO) and silver (Ag) nanoparticles also have been synthesized and evaluated. The significant results obtained from this research are as follows:

(1) Barium titanate/carbon nanotube (BTO/CNT) hybrid nanocomposites were fabricated by sol-gel method. The BTO/CNT hybrid nanomaterials were characterized using X-ray diffraction (XRD), transmission electron microscopy (TEM), field emission scanning electron microscopy (FE-SEM), Raman and X-ray photoelectron spectroscopy (XPS). The BTO/CNT hybrid nanomaterials were then loaded in paraffin wax with different weight percentage, and pressed into toroidal shape with thickness of 1.0 mm to evaluate their complex permittivity and complex permeability using vector network analyzer. The reflection loss (*R.L.*) of the samples was calculated according to their measured complex permittivity and permeability. Samples with various weight fractions were prepared and evaluated. Improved *R.L.* can be obtained with higher weight fraction and this greatly affected the absorption ability. The BTO/CNT 60 wt.% hybrid nanocomposites with thickness of 1.1 mm showed a minimum *R.L.* of ~-56.5 dB (over 99.999% absorption) at 13.2 GHz and was the best absorber when compared with the other samples of different thickness. The reflection loss peak shifted to lower frequency and wider response bandwidth can be obtained as the thickness of the samples increased. The EM wave absorption properties of double-layer BTO/CNT nanocomposites where BTO/CNT nanopowders were incorporated into polyurethane (PU) matrix were also evaluated. The minimum *R.L.* of single-layer BTO/CNT 30 wt.% nanocomposites sample with a thickness of 1.1 mm reached ~-30.3 dB (over 99.9% absorption) at 13.8 GHz, and the bandwidth of the *R.L.* less than -10 dB (over 90% absorption) was 1.5 GHz. The double-layer composites consist of BTO/CNT 30 wt.% (Layer 1) with thickness of 1.0 mm and BTO 30 wt.% (Layer 2) with thickness of 0.3 mm showed a minimum

R.L. of ~ 63.7 dB (over 99.9999% absorption) at 13.7 GHz, and the bandwidth of the *R.L.* less than -10 dB was 1.7 GHz. The *R.L.* improved and wider response bandwidth can be obtained with the double-layer composites. This proves that the thickness of samples can be manipulated to produce absorption bands at different frequencies to design highly effective EM wave absorbers. Furthermore, the high interfacial area of the nanomaterials results in high interfacial polarization and multiple scattering, which improves EM wave absorption. Influence of weight fraction, behavior of permittivity and permeability, nanoeffects, and so on are discussed to understand the EM wave absorption mechanism.

(2) Ag nanoparticles fabricated by chemical reduction process were grafted onto the surface of CNTs to prepare hybrid nanocomposites. The Ag/CNT hybrid nanomaterials were characterized using TEM, XPS, and Raman spectroscopy. The Ag/CNT hybrid nanomaterials were then loaded in paraffin wax, and pressed into toroidal shape with thickness of 1.0 mm to evaluate their complex permittivity and complex permeability. The *R.L.* of the samples was calculated using their measured complex permittivity and permeability. The minimum *R.L.* of the Ag/CNT hybrid nanocomposite sample with a thickness of 1.0 mm reached ~ 21.9 dB (over 99 % absorption) at 12.9 GHz, and also exhibited a wide response bandwidth where the frequency bandwidth of the reflection loss of less than -10 dB (over 90 % absorption) was from 11.7 to 14.0 GHz. The Ag/CNT hybrid nanocomposite with thickness of 6.0 mm showed a minimum reflection loss of ~ 32.1 dB (over 99.9 % absorption) at 3.0 GHz and was the best absorber when compared with the other samples of different thickness. The reflection loss shifted to lower frequency as the thickness of the samples increased. Interestingly, the Ag/CNT hybrid nanomaterials also exhibited two *R.L.* peaks, at low and high frequency region, as the thickness of the sample increased, and they also showed wide minimum *R.L.* of over -10 dB. The capability to modulate the absorption band of these samples to suit various applications in different frequency bands simply by manipulating their thickness indicates that these hybrid nanocomposites could be a promising EM wave absorber. Optimization of the EM wave absorber that is able to perform at certain frequency and broad frequency range can be realized by controlling the absorber's design.

(3) The electroactive nanocomposite films were fabricated using PU with modified CNTs as the filler. The CNTs were modified using microwave-induced polymerization and mild hydrothermal route and they were found to be highly dispersed in polar solvents such as dimethylformamide. The modified CNTs contained more carboxyl and hydroxyl groups and showed less aggregation than that of the pristine CNTs. The modified CNTs were characterized using TEM, FE-SEM, thermogravimetric analysis (TGA), and XPS. To evaluate these films we mainly focused on electrical properties such as actuation behavior, volume resistivity, dielectric spectroscopy, impedance analysis, and space charge measurements. We found that the PU/CNT films bent toward the cathode when an electric field was applied and they reverted to its original position when the electric field was removed. Upon the incorporation of CNTs as the filler for the polymer, the electrical properties of the films improved significantly. The highly polarized films had a high relative permittivity, and this produced a higher Maxwell pressure, which assisted the actuation. Asymmetric charge accumulation was observed from space charge measurements in some of the films and this explains the bending deformation and the actuation behavior. This helped us to understand the actuation mechanism and bending deformation direction of this kind of electroactive nanocomposite materials. Furthermore, by considering the factors that affect the actuation, highly effective actuator material that shows high performance and can be applied for advanced materials in the future could be designed.