

# Study of Thermoelectric Properties of Nickel Germano-silicides Obtained on the Basis of Bulk Solid Solutions of Silicon-germanium

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## To cite this article:

Nuriddin Azadovich Matchanov, Komiljon Abdusharipovich Bobojonov, Dilmurad Shamuradovich Saidov, Shokhzod Komiljonovich Abdusharipov, Alisher Erkaboevich Rajabov. Study of Thermoelectric Properties of Nickel Germano-silicides Obtained on the Basis of Bulk Solid Solutions of Silicon-germanium. *American Journal of Physics and Applications*. Vol. 10, No. 2, 2022, pp. 45-50.  
doi: 10.11648/j.ajpa.20221002.14

**Received:** January 15, 2022; **Accepted:** February 4, 2022; **Published:** April 9, 2022

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**Abstract:** The effect of thermal annealing in vacuum (in the temperature range of 200-900°C) on the electrical conductivity of the structure of a nickel film - a single-crystal layer of germanosilicide has been studied. Nickel films 15 nm thick were sputtered by adsorption sputtering in vacuum. The possibility of forming conductive layers with a surface resistance of about 4.9 mOhm \* cm and on their basis bulk Si<sub>1-x</sub>Ge<sub>x</sub> single crystals with a thickness of 4 μm by mechanical processing and chemical etching, as well as deposition in a vacuum environment of 10<sup>-5</sup> -10<sup>-7</sup> Torr. The surface resistance of the film and the surface morphology (and chipping) were studied, the phase state of nickel germanium silicide films was studied by X-ray diffraction, the study of thermoelectric properties and the comparison of Ni-SiRH and Ni-Si<sub>1-x</sub>-Ge<sub>x</sub> samples as thermoelectric materials. The thermopower values were obtained experimentally in the temperature range 36-107°C, and the role of the nanoscale film and substrate in the formation of the thermoelectric effect was discussed. Increasing the thermoelectric figure of merit due to nickel with decreasing surface resistance and due to phonon scattering on surfaces and heterostructures such as nickel germanosilicide, superlattices and layer/substrate systems. The best quality factor ZT = 1.98 ± 0.05 was obtained for Ni-(Si<sub>0.95</sub>Ge<sub>0.05</sub>) at T = 300K. The maximum difference with a value of ΔT<sub>max</sub> = 326.84K.

**Keywords:** Thermoelectricity, Solid Solution, Alloying, Clusters, Solid-State Reactions, Nickel Silicide, Germane-Nickel Silicide, Nanoclusters, Agglomeration

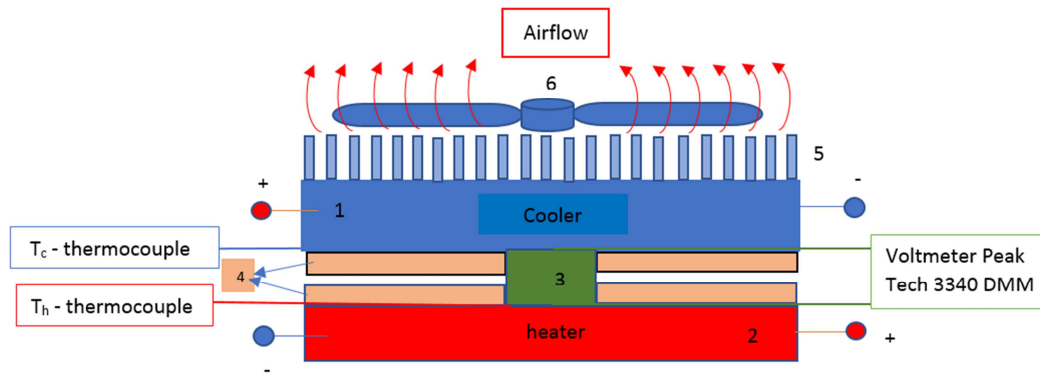
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## 1. The Purpose of the Experiment

The main goal of the experiment is to search for new alternative portable energy sources. The most urgent task these days is recharging mobile devices like cell phones, smartphones, tablets, etc., for this we need thermoelectric converters at low temperature drops. In this study, we are interested in the problem of autonomous power supply of low-power electronic devices (wireless sensors, mobile

communications) due to the accumulated energy collected in the presence of natural minimum temperature differences of the order of 27-100°C.

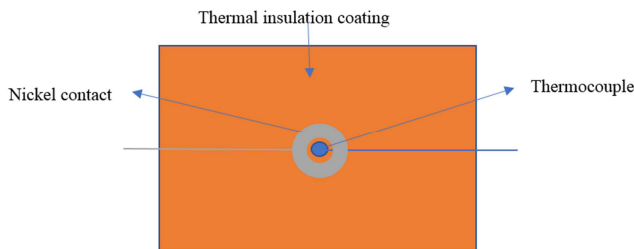
Thermoelectric conversion allows the use of almost any source of heat flux, in which the use of other conversion methods is impossible. There is always a natural temperature difference: the temperature of a person's body is the temperature of air, water, earth, air temperature is the temperature of flowing water, etc. [25].



**Figure 1.** Device for measuring thermopower: 2- Cooler and heater (Peltier element) ( $T_c$ ) and ( $T_h$ ) (TES1-127110), 3. Ni-Si<sub>1-x</sub>Ge<sub>x</sub> (Sample), 4. Thermal insulation coating, 5. Cooler registers, 6. Cooler.

## 2. Measurement Technique

In this work, thermoelectric converters were investigated by the Harman method [1] based on a semiconductor solid solution Ni-(Si<sub>1-x</sub>Ge<sub>x</sub>). In the last 10 years, nickel with semiconductors has been a promising thermoelectric [23, 24] for the medium (200–900°C) temperature range and has a number of competitive advantages: low cost, high availability, well-developed production methods, and low toxicity. The latter, in comparison with analogs, is one of the main advantages of the Ni-Si-Ge system [2, 27]. In addition, there is a high level of understanding of the electronic properties in the bulk of Si<sub>1-x</sub>Ge<sub>x</sub> [29]. Development and research of thermoelectric materials based on Si<sub>1-x</sub>Ge<sub>x</sub> is the subject of research by a number of research teams in Uzbekistan, Russia and abroad [6-10].



**Figure 2.** Thermopower measuring device: arrangement of contacts and thermocouples.

In order to obtain good thermoelectric power characteristics, we needed a modern device for measuring thermoelectric power [1, 11] as in Figure 1, we used Seebeck and Peltier elements as a cooler and a heater. As a thermal insulator, we

applied on the surface of the Seebeck and Peltier element (TES1-12710) a thermal coating with a thickness of 2.5 mm with heat resistance RTV-silicone-2 auto-sealant AUTOSIL - 11225, as a good contact we took copper with coated nickel (since we have the material Ni-(Si<sub>1-x</sub>Ge<sub>x</sub>) in the form of a washer with a circle diameter of 8 mm and a hole diameter of 1.5 mm (referring to 7 mm samples) as shown in Figure 2. A thermocouple was placed on the hole in the center of the contact washer to measure the temperature.  $T_h$  and  $T_c$  as well as thermoelectromotive force (thermoEMF) were measured with a digital universal device PeakTech 3340 DMM [28].

## 3. Results and Discussion

Table 1 ZT shows the experimental temperature dependences of the Z solid solution Ni- (Si<sub>0.95</sub>Ge<sub>0.05</sub>) at various concentrations of current carriers. It can be seen from the results that the maximum Z grows rapidly with increasing concentration of current carriers up to  $2.13 \cdot 10^{19} \text{ cm}^{-3}$ . With a further decrease in the temperature difference between the hot and cold ends, the concentration decreases Figure 3. All the features on the temperature dependences of Z can be associated with the complex structure of the conduction band and can easily be qualitatively explained within the framework of the model of the conduction band proposed in [12]. Thermoelectric efficiency is high and the figure of merit reaches  $ZT = 1.98$  and the efficiency is 3.6%, and this is due to a decrease in the surface resistance of Figure 4 due to nickel and the type of conductivity table 1, and the appearance of films of nickel germanosilicides [3].

**Table 1.** Comparison of thermoelectric properties of Ni-SiHR and Ni- (Si<sub>1-x</sub>Ge<sub>x</sub>).

Sample number	Temperature annealing, °C	Temperature difference $\Delta T$ , K	Potential difference, mV	Seebeck coefficient $\alpha$ , mV/K
136-13	500	37	40.9	1.105
139-10	400	36	27.4	0.76
139-7	300	37	70.6	1.96
132-00	200	36	32.1	0.89
139-15	600	37	16.7	0.45
139-17	700	35	13	0.37
139-24	800	36	3.7	0.1
139-30	900	37	7.6	0.2
Si-HR	300	37	9.6	0.25

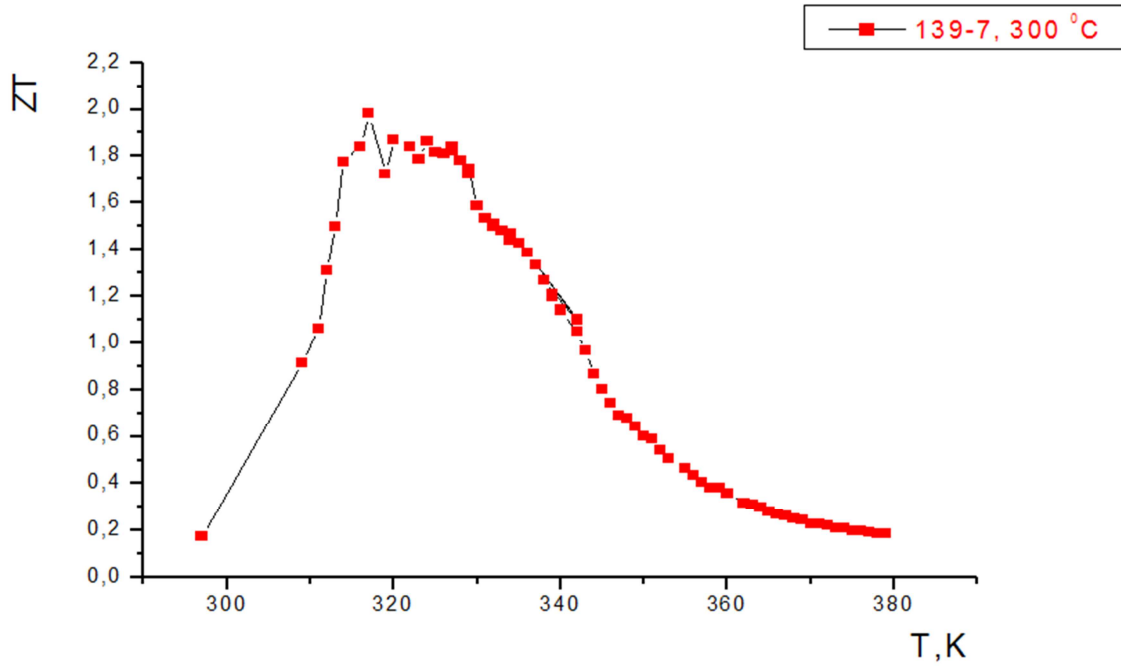


Figure 3. Thermoelectric figure of merit Ni- ( $\text{Si}_{1-x}\text{Ge}_x$ ) (n-type) at an ambient temperature of 300°C.

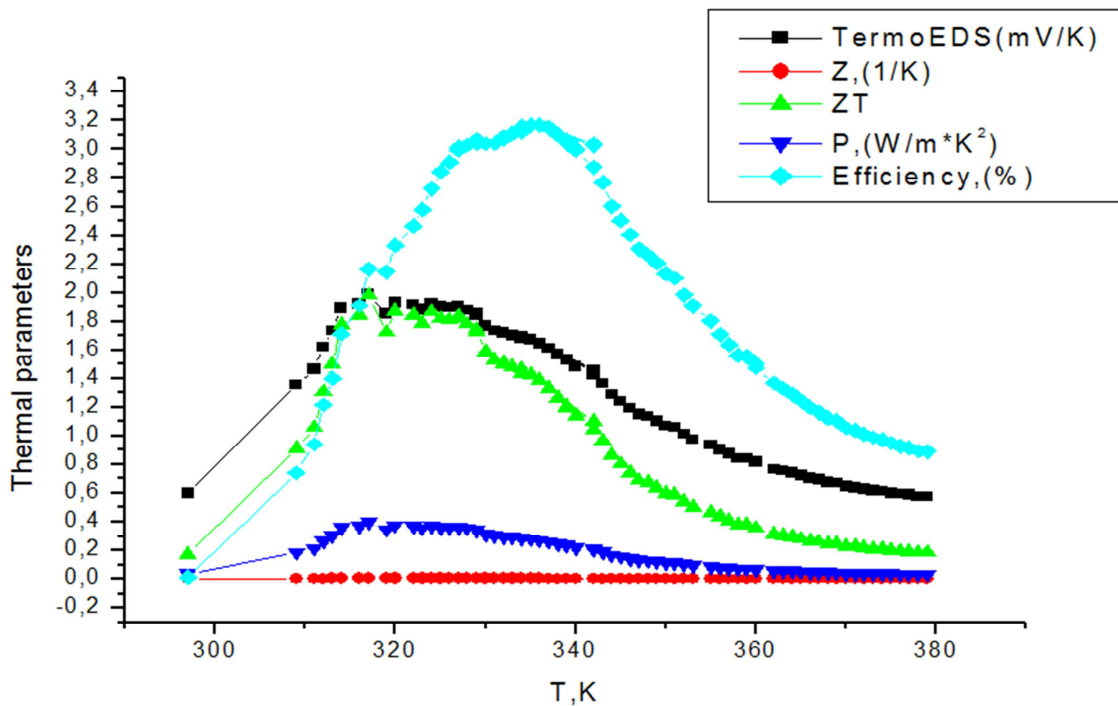
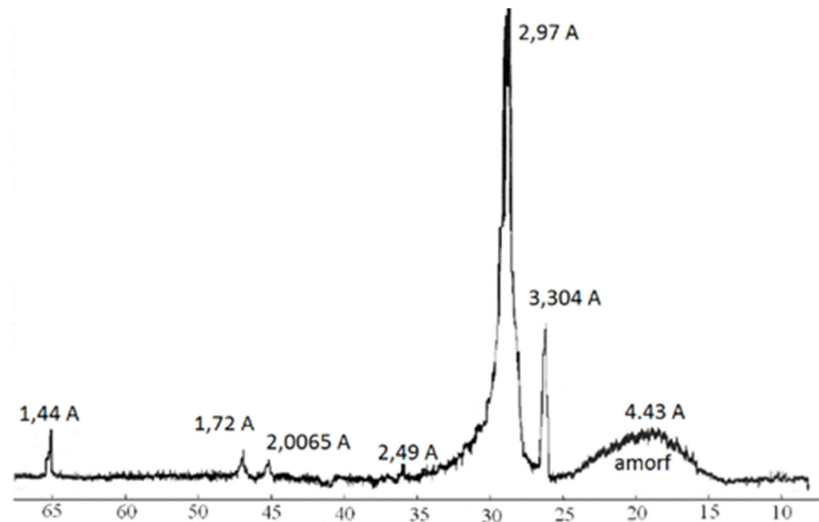


Figure 4. Thermal characteristics of Ni- ( $\text{Si}_{1-x}\text{Ge}_x$ ) (n-type) at an ambient temperature of 300°C.

In the solid-phase reaction, the most surprising thing is the appearance of nickel germane-silicide and the reaction continues until the formation of an amorphous layer [13-15, 26]. The optimum temperature for the Ni- $(\text{Si}_{1-x}\text{Ge}_x)$  film is 300°C, table 1 and Figure 6. In the bulk of a  $\text{Si}_{1-x}\text{Ge}_x$  single crystal with nickel atoms, hetero-structures will appear at 200°C with a mutual solid-phase reaction, and with an increase in the annealing temperature, the number of Nano-layers will grow to the appearance of nickel germanosilicide [16] up to 700°C [3-5], and after that an amorphous layer begins to

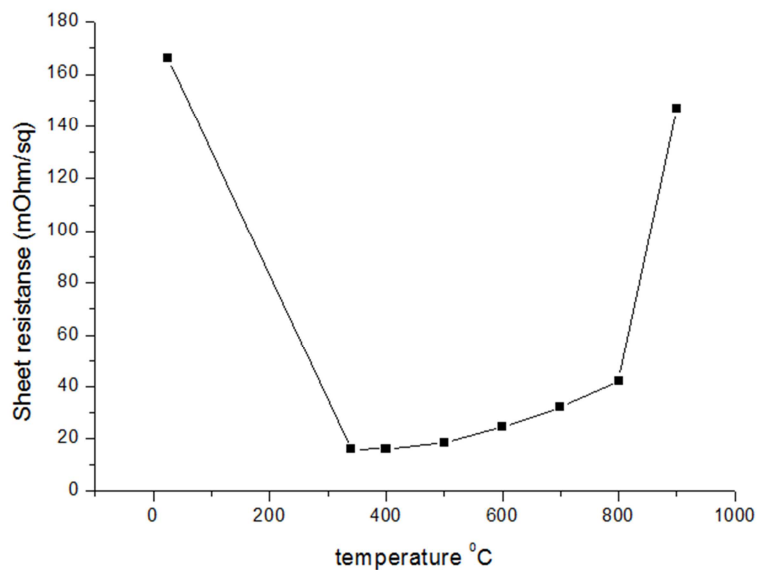
Figure 5 at the same time on the surface of the appearance of a nucleating island and a nanoisland of germano-silicides of nickel, this process occurs by agglomeration of the surface and [17, 16] Figure 7 begins the destruction of the structure and nickel atoms diffuse deeper [22]. These completely new nanomaterials for future super technology depend on surface and bulk nanofilms [18]. And we can add that nickel silicide (Ni-SiHR) was obtained due to the high surface resistance, the figure of merit  $ZT = 0.00042$ . This completely proves the main reason for this absence of a nickel-hermonide film [3, 30, 31].



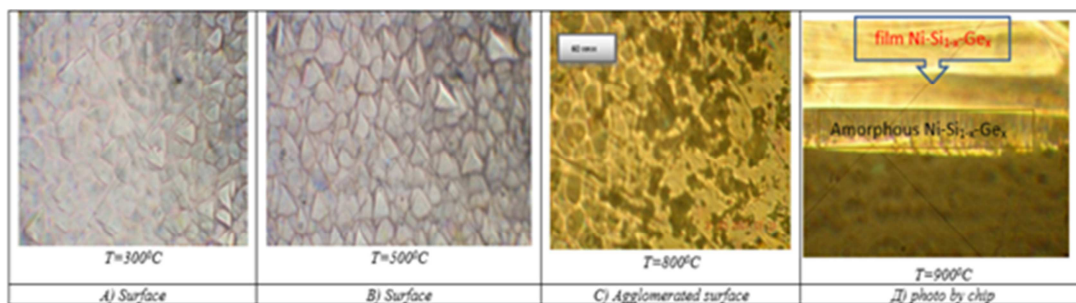
**Figure 5.** X-ray reflection spectrum from a Ni- ( $\text{Si}_{0.93}\text{Ge}_{0.07}$ ) film formed on a  $\text{Si}_{0.93}\text{Ge}_{0.07}$  substrate at an annealing temperature of 900°C. Reflection peaks corresponding to nickel germane-silicide 65-35.

Since we know that germanium is an expensive material than silicon, we decided to get a material with a small germanium composition of up to 0.5%, it is even more important that Ni reacts more easily with Si than with Ge. On the other hand, if the composition of germanium is less then  $\Delta T_{\text{max}}$  and  $\eta$  will mix in the region of low

temperatures [19, 20] and this is confirmed by tabular data, table 1. The maximum concentration of nickel in the near-surface enriched layer weakly depends on the diffusion temperature, and during subsequent heat treatments at temperatures below 900°C the enriched layer is retained [21, 32].



**Figure 6.** Surface resistance of annealed at temperatures of 300-900°C.



**Figure 7.** Surface images from the MIM-8 microscope.

## 4. Conclusion

The preparation of a thermoelectric element based on the contact "nickel-silicon" and "nickel-cremium-germanium" is investigated. Studied solid-phase reactions in the range of temperatures 200-900°C. The Zeebeck coefficients were determined and the thermoelectric parameters were calculated as the figure of merit, the maximum temperature difference, power and efficiency, in the range 36-107°C. The threshold values of thermal parameters for the lowest temperature of the human body (36.6°C) were obtained. The surface morphologies were studied and the analysis of the structures according to the DRON was done. The surface morphology was also studied for the cleavage.

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## Biography



**Komiljon Abdusharipovich Bobojonov**, physics teacher and independent researcher at the Urgench branch of the University. Information Technology named after Muhammad al-Kharezmi. *His scientific research is the study of thin films such as nickel germanosilicide's.*