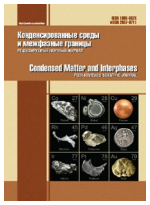

ISSN 1606-867X (Print)
ISSN 2687-0711 (Online)
Condensed Matter and Interphases
Kondensirovannye Sredy i Mezhfaznye Granitsy
https://journals.vsu.ru/kcmf/

Original articles

Research article
https://doi.org/10.17308/kcmf.2022.24/9264

Preparation and characterization of Ge-Ni-Te nanocomposite

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Abstract

$Ni_xGe_{50-x}Te_{50}$ with x = 2, 4, 6, 8, 10, 15 and 20 at% ternary nanocomposite prepared using multistage solid-state direct reaction. Nanocrystalline nature was studied by X-ray powder diffraction, results reviled that, the main phase is rhombohedral GeTe polymorph, and the second major phase is hexagonal Ni_3GeTe_2. The calculated average crystallite size of the whole constituents in prepared samples is within the range of 47.3-83.8 nm. Optical properties evaluated from diffuse reflection measurements and the calculated bandgap of all samples are nonmonotonically changes with Ni content from 1.45 to 1.62 eV with the direct allowed transition.

Keywords: Ni-Ge-Te, Nanocomposite, Structural and optical properties, Diamagnetic

Funding: This work was supported by the American University in Cairo (AUC) project № SSE-PHYS-S.E.- F.Y17- F.Y18-F.Y19-RG(2)-2016-Feb-10-08+45-59 & project № SSE-PHYS-H.O- F.Y20- RG(2-19)-2018-Dec-08-03-06-32-.FZGU-2020-0036. As well as technical support from Nanomaterials Laboratory at Physics Department, Faculty of Science, Al-Azhar University (Girls Branch) and National Research Centre (Cairo, Egypt).

For citation: *Mahdy I. A., El Sheikh S. M., Omar H. A., Seredin P. V., Mahdy M. A.* Preparation and characterization of Ge-Ni-Te nanocomposite. *Condensed Matter and Interphases*. 2022;24(2): 243–249. https://doi.org/10.17308/kcmf.2022.24/9264

Для цитирования: Махди И. А., Эль Шейх С. М., Омар Х. А., Середин П. В., Махди М. А. Получение и исследования нанокомпозитов Ge-Ni-Te. *Конденсированные среды и межфазные границы*. 2022;24(2): 243–249. https://doi.org/10.17308/kcmf.2022.24/9264

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1. Introduction

Diluted magnetic semiconductors (DMS) are the focus of great interest due to their exceptional potential for implementation in the spintronic industry. Their importance lies in the fact that it provides scientific information and potential technological applications of semiconductors [1–4] for tunable ferromagnetic devices [5]. Among the IV–VI semi-magnetic semiconductors, only a few compounds (PbSnMnTe, SnMnTe, GeCrTe, and GeMnTe) show the ferromagnetic (FM) ordering [6]. The interaction between magnetic Mn ions doped GeTe films is ferromagnetic although Ni-doped GeTe films are paramagnetic [7] and $Ni_{5.42}GeTe_2$, crystallized in the tetragonal system [8]. Both crystallization speed and thermal stability of GeTe are significantly increased by Ni doping. The carrier concentration in multiferroic diluted magnetic semiconductors can control its magnetic properties; which may provide some new opportunities for novel spintronic and magnetoelectronic materials and devices. The magnetic, magneto-optical, and transport properties of the DMS materials, which are recently treated as spintronic model materials, are controlled via three coupled subsystems: free carriers, lattice excitations (phonons), and magnetic ions [9]. The spin and energy transfer between these coupled systems controls spin dynamics in DMS, where the magnetic ions and free carriers (electrons and holes) have non-vanishing spins [10]. Cao L. et al., [11, 12] studied the effect of Ni-doped GeTe thin films and demonstrated the possibility of using it in high-temperature phase-change memory (PCM) applications. This is due to the distinctive characteristics of its ability to low power consumption, retain data for a long time (10- years), and consider an excellent electric switching speed of 6 ns. Due to its high crystallization temperature and good data retention ability, a short electrical pulse of up to 6 ns can achieve a reverse switch between the SET and RESET states.

The major goal of this research is to study the effect of replacing Ge with Ni in a GeTe system with various Ni concentrations (2-20%) on the structural and optical properties of GeTe, as well as their magnetic response as the Ni ratio changes.

2. Methods of production and studying of the samples

In the present work the $Ni_xGe_{50-x}Te_{50}$ with $x = 2$, 4, 6, 8, 10, 15 and 20 at% ternary compound has been prepared in an evacuated quartz ampoule with 14 mm diameter by the conventional solid-state direct reaction by multistage preparation from high purity crystalline Ge 99.999% Ge, Ni 99.999%, and Te 99.999% grade Sigma Aldrich. Starting Ge, Te and Ni were mixed in sealed quartzes ampoule evacuated under argon atmosphere for preventing oxidation, and then prepared with their direct reaction at 1000±1 °C for 24 hr and then cooled at a cooling rate 2 °C/min to obtain the initial ingot. In the second stage, the obtained ingots were ground in an agate mortar and then loaded in a pressing die with 13 mm diameter at a pressure of 5 tons for 5 min, pellet samples were sealed and evacuated again. Samples were heated in a vertical furnace to reduce the temperature gradient and then thermally treated at 700 ± 1 °C for 10 days without breaking, and then cooled with a cooling rate of 2 °C/min to reach room temperature. finally, the second stage was repeated once at 1000±1 °C for 150 hr. The crystalline nature of all compounds is investigated using [Bruker X-Ray Diffractometer D8] with (Cu$K\alpha$) radiation over [4: 90°] angular range with a normal scan, step size 0.03 and wavelength ~ 1.54061 Å. Diffuse reflection of all compounds was measured using a (Jasco V-570) spectrophotometer. The magnetic properties of all samples were investigated using a vibrating sample magnetometer (Lakeshore 7410).

3. Results and their discussion

3.1. Structural properties of Ni-Ge -Te

Structural properties of the prepared samples are investigated using XRD, Results of the X-ray diffraction pattern depicted in Fig. 1. phase analysis shows that the $N_xGe_{1-x}Te$ has a multi-phase nanocrystalline structure. The main phase of all compositions is rhombohedral GeTe modification identified by the ICDD card (No 47-1079) GeTe. The second phase which appeared in the composite is the ternary phase that was identified as a Ni_3GeTe_2 hexagonal crystal with lattice parameters: a = 3.911 Å, c = 16.022 Å – which was identified by ICDD card

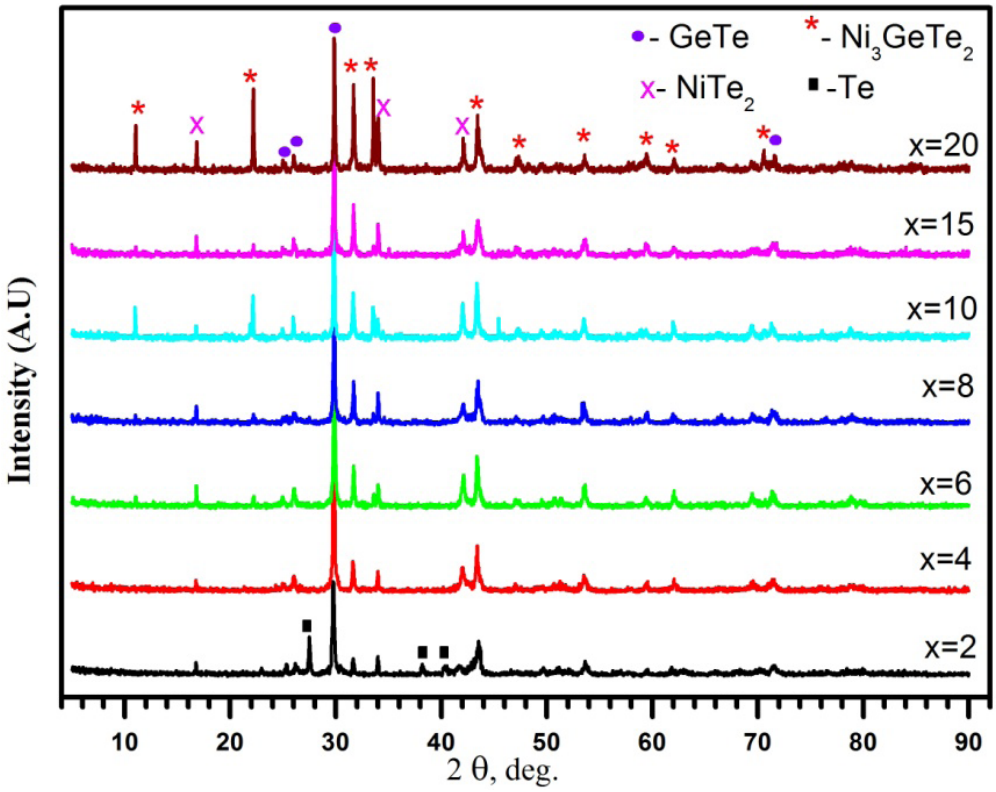

Fig. 1. X-ray difrractogram of $Ni_xGe_{50-x}Te_{50}$ nanocomposite with different concentrations

No (01-075-5621). The additional secondary phases are identified as a $NiTe_2$ hexagonal system of lattice parameters: a = 3.854 Å, c = 5.2604 Å – which was identified by ICDD card No (88-2278). But at low values of Ni content x = 2 at%, there is some precipitation of Te which indicated by the diffraction lines at 2θ = 22.97, 27.50, and 40.34 diffraction lines, all Te peaks are disappeared at different concentrations from x = 4 to x = 20 at% as depicted in Fig. 1. The highly intensive diffraction peak of the main GeTe phase was used to determine crystallite size D using Scherrer's equation [13, 14]:

$$D = \frac{k\lambda}{\beta \cos(\theta)}, \quad (1)$$

where the shape factor $k \approx 0.9$, and β: full width at half maximum (FWHM) of the diffraction peak (radians), λ = 1.5405 Å represents the wavelength of (Cu$K\alpha$) radiation, θ is the Bragg angle of the XRD peaks. Moreover, the average crystallite size overall the different phases that appeared in the composite and strain ε established in the preparation process were evaluated using the William-son–Hall, W–H, plot as shown in Fig. 2 using the following equation [15, 16]:

$$\beta\cos\theta = \frac{k\lambda}{D} + 4\varepsilon\sin\theta. \quad (2)$$

The calculated crystallite size from both Scherrer's and W–H, plot confirms the nanocrystalline nature of the prepared composite. The obtained result of crystallite size and strain ε formed in nanocomposite with different compositions during the preparation process indexed in Tabl. 1.

3.2 Optical band gap calculations using Diffuse reflection

Diffuse reflection is one of the employed methods for optical characterization. The electronic transitions of solid materials can be described by using the technique of optical diffuse reflection [17]. Diffuse reflection spectra of Ge-Ni-Te samples were measured in the spectral range from 190 to 2500 nm and represented in Fig. 3. The absorption spectra can be obtained by using the following relation of Kubelka–Munk [17, 18]:

$$F(R) = (1-R)^2 / 2R, \quad (3)$$

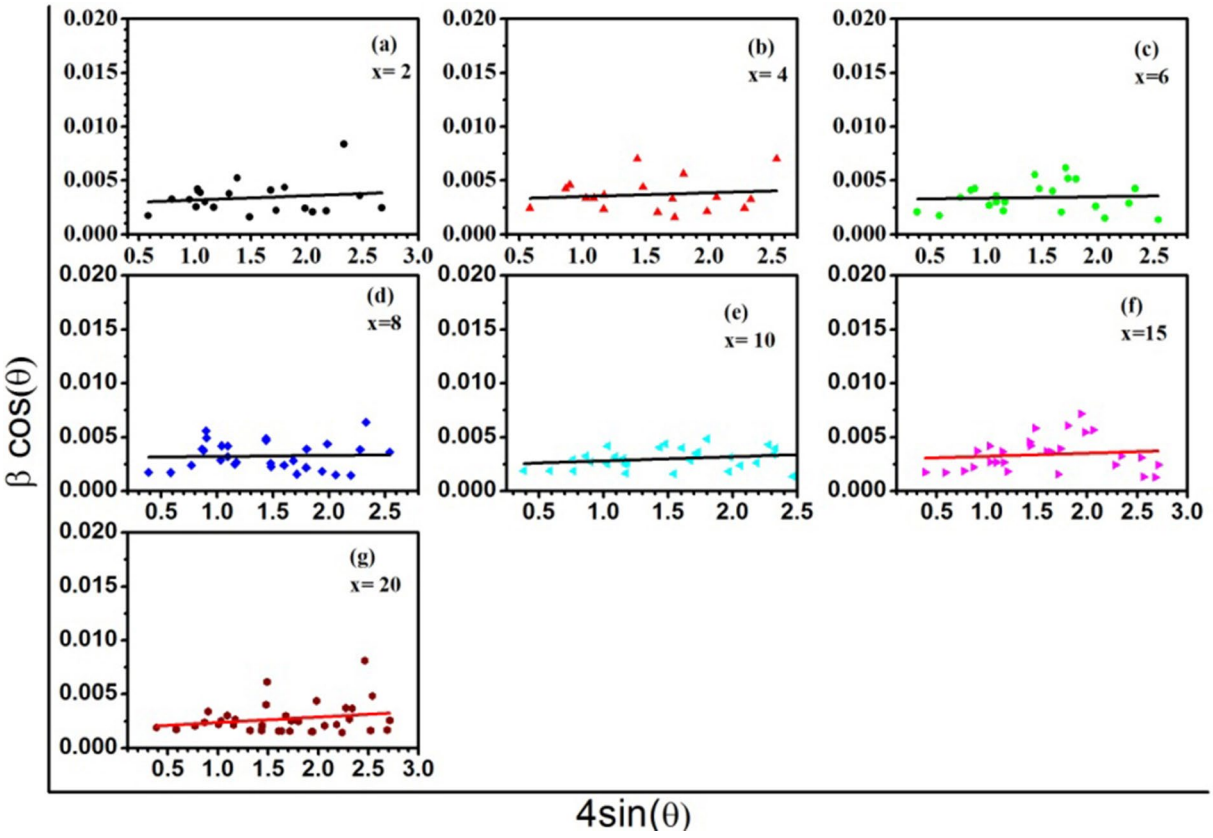

Fig. 2. Williamson–Hall plot of $Ni_xGe_{50-x}Te_{50}$ nanocomposite with different concentrations

where $F(R)$ is the Kubelka–Munk function and R represents the measured diffuse reflection. Fig. 3 represents the plot of $(F(R)*E)^2$ as a function of photon energy E [19]. The optical band gap for each composite was determined from the intercept of the linear part with the energy axis as seen in Fig. 1, the energy gap evaluated is listed in Tabl. 1. The optical band gap of all examined samples is greater than those of GeTe in microcrystal ~0.73–0.95 eV [20]. The redshift that appeared in the bandgap of nanocomposite can be attributed to the particle size of the prepared nanocomposite.

3.3. Magnetic properties of Ni-Ge-Te.

Magnetic properties of $Ni_xGe_{50-x}Te_{50}$ nanocomposite system were measured at room temperature. The resultant data of magnetic moment as a function of the applied magnetic field is depicted in Fig. 4 for different compositions. All results show that all samples have the predominant diamagnetic effect despite the presence of the paramagnetic phase Ni_3GeTe_2 [21, 22] as a secondary phase as mentioned above. That diamagnetic behavior can be attributed to the main GeTe phase which is characterized as a diamagnetic material [23].

Table 1. The evaluated crystallite size and optical gap of nanocomposite samples with different compositions

Ni at%	D nm, Scherrer	D nm, W-H plot	Strain ε % ×10^{-4}	E_g (eV)
2	55.4	32.7	3.9	1.54
4	55.4	49.1	3.49	1.51
6	47.8	51.38	1.3	1.62
8	49.4	48.36	0.9	1.57
10	64.2	54.8	3.9	1.45
15	52.4	51.3	2.8	1.54
20	83.3	54.8	5.1	1.45

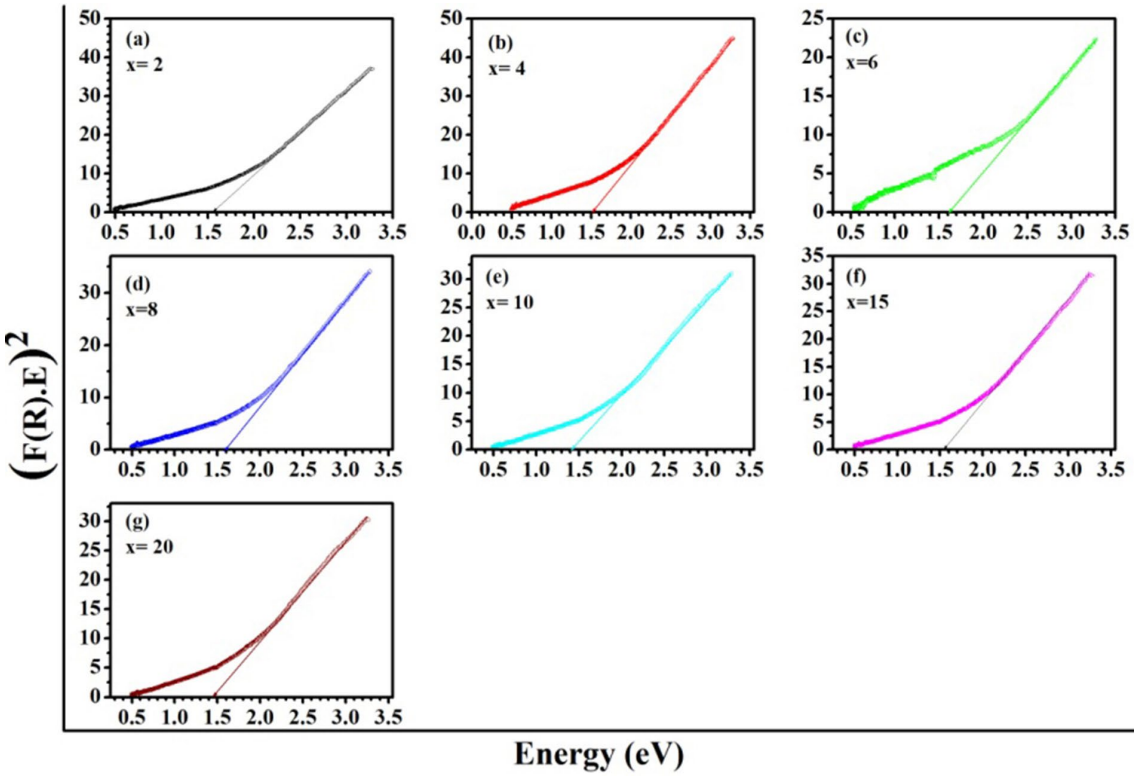

Fig. 3. Representation of $(F(R)^*E)^2$ as a function of photon energy E in eV of $Ni_xGe_{50-x}Te_{50}$ nanocomposite with different concentrations

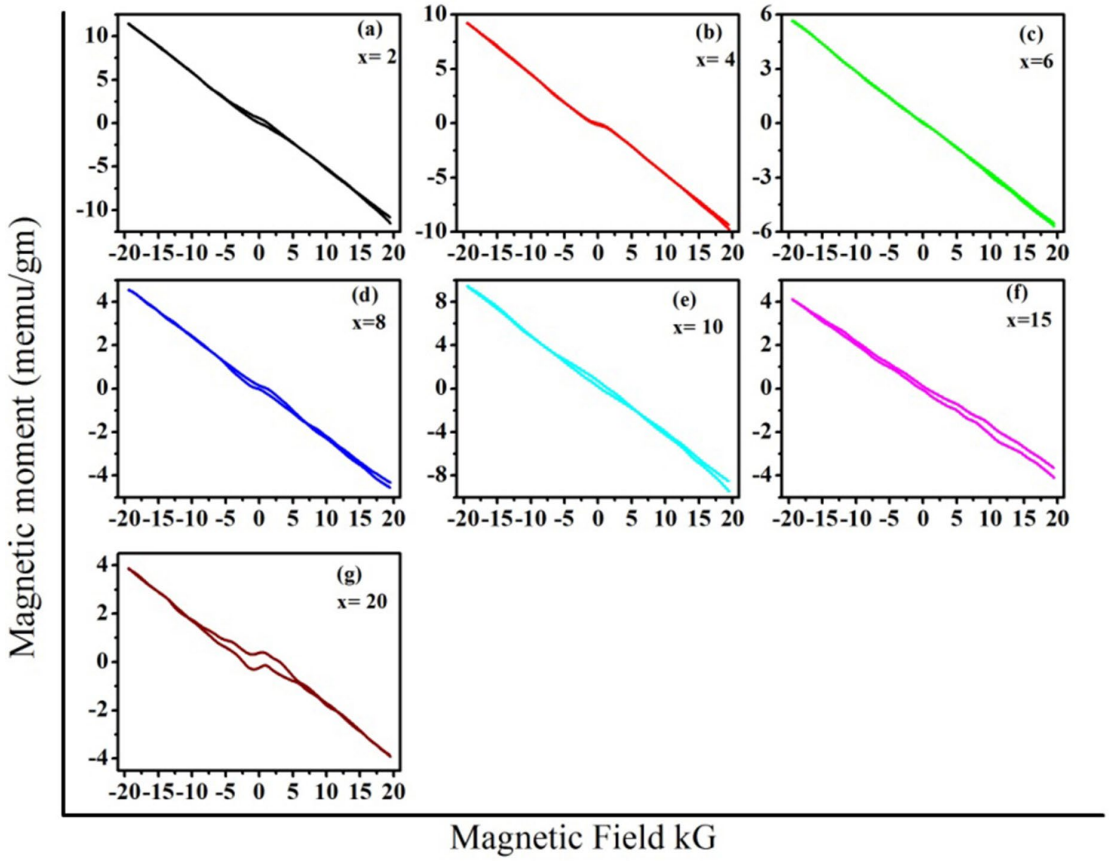

Fig. 4. Results of the magnetic moment as a function of the applied magnetic field of $Ni_xGe_{50-x}Te_{50}$ nanocomposite with different concentrations

4. Conclusions

The prepared $Ni_xGe_{50-x}Te_{50}$ alloy, x = 2, 4, 6, 8, 10, 15, 20 at% identified as a nanocomposite form with a multiphase structure of average crystallite size 47.8–83.8 nm. Optical properties of such nanocomposites show a direct allowed electronic transition with optical gap changes in the range of 1.45 to 1.62 eV with irregular change which can be attributed to the multiphase structure. Magnetic measurements confirm the diamagnetic behavior of all nanocomposite samples, which can be attributed to the predominant GeTe rhombohedral crystalline modifications.

Author Contributions

Iman A. Mahdy – conceptualization, scientific management, research concept, methodology development, formal analysis, writing – review & editing, visualization, final conclusions. S. M. El Sheikh – resources, supervision, project administration. Hosny A. Omar – resources, supervision, project administration. Pavel V. Seredin – writing the Russian version. Manal A. Mahdy – investigation, formal analysis, methodology, writing – review & editing.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Received 22.02.2022; approved after reviewing 04.03.2022; accepted for publication 15.05.2022; published online 25.06.2022