



# Determination of the Characteristics of Thermoelectrics with High and Ultra-high Values of the Seebeck Coefficient

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## **Authors' contributions**

*This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.*

## **Article Information**

### **Open Peer Review History:**

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**Original Research Article**

**Received: 23/05/2024**

**Accepted: 25/07/2024**

**Published: 30/07/2024**

## **ABSTRACT**

The most important problems facing society today is energy supply of humanity. One of the clear examples of direct energy conversion is thermoelectric effect, which consists in obtaining electric potential using a temperature difference. The obtaining and study of materials with high values of the Seebeck coefficient is one of the main factors for the efficient operation of thermoelectric devices.

This article analyzes works in which high and ultra-high values of the Seebeck coefficient were obtained. The new formula is used to calculate resistivity, carrier concentration or Seebeck coefficient in cases where one of these parameters is not measured or calculated. Important thermoelectric parameters of materials are electronic quality- and scaled power factors.

The scaled power factor (SPF) of a number of thermoelectric materials is analyzed. The graph of its

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dependence on the Seebeck coefficient agrees well and complements the graph known from the literature for a large number (more than 3500) of samples towards higher values of the Seebeck coefficient.

Based on the values of SPF and resistivity (or conductivity), the electronic quality factor of some materials is calculated. Its temperature dependence provides information about the mechanism of charge carrier scattering.

The universal electrical conductivity is calculated. Its temperature dependence and the dependence of the Seebeck coefficient on it confirm that the electronic quality factor scales the thermoelectric parameters of materials.

**Keywords:** Thermoelectric; high and ultra-high values of the seebeck coefficient.

## 1. INTRODUCTION

Devices based on the Seebeck effect are widely used in industrial and household generators, navigation (including space) systems, solar energy converters, etc. The Seebeck coefficient (S) is a measure of the magnitude of the induced voltage. Obtaining and researching of materials with its high values is one of the main factors for the effective operation of the above-mentioned devices (Values of the Seebeck coefficient exceeding  $2 \cdot 10^{-4} \text{V/K}$  are considered to be high values.) An important factor of thermoelectrics is also the scaled power factor, which is given by the expression [1]:

$$B_S = S_r \left[ \frac{S_r e^{(2-S_r)}}{1+e^{-5(S_r-1)}} + \frac{\pi^2}{3[1+e^{5(S_r-1)}]} \right], \quad (1)$$

where  $S_r = (q_e/k_B)S \cong 1.1605 \cdot 10^4 S$  is the reduced Seebeck coefficient, and  $q_e$ ,  $k_B$  are elementary charge and Boltzmann's constant, respectively.

The paper [1] shows also the dependence of the scaled power factor on the Seebeck coefficient for a large number (more than 3500) of samples at  $(0 < S \leq 6) \cdot 10^{-4} \text{V/K}$ . Values of S greater than  $2 \cdot 10^{-4} \text{V/K}$  can already be considered high values of the Seebeck coefficient. In a number of works, ultra-high values of this coefficient ( $> 1 \cdot 10^{-3} \text{V/K}$ ) are given. Here we will consider works in which high and ultra-high values of the Seebeck coefficient were obtained. In such cases, instead of equation (1) you can use:  $B_S \cong S_r^2 e^{(2-S_r)}$ , the accuracy of which increases with increasing Seebeck coefficient, and instead of equation (2) (see below) you can use:  $S_r^2 e^{(2-S_r)} \cong 1.087 \cdot 10^{-9} (\rho n)^{2/3} S$ .

Using the Seebeck coefficient, the effective mass of charge carriers is calculated, and then the mobility is calculated using the formulas given in [2-5].

## 2. METHODOLOGY

As already mentioned in the introduction, in this article we consider works in which high and ultra-high values of the Seebeck coefficient were obtained. We present data for a certain number of different thermoelectric materials. The relevant methods can be found in the cited references. Below we provide data on the method of preparing our samples a silicon-germanium alloy.

We made SiGe alloys with n-type conductivity. The method of joint grinding of the components and vacuum hot pressing of the obtained ultradisperse powder was used. The alloying element for n-type conduction was phosphorus (0.5 wt.%). The concentration of charge carriers was  $3.2 \cdot 10^{20} \text{cm}^{-3}$ . It was defined by measuring the Hall constant at room temperature. The grain sizes of the powder were evaluated with an optical microscope "Nicon" and an X-ray diffractometer DRON-3M. Ultradisperse powder consisted mainly of 60–80 nanometer Si and Ge grains. The resulting powder was pressed in a high-temperature vacuum induction pressure chamber at a temperature of 1200–1320°C and a pressure of  $480 \text{ kg} \cdot \text{cm}^{-2}$  for 20–30 minutes. The matrix and punches are made of high strength graphite. From the obtained briquettes, profiled samples were cut out on a diamond-cutting disk device.

## 3. RESULTS AND DISCUSSION

Fig. 1a,b shows the results calculated from the data of the literature [6-16] for such values of the Seebeck coefficient.<sup>(\*)</sup> These data extend well the graph presented in [1] towards higher values of S. It should be noted that some of the works cited above do not mention the values of the Seebeck coefficient, because the emphasis is on the study of other parameters. However, of the proposed equation [17]: 
$$\frac{e^{(11605S-2)}}{S[1+e^{-5(11605S-1)}]} +$$

$\frac{3527.5}{1+e^{5(11605S-1)}} \cong 1.087 \cdot 10^{-9} (\rho n)^{2/3}$ , S can also be calculated using the values of  $\rho$  (specific resistivity) and  $n$  (charge carrier concentration). This equation can be used for the complete analysis of the papers in which one of the values of S,  $\rho$  or  $n$  is not given. (\*\*).

Using experimentally measured values of the Seebeck coefficient or values calculated by equation (2), as well as the value of specific resistivity (or specific electrical conductivity  $\sigma=1/\rho$ ), it is possible to calculate such an important parameter of thermoelectric materials as the electronic quality factor:  $B_E = \sigma S^2 / B_S$ . By means of its temperature dependence, it is possible to obtain information on the mechanism of scattering of charge carriers in the material [1]. After calculating the electronic quality factor, it is possible to calculate the universal electrical conductivity:  $\sigma' = (q_e / k_B)^2 \sigma / B_E \cong 1.347 \cdot 10^8 \sigma / B_E$ .

In order to substantiate the reliability of the above formulas at high values of the Seebeck

coefficient, let us consider some data from the literature.

In work [18] N-type  $\text{Si}_{0.96}\text{Ge}_{0.04}$  was irradiated with  $^{60}\text{Co}$  gamma photons and the electrical resistance, concentration, and mobility of charge carriers were measured. Seebeck coefficient and other thermoelectric characteristics were not studied in this work. These parameters were calculated in [19] for the temperature range (250-430) °C. Data up to 25°C has been added here (Fig. 2).

The dependences  $B_E - t$  and  $\sigma' \cdot t$  differ significantly from the data for  $\text{Si}_x\text{Ge}_{1-x}$  samples without irradiation (Fig. 3) and from them it is difficult to conclude about the mechanism of charge carrier scattering (see footnote (\*\*)).

In work [20], all parameters of ceramic  $\text{Na}_{0.55}\text{Co}_{0.9}\text{W}_{0.1}\text{O}_2$  were measured or calculated, except for the effective mass ( $m^*$ ) and mobility ( $\mu$ ) of charge carriers. The ratio  $m^*/m_0$  ( $m_0$  - rest mass) can be calculated using the formula:

$$\frac{m^*}{m_0} \cong 1.059 \cdot 10^{-15} \left( \frac{n^{2/3}}{T} \right) \left\{ \frac{3[e^{(S_r-2)} - 0.17]^{2/3}}{1+e^{-5(S_r-S_r^{-1})}} + \frac{S_r}{1+e^{5(S_r-S_r^{-1})}} \right\} \cong \frac{6.608 \cdot 10^{-15}}{T} [ne^{(S_r-2)}]^{2/3} \quad (3)$$

where T is absolute temperature. The value of  $m^*/m_0$  calculated using Eq. (3) at 1167K turned out to be  $3.33 \cdot 10^{-3}$ . This value could be considered very (unacceptable) low, but then, using the formula  $\mu = (m^*/m_0)^{-3/2} \mu_W$  ( $\mu_W$  - weighted mobility), a very real value of mobility was obtained:  $\mu \cong 3.25 \cdot 10^{-2} \text{m}^2/\text{V}\cdot\text{s}$ .

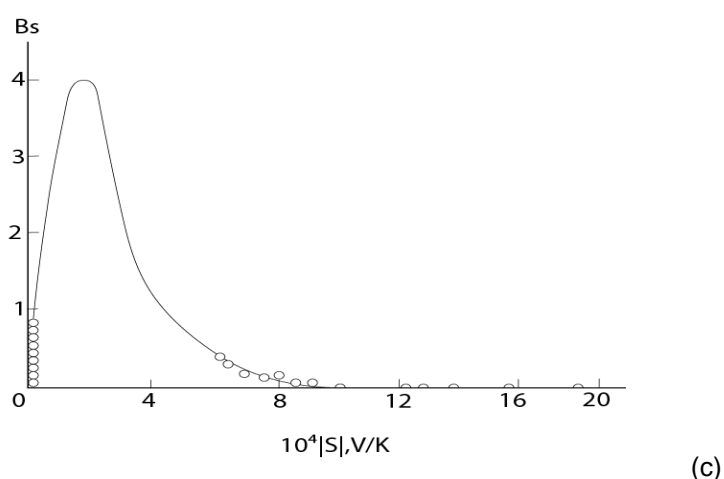
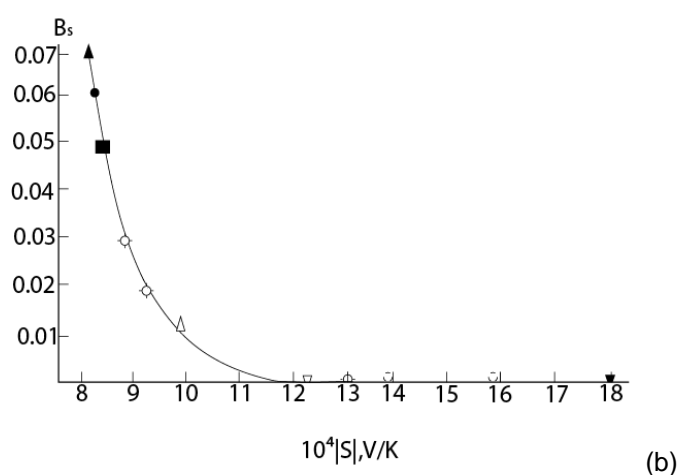
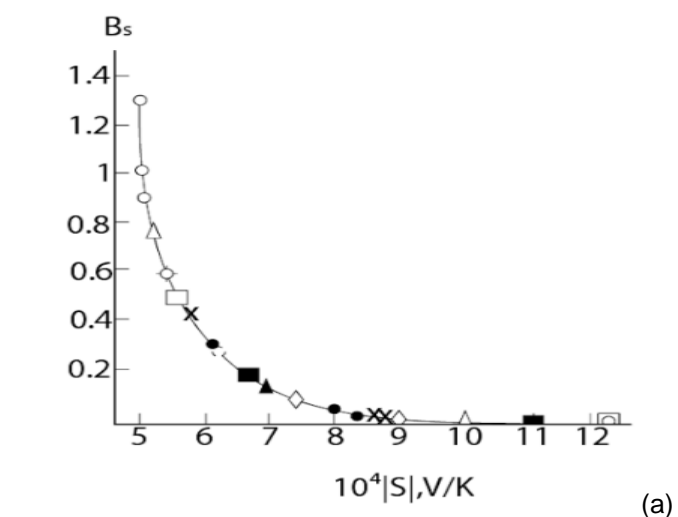
In work [21], all parameters of  $\text{Cu}_{1.98}\text{S}_x\text{Se}_{1-x}$  were determined, except for the electronic quality factor and universal electrical conductivity. From the temperature dependence of  $B_E$  we calculated, it is difficult to draw a definite conclusion about the mechanism of charge carrier scattering. And shown in Fig. 3(b) the temperature dependence of the universal electrical conductivity, together with the data for  $\text{Si}_x\text{Ge}_{1-x}$ , shows that all experimental points form a regular set, despite the differences in the samples. This confirms that the electronic quality factor  $B_E$  scales the electrical conductivity of materials. (\*\*\*)

## FOOTNOTES:

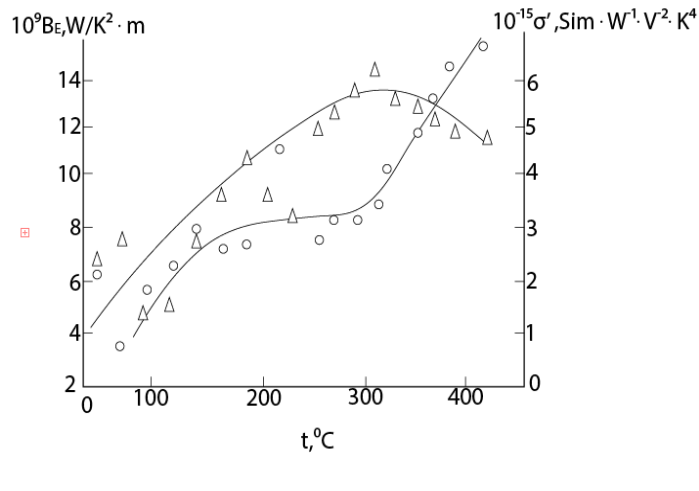
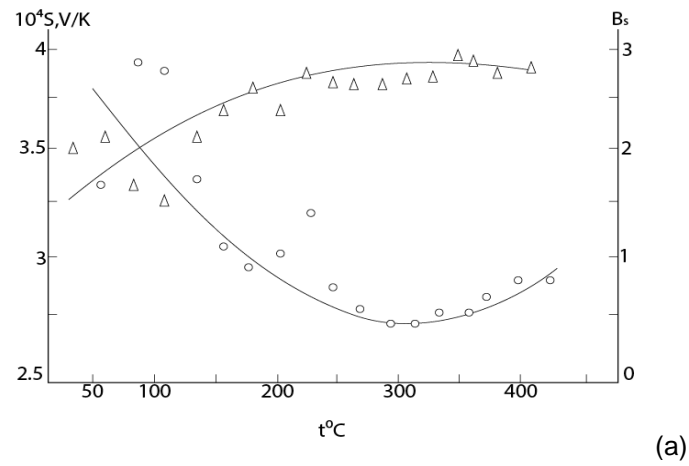
(\*) Points corresponding to higher values of S according to some of the works cited above are not shown based on the scale of the figure. Much higher values were obtained for polymers PEDOT:PSS [22], O+NaOH [23], PSSH+PANI-CNT [17], NFC-PSSNa [24], Nafion, S-PEEK [25], PEDOT-Tos+CNT [26], (poly(3-hexylthiophene)) $_x\text{B}_{1-x}$  [27], as well as for Si(ICs) [28],  $\text{CsGeX}_3$  ( $x=\text{F, Cl, Br}$ ) [29,30] and others.

(\*\*) The shape of the curves in Fig. 3(a) indicates that in  $\text{Si}_x\text{Ge}_{1-x}$ , with increasing temperature, additional scattering is accompanied by convergence of the bands, and then by bipolar effects.

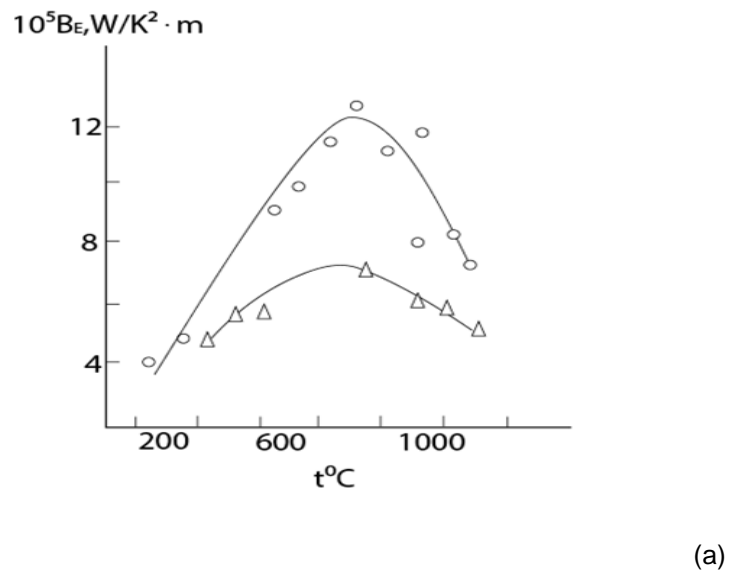
(\*\*\*)  $B_E$  scales the electrical conductivity also in the  $\sigma'$ -S and  $\sigma'$ - $\sigma$  coordinates [31-33].

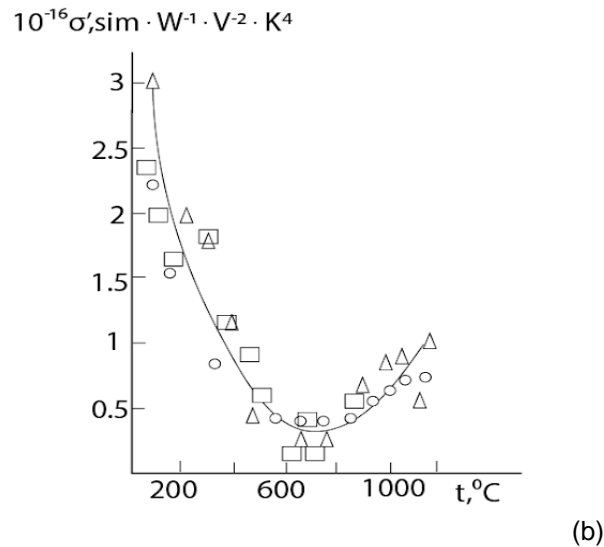


**Fig. 1. (a) Dependence of the scaled power factor on the Seebeck coefficient, calculated according to the data for following thermoelectrics: (o)Cu<sub>3</sub>Sb<sub>1-x</sub>Bi<sub>x</sub>Se<sub>4</sub> [6], (Δ)SrTiO<sub>3</sub> [7], (□)CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> [8], (○) SiGe [9], (●) SnSe<sub>1-x</sub>Te<sub>x</sub> [10], (■) GCuO [11], (◊) PbSb<sub>x</sub>Te<sub>1-x</sub> [12], (◻) PtSe<sub>2</sub> [13], (▲) CuGeTe [14], (▼) Si [15]; (x) La<sub>0.95</sub>Sr<sub>0.05</sub>CoO<sub>3</sub> [16]; solid line is a fragment of the averaged curve given in [1]; (b) the same - in another scale on the ordinate axis; (c) - curve from [1] to the right with points from Figs. (a) and (b), points on the left correspond to ultra-low values of the Seebeck coefficient (unpublished data)**



**Fig. 2. (a) Dependences (o)  $S - t$  and ( $\Delta$ )  $B_s - t$  for  $Si_{0.96}Ge_{0.04}$  irradiated with gamma photons, ( $t$  – isochronal annealing temperature). (b)  $B_E - t$  (o) and  $\sigma' - t$  dependences ( $\Delta$ )**





**Fig. 3. (a)  $B_E - t$  dependences for  $Si_{0.7}Ge_{0.3}$ : (o) N-type, ( $\Delta$ ) P-type [31,32].(b)  $\sigma' - t$  dependences for N-type: (o)  $Si_{0.7}Ge_{0.3}$  and ( $\Delta$ )  $Si_{0.83}Ge_{0.17}$  and ( $\square$ )  $Cu_{1.98}S$  [19,32,33]**

#### 4. CONCLUSION

This Article analyzes works in which high and ultra-high values of the Seebeck coefficient were obtained. A new formula is used to calculate resistivity, carrier concentration or Seebeck coefficient in cases where one of these parameters is not measured or calculated. The scaled power factor, universal electrical conductivity and electronic quality factor of a number of thermoelectric materials has been analyzed. Temperature dependence of electronic quality factor indicates that in alloy  $Si_xGe_{1-x}$  with increasing temperature additional scattering is accompanied by band convergence, and then by bipolar effects. Temperature dependence of universal electrical conductivity confirm that the electronic quality factor scales thermoelectric parameters. The data for  $B_s$  extend well the graph presented in the literature towards higher values of  $S$ .

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#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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