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RESEARCH ARTICLE

MONOLYTIC MODULE BASED ON $Si_{0.7}Ge_{0.3}$ ALLOY FOR THERMOELECTRIC GENERATOR

Guram Bokuchava[1], Karlo Barbakadze[1] and Irakli Nakhutsrishvili[2]*

[1]Sukhumi Institute of Physics and Technology
[2]Institute of Cybernetics of Georgian Technical University

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ABSTRACT

Made n- and p-type $Si_{0.7}Ge_{0.3}$ thermoelectric alloys and their thermoelectric characteristics are studied. These alloys are cut into profiled plates, monolithic packages are assembled from them, switching plates are glued to the lower and upper bases of the packages, commutated packages are cut into 4-5 mm thick plates and monolithic thermoelectric modules are assembled from them. Electrical insulation nodes of thermoelectric modules are made on the basis of AlN and graphite plates. These nodes are connected to modules, resulting in monolithic samples. The energy characteristics of monolithic thermoelectric modules are studied.4 n- and p-type alloy plates (2 n-type and 2 p-type) were taken to make a mini monolithic thermoelectric module containing 16 branches. They were arranged in n-p-n-p order. A mini-monolithic thermoelectric module containing 24 branches was made using a similar technology. The difference is that the monolithic package is made of 6 plates.The electric power of thermoelectric modules is calculated by the electromotive force, external electric resistance and electric voltage generated by the module. On the basis of temperature dependences of specific electrical conductivity, thermal conductivity and Seebeck coefficient, values of figure of merit ZT were calculated. ZT for n-$Si_{0.7}Ge_{0.3}$ is ~25% higher than for p-$Si_{0.7}Ge_{0.3}$ at the same temperatures. An alloy of this composition, Si_xGe_{1-x} (x=0.7), was chosen because it, together with highly compatible tungsten, gives a switching junction with low electrical resistance.

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INTRODUCTION

Thermoelectric generators (TEGs) are widely used for uninterrupted electrical service of special facilities located in remote regions of the earth, underwater and in outer space. Their use is also relevant for non-electrified regions. SiGe alloys are one of the important high-temperature thermoelectric materials [1,2]. Their working temperature reaches up to 1100°C and is characterized by high efficiency: $Z = \sigma S^2/k \cong (0.6\text{-}1.0)\ 10^{-3}\ K^{-1}$ (σ - specific electrical conductivity, S and k - Seebeck and thermal conductivity coefficients). Considering these characteristics, the potential energy conversion efficiency of thermoelectric SiGe alloys reaches 15%. In addition, SiGe alloys can work in vacuum, inert gas environment and air, which is very important in terms of ease of operation of thermoelectric generator. Thermoelectric SiGe alloys are solid solutions of Si and Ge, whose thermal conductivity is 8-10 times lower than the thermal conductivity of individual components [3]. SiGe alloys with optimal concentration of charge carriers are obtained by adding alloying elements (phosphorus for n-type and boron for p-type). The main goal of alloying is to obtain alloys with the maximum power factor (σS^2). The synthesis of thermoelectric SiGe alloys is traditionally performed by co-melting the components. Melting takes place in a quartz crucible in an inert gas environment at a temperature of 1350-1450°C for 10-20 minutes. The resulting alloy adheres to the quartz and upon cooling, both the crucible and the alloy shatter. Homogenization of the alloy is carried out by zone melting. But the cost of the SiGe alloy produced by this method, taking into account the quartz crucible and the technological operations used, is relatively high.We have developed a method for the synthesis of n- and p-type SiGe alloys using a relatively cheap powder technology. It consists in the joint grinding of the components and hot vacuum pressing of the resulting ultradispersed powder. Polycrystalline Si and Ge are used as the main alloy components, and amorphous phosphorus powder (for n-type) and amorphous boron powder (for p-type) are used as alloying components. On the basis of SiGe and Ge/SiGe alloys, modules for TEG were created both earlier and recently [4,5].

Experimental

Preparation of $Si_{0.7}Ge_{0.3}$alloys: To create a thermoelectric module, n- and p-type $Si_{0.7}Ge_{0.3}$ alloys containing alloying substances with a concentration of $3.2{\cdot}10^{26}\ m^2/V{\cdot}sec$ were fabricated by the vacuum hot pressing method.Massive wastes of Si and Ge were crushed with a steel rod and sieved through a (with 0.2 mm cells) sieve. Then it was loaded into the mill chamber ("REC" PM-100 SM) and ground for 20-25 hours. The powder grain size was assessed using an optical microscope (Nicon) and an X-ray diffractometer (DRON-3M). The disperced $Si_{0.7}Ge_{\ 0.3}$ alloy powder produced in the indicated mode consisted mainly of Si and Ge grains of size 60–80 nm. The resulting powder was pressed in a high-temperature vacuum induction pressure chamber at a temperature of 1200-1320°C and a pressure of 480 $kg{\cdot}cm^{-2}$ for 20-30 minutes. The matrix and punches are made of high-strength graphite (MG). From the obtained briquettes, profiled samples were cut out on a diamond-cutting disk device. Photo of briquette is shown in Fig.1.

Fig.1. Photo (x1) of briquette compacted from ultradispersed$Si_{0.7}Ge_{0.3}$+$P_{0.5}$ (wt.%) alloy powder at 1300°C

Monolithic thermoelectric module: Graphite switching plates were attached to the ends of the alloy branches.The switched sample was placed in the vacuum chamber of an induction furnace, and probes were placed in its switching plates to measure the temperature and electromotive force.One side of the module was heated by a flame generated by gas combustion, which directly hit the surface of the module. On the other side, the module was cooled by running water. Chromel-alumel thermocouples were placed on the hot and cold ends of the module. The monolithic thermoelectric module's cold side electrical insulation node was fabricated using AlN and graphite plates. Both of them are thermomechanically combined with SiGe alloys in a wide temperature range, which is very important for creating a thermostable TEG. Figure 2 shows photos of modules from 16 and 24 branches.

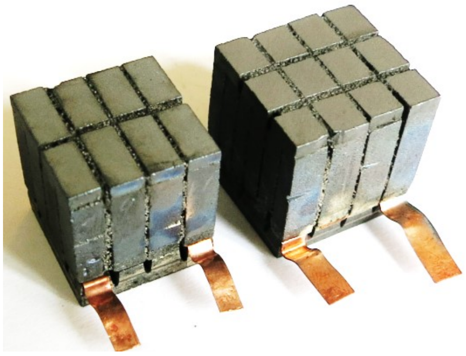


Fig. 2. Photos of modules from 16(2.0x2.0x2.3 cm) and 24 (2.2x2.2x2.3 cm) branches

4 n- and p-type alloy plates (2 n-type and 2 p-type) were taken to make a mini monolithic thermoelectric module containing 16 branches. They were arranged in n-p-n-p order. A mini-monolithic thermoelectric module containing 24 branches was made using a similar technology. The difference is that the monolithic package is made of 6 plates.

RESEARCH OF MODULES

The tables show the measurement results.

Module №1. 16 branches

t_h°C	t_c°C	(t_h-t_c)°C	E, V	S, μV·K^{-1}	R,Ohm	U, V	P, W
410	25	385	1.39	225	0.3	0.62	1.28
455	25	430	1.53	222	0.3	0.68	1.54
485	25	460	1.63	222	0.3	0.72	1.73
509	25	484	1.63	218	0.3	0.74	1.83

Module №2. 16 branches

t_h°C	t_c°C	(t_h-t_c)°C	E, V	S, μV·K^{-1}	R,Ohm	U, V	P, W
395	40	355	2.02	356	0.5	0.96	1.84
424	45	379	2.31	380	0.5	1.07	2.29
443	48	395	2.35	372	0.5	1.1	2.42
483	50	433	2.52	363	0.5	1.15	2.64

Module №3.24branches

t_h°C	t_c°C	(t_h-t_c)°C	E, V	S, μV·K^{-1}	R,Ohm	U, V	P, W
523	59	464	2.34	210	0.5	1.22	2.97
619	72	544	2.61	200	0.5	1.39	3.86
633	79	554	2.76	207	0.5	1.45	4.20
674	84	590	2.89	204	0.5	1.5	4.50

Module №4.24branches

t_h°C	t_c°C	(t_h-t_c)°C	E, V	S, μV·K^{-1}	R,Ohm	U, V	P, W
455	58	397	1.37	144	0.4	0.77	1.48
497	62	435	1.48	142	0.3	0.713	1.69
584	67	517	1.72	138	0.3	0.82	2.24
652	74	578	1.912	138	0.3	0.904	2.74

The electric power (P) of thermoelectric modules is calculated by the formula $P=E^2/R=U^2/4R$, where E is electromotive force, R is theexternal electric resistance and U is the electric voltage generated by the module. As can be seen from the table, the value of P reaches 4.5 W (module N3 from 24 branches).

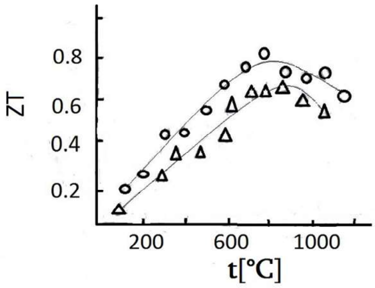


Fig. 3. Temperature dependences of ZT of: (o) n-type $Si_{0.7}Ge_{0.3}$and (Δ) p-type $Si_{0.7}Ge_{0.3}$

Also, on the basis of temperature dependences of specific electrical conductivity ($\sigma=1/\rho = \square/R$), thermal conductivity and Seebeck coefficient, values of figure of merit $ZT= (\sigma S^2/k)T$(T – absolute temperature) were calculated (Fig.3). As we see, ZT for n-$Si_{0.7}Ge_{0.3}$ is ~25% higher than for p-$Si_{0.7}Ge_{0.3}$ at the same temperatures.

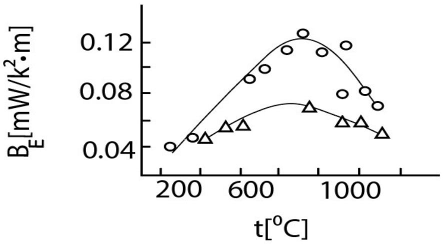
CONCLUSION

$Si_{0.7}Ge_{0.3}$ thermoelectric alloys of n- and p-type were fabricated and their thermoelectric characteristics were studied. These alloys are cut into profiled plates, monolithic packages are assembled from them, switching packages are glued to the lower and upper bases of the packages. These packages are cut into plates 4-5 mm thick and monolithic thermoelectric modules are assembled from them. Electrical insulation units of thermoelectric modules are made on the basis of AlN and graphite fittings. These nodes are connected to thermoelectric modules, resulting in monolithic thermoelectric modules. The energy characteristics of these modules have been studied.

Appendix

We investigated the important characteristic of thermoelectric materials, electronic quality factor (B_E). A rigorous definition of B_E is given by the formula $B_E=(\hbar N_V C_l/3\pi^2 m{*}\xi^2)(k_B/q)^2$, where ħ is reduced Planck's constant, C_l is a combination of elastic constants, ξ is the deformation potential coefficient , m* - electron rest mass, N_V - band degeneracy, q -elementary charge, k_B - Boltzmann's constant. To calculate B_E,based on experimental data (Seebeck coefficient, specific electrical conductivity), the formula is used: $B_E = \sigma S^2/B_S$, where B_S is scaled power factor: $B_S = \frac{q}{k_B}\left[\frac{\frac{qS}{k_B}e^{2-\frac{qS}{k_B}}}{1+e^{-5\left(\frac{qS}{k_B}-1\right)}} + \frac{\frac{\pi^2}{3}S}{1+e^{5\left(\frac{qS}{k_B}-1\right)}}\right]$ [6,7].

The temperature dependence of B_E has a form (Fig.4) that, according to the literature [6], indicates the presence of additional effects, such as band convergence, bipolar effects, additional scattering.



We calculated the effective masses (m*) of charge carriers for some temperatures. The following formula are used for the calculation [7]:

$$m^* \cong \frac{h^2}{2k_BT}\left[\frac{3n}{16\sqrt{\pi}}\left(e^{S_r-2} - 0.17\right)\right]^{2/3}. \quad (1)$$

(Formula (1) is fair when |S|>0.75·10-4V/°K. In our case S=(1.08÷3.07)·10-4V/°K.)

Values m^*/m_0 (m_0 - rest mass) calculated from Eq.(1) for different temperatures are shown in the table 5.

Table 5. Values of m^*/m_0 in $Si_{0.7}Ge_{0.3}$ for different temperatures

t,℃	m^*/m_0	
	$n - Si_{0.7}Ge_{0.3}$	$p - Si_{0.7}Ge_{0.3}$
30	-	1.87
155	2.98	-
230	-	1.65
325	-	1.6
430	-	1.62
445	3.06	-
530	-	1.62
550	3.8	-
625	-	1.66
730	-	1.71
740	4.59	-
855	3.76	-
885	-	-
940	4.16	-
990	2.82	-
1055	2.45	-
1030	-	1.28
1130	1.79	-
1135	-	1.3

The obtained values of are approximate to the corresponding values for some thermoelectrics [8, 9]. The mobility (μ) of charge carriers were also experimentally investigated. For the p-type $Si_{0.7}Ge_{0.3}$, an empirical equation was obtained: $\mu \cong \frac{1}{2}T^{-3/2}$ (T – absolute temperature), which indicates of the phonon scattering of charge carriers [10]. But for n- type $Si_{0.7}Ge_{0.3}$ it is impossible to draw an unambiguous conclusion from the experimental data. Values of μ were obtained on average 0.6 m^2/V·sec. They are approaching data for SiGe alloy at 300°K with the same type of conductivity and concentration of charge carriers [3] and for other alloys at different temperatures [11,12].

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