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**COMPUTER METHOD OF
DESCRIPTION OF THE TECHNOLOGIES
AND PROPERTIES OF Bi_2-Te_3 -BASED THERMOELECTRIC
MATERIALS OBTAINED BY THE PRESSING METHOD**

This paper presents the results of the study of literary sources describing the technologies and properties of thermoelectric materials obtained by the pressing method. The results of one of the stages of creating a software product for the description of the production technologies and properties of thermoelectric material based on Bi-Te compounds are given. Bibl. 13, Fig. 2, Table 1.

Key words: hot pressing method, cold pressing method, dynamic elements, bismuth telluride

Introduction

Thermoelectric materials science is an important direction in the development of thermoelectricity, since advances in this area as a whole determine the possibilities and versatility of practical uses of thermoelectric energy conversion [1]. Increasing the efficiency of thermoelectric converters is a rather important and widespread problem. The characteristics of thermoelectric materials can be determined by the formula:

$$Z = \frac{\alpha^2 \sigma}{\kappa}, \quad (1)$$

where α is the Seebeck coefficient, σ is electrical conductivity, κ is thermal conductivity.

One of the ways to obtain thermoelectric materials is the pressing method. An important advantage of pressed thermoelectric materials (TEMs) based on Bi_2Te_3 is their high mechanical strength compared to crystallizing materials from a melt. In addition, powder metallurgy contributes to increased productivity and savings. Materials obtained by powder pressing, as a rule, have a lower value of figure of merit Z due to misorientation of grains in the bulk of the material [2]. The purpose of this work is to study the thermoelectric characteristics of solid solutions based on bismuth telluride

obtained by pressing, as well as application of a modified computer program to study the pressing method and characteristics of thermoelectric materials based on $Bi-Te$ compounds.

Dependence of the thermoelectric characteristics of Bi_2Te_3 -based materials obtained by the pressing method

Pressed Bi_2Te_3 -based materials are obtained from a powder of a preliminarily synthesized material from a mixture of powders of initial components taken in a stoichiometric ratio [3]. Two pressing methods are used: cold pressing, which consists in briquetting the powder in a cold mold, followed by sintering in vacuum, in an atmosphere of hydrogen, an inert gas, and hot pressing of the powder in a heated mold with additional annealing of the sample. Materials obtained by the pressing methods have increased strength due to grain boundaries that prevent the propagation of cracks along cleavage planes. In addition, this method is relatively inexpensive. One of the important thermoelectric characteristics of pressed materials is the ability to withstand shock loads and thermal stresses. The thermoelectric characteristics of $Bi-Te$ -based materials obtained by pressing are indicated in Table 1.

Table 1

*Production technologies and properties of thermoelectric materials
obtained by the pressing method*

Working temperature, K	$Z, 10^{-3}, K^{-1}$	$\alpha, mV/K$	$\sigma, \Omega m^{-1} cm^{-1}$	$\kappa, W/m \cdot K$	Material type	Material composition	Ingot length, mm	Ingot diameter, mm	Pressing pressure, MPa	Pressing temperature, K	Reference:
613-723	1.7	160	-	1.4	N	$(Bi_2Te_3)_{0.95}$ $(Bi_2Se_3)_{0.05}$	-	10 × 1	60	-	[1]
450-530	1.4	290	454	1.3	P	$Bi_{0.5}Sb_{1.5}Te_3$	-	-	30	480	[2]
100-250	1.4	215	-	1.2	P	$BiSbTe$	-	1.25 - 25	-	-	[3]
380, 400, 420	2.69	223	-	0.95 1.09 1.17	P	$Bi_{0.5}Sb_{0.5}$ $Te_{0.5}$	-	30	200	-	[4]
700	3	-	-	-	N	$Bi_{0.5}Sb_{0.5}$ $Te_{0.5}$	5 × 10	-	30	400-585	[5]
400-500	3	226	780	1.17	P	$Bi_2Te_3-Bi_2Se_3$	-	-	700	450	[6]
25-250	0.45	171	0,25	0.55	P	25% Bi_2Te_3 + 75% Sb_2Te_3	-	-	70	-	[7]

Continuation of Table

623-773	1.92	235	533	1.53	<i>P</i>	$Bi_{0.5}Sb_{1.5}Te_3$ (120:1)	$5 \times 5 \times 10$	-	500	500	[8]
533-693	0.71	180	-	0.61	<i>N</i>	$(Bi_{0.25}Sb_{0.75})_2Te_3$	20×13	-	-	-	[9]
200-700	2.52	180	900	1.4	<i>N</i>	$Bi_2Te_{2.88}Se_{0.12}$ $Bi_{0.52}Sb_{1.48}Te_3$	$30 \times 30 \times 20$	-	-	-	[10]
200-700	2.41	180	900	1.4	<i>P</i>	$Bi_{0.52}Sb_{1.48}Te_3$	$30 \times 30 \times 20$	-	-	-	[10]
200-600	1.65	175	890	16.5	<i>N</i>	$Bi_2Te_{2.3}Se_{0.7}$	-	-	-	-	[11]
200-600	2.45	182	1250	16.9	<i>P</i>	$Bi_{0.56}Te_{2.9}$ $Sb_{1.44}Se_{0.1M}$	-	-	-	-	[11]
300-550	3	-	-	-	<i>P</i>	$Bi_{0.4}Sb_{16}$ $Te_3 + Pb$	-	-	800	-	[12]
300-550	2	-	-	-	<i>P</i>	$Bi_{0.4}Sb_{1.6}Te_3$	-	-	800	-	[12]
100-400	3.12	171	-	-	<i>P</i>	$Sb_{1.51}Bi_{0.49}Te_3$	$4 \times 2 \times 2$	-	1200	-	[13]

All the data in the table were implemented in the software product to describe the technologies and properties of the thermoelectric $Bi-Te$ - based material. Further updating the software product database will be described in future articles.

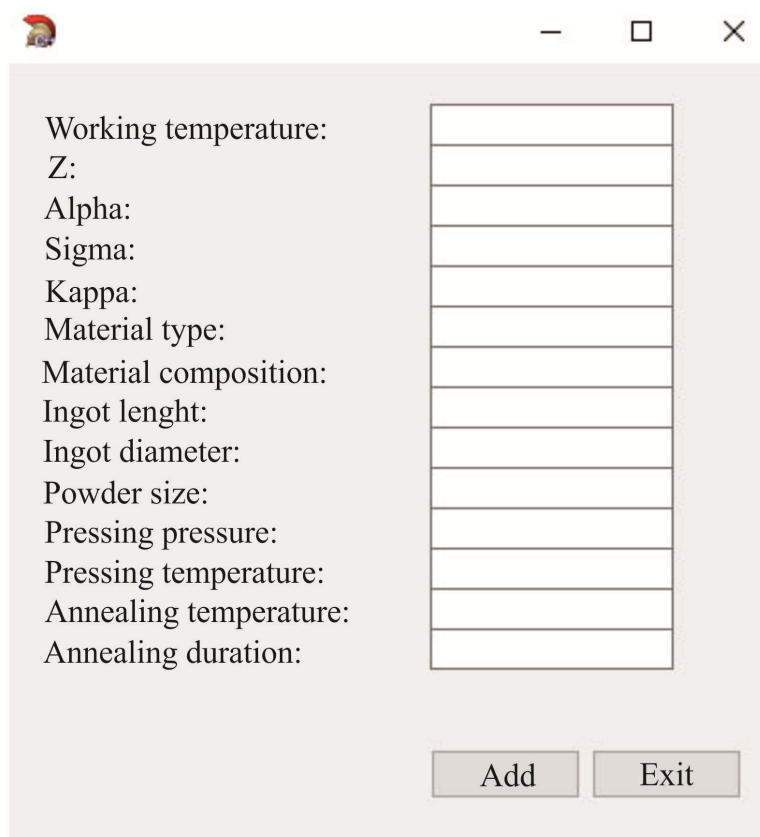
Further updating the software product to describe the technologies and properties of $Bi-Te$ -based thermoelectric material

Currently, the function of adding new records has been implemented into the software product, which contains data on the growing technology and characteristics of the thermoelectric material based on $Bi-Te$ compounds. The general algorithm of this function is as follows.

- Calling the add function by the user.
- Creation of a dynamic form and all its components, according to the chosen method of obtaining thermoelectric materials.
- After the user enters all the necessary data about the mode of obtaining thermoelectric material, the program checks the correctness of the data.
- The program switches to data adding mode.
- A new record is created in the database.
- The program switches to working mode.

- Delete the dynamic form and all its components.

The general view of record adding window is presented in Fig. 1.



The screenshot shows a standard Windows-style window with a title bar containing a small icon and three control buttons (minimize, maximize, close). The main area of the window is light gray and contains a list of labels on the left and a column of empty text input boxes on the right. The labels are: 'Working temperature:', 'Z:', 'Alpha:', 'Sigma:', 'Kappa:', 'Material type:', 'Material composition:', 'Ingot lenght:', 'Ingot diameter:', 'Powder size:', 'Pressing pressure:', 'Pressing temperature:', 'Annealing temperature:', and 'Annealing duration:'. At the bottom right of the window, there are two buttons labeled 'Add' and 'Exit'.

Fig. 1. General view of record adding window.

The function of editing existing records was also implemented. The general algorithm of this function is as follows.

- Calling the editing function by the user.
- Creation of a dynamic form and all its components, according to the chosen method of obtaining thermoelectric materials.
- Transfer of information from the selected record to the editing window.
- After the user makes all the necessary corrections in the thermoelectric material acquisition mode, the program checks the correctness of the data.
- The program switches to data editing mode.
- Editing of the selected record in the database.
- Transition of the program into working mode.
- Delete the dynamic form and all its components.

The general view of record editing window is presented in Fig. 2

It should be noted that depending on the chosen method of obtaining thermoelectric material, a corresponding window for adding and editing records about the mode of obtaining thermoelectric material is created. Further development of the software product will be described in future articles.

Working temperature:	700
Z:	3
Alpha:	
Sigma:	
Kappa:	
Material type:	
Material composition:	N
Ingot lenght:	Bi2Te3-Bi2Se3
Ingot diameter:	5*10
Powder size:	
Pressing pressure:	30
Pressing temperature:	400-585
Annealing temperature:	
Annealing duration:	

Edit Exit

Fig. 2. General view of record editing window.

Conclusions

1. A study of literary sources describing *Bi-Te*-based thermoelectric materials obtained by pressing was carried out.
2. The research data were added to the database of the software product to describe the technologies and properties of obtaining *Bi-Te*-based thermoelectric material.
3. New functions were introduced into the software product to describe the technologies and properties of obtaining *Bi-Te*-based thermoelectric material.
4. Further versions of the software product will be described in the future articles.

References

1. Yang J.Y, Fan X.A., Chen R.G., Zhu W., Bao S.Q., Duan X.K. (2006). Consolidation and thermoelectric properties of n-type bismuth telluride based materials by mechanical alloying and hot pressing. *Journal of Alloys and Compounds*, 270 - 273.
2. Haoxiang Wei, Jiaqi Tang, Dongyan Xu. Effect of abnormal grain growth on thermoelectric properties of hot-pressed *Bi_{0.5}Sb_{1.5}Te₃* alloys. // *Journal of Alloys and Compounds*. – 2020.
3. Poudel Bed, Hao Qing, Ma Yi, Lan Yucheng, Minnich Austin, Yu Bo, Yan Xiao, Wang Dezhi, Muto Andrew, Vashae Daryooshe, Chen Xiaoyuan, Liu Junming, Dresselhaus Mildred S., Chen Gang, Ren Zhifeng (2008). High-thermoelectric performance of nanostructured bismuth antimony telluride bulk alloys, *Journal of Alloys and Compounds*, 634-638.

4. Seo J., Park K., Lee D. and Lee C. (1998). Microstructure and thermoelectric properties of P-TYPE $Bi_{0.5}Sb_{0.5}Te_{0.5}$ compounds fabricated by hot pressing and hot extrusion. *Scripta Materialia*, 38 (3), 477 - 484.
5. Ha Heon Phil, Oh Young Joo, Hyun Dow Bin and Yoon Eui Pak (2002). Thermoelectric properties of n-type bismuth telluride based alloys prepared by hot pressing and zone melting method. *Int. J. Soc. Mater. Eng. Resour.*, 10(2), Sept (2002).
6. Kim Taek-Soo Kim, Kim Ik-Soo, Kim Taek-Kyung, Hong Soon-Jik, Chun Byong-Sun (2002). Thermoelectric properties of p-type $25\%Bi_2Te_3+75\%Sb_2Te_3$ alloys manufactured by rapid solidification and hot pressing. *Materials Science and Engineering*, B90, P. 42 - 46.
7. Shi Jianxu, Chen Hualing, Jia Shuhai, Wang Wanjun (2018). Rapid and low-cost fabrication of thermoelectric composite using low-pressure cold pressing and thermocuring methods. *Materials Letters*, 299 - 302.
8. Kavei G., Ahmadi K and Seyyedi A. (2011). Hot pressing effect on $(Bi_{0.25}Sb_{0.75})_2Te_3$ mechanical and thermoelectric properties. *Bull. Mater. Sci.*, 34 (7), 1591 - 1597.
9. Fan Xi'an, Rong Zhenzhou, Yang Fan, Cai Xinzhi, Han Xuewu, Li Guangqiang (2015). Effect of process parameters of microwave activated hot pressing on the microstructure and thermoelectric properties of Bi-Te-based alloys. *Journal of Alloys and Compounds*, 282-287.
10. Shtern Yu.I. (2008). Issledovaniie elektyofizicheskikh svoistv i opredeleniie mekhanizmov teplo- i elektroprovodnosti v termoelektricheskikh materialakh na osnove Bi_2Te_3 [Study of electrophysical properties and determination of the mechanisms of thermal and electrical conductivity in Bi_2Te_3 -based thermoelectric materials]. *National Research University of Electronic Technology (MIET)* [in Russian].
11. Simkin A. V., Biriukov A. V., Repnikov N. I., Khovailo V. V. (2015). Termoelektricheskaia effektivnost nizkotemperaturnykh generatornykh materialov i vozmozhnosti yeio povysheniia [Thermoelectric figure of merit of low-temperature generator materials and possibilities of its improvement]. *Bulletin of Cheliabinsk State University*, 7. Physics. Issue 20, 21–29.
12. Ben-Yehuda O., Shuker R., Gelbstein Y., Dashevsky Z. and Dariel M. P. (2007). Highly textured Bi_2Te_3 -based materials for thermoelectric energy conversion. *Journal of Applied Physics*, 101,113707.
13. Navratil J., Sary Z. and Plechzek T. (1996). Thermoelectric properties of p-type antimony bismuth telluride alloys prepared by cold pressing. *Materials Research Bulletin*, 31 (12), 1559 - 1566.

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КОМП'ЮТЕРНИЙ МЕТОД ОПИСУ ТЕХНОЛОГІЙ ТА ВЛАСТИВОСТЕЙ ТЕРМОЕЛЕКТРИЧНИХ МАТЕРІАЛІВ НА ОСНОВІ $\text{Bi}_2\text{-Te}_3$, ОТРИМАНИХ МЕТОДОМ ПРЕСУВАННЯ

У даній роботі наводяться результати дослідження літературних джерел в яких описуються технології та властивості термоелектричних матеріалів отриманих методом пресування. Наводяться результати одного з етапів створення програмного продукту для опису технологій отримання та властивостей термоелектричного матеріалу на основі сполук Bi-Te . Бібл. 13. рис. 2. табл. 1.

Ключові слова: метод гарячого пресування, метод холодного пресування, динамічні елементи, телурид вісмуту.

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КОМПЬЮТЕРНЫЙ МЕТОД ОПИСАНИЯ ТЕХНОЛОГИЙ И СВОЙСТВ ТЕРМОЭЛЕКТРИЧЕСКИХ МАТЕРИАЛОВ НА ОСНОВЕ $\text{Bi}_2\text{-Te}_3$, ПОЛУЧЕННЫХ МЕТОДОМ ПРЕССОВАНИЯ

В данной работе приводятся результаты исследования литературных источников, в которых описываются технологии и свойства термоэлектрических материалов полученных методом прессования. Приводятся результаты одного из этапов создания программного продукта для описания технологий получения и свойств термоэлектрического материала на основе соединений Bi-Te . Библ. 13. рис. 2. табл. 1.

Ключевые слова: метод горячего прессования, метод холодного прессования, динамические элементы, теллурид висмута.

References

1. Yang J.Y, Fan X.A., Chen R.G., Zhu W., Bao S.Q., Duan X.K. (2006). Consolidation and thermoelectric properties of n-type bismuth telluride based materials by mechanical alloying and hot pressing. *Journal of Alloys and Compounds*, 270–273.
2. Wei Haoxiang, Tang Jiaqi, Xu Dongyan (2020). Effect of abnormal grain growth on thermoelectric properties of hot-pressed $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ alloys. *Journal of Alloys and Compounds*.

- Haoxiang Wei, Jiaqi Tang, Dongyan Xu. Effect of abnormal grain growth on thermoelectric properties of hot-pressed $Bi_{0.5}Sb_{1.5}Te_3$ alloys. // *Journal of Alloys and Compounds*. – 2020.
3. Poudel Bed, Hao Qing, Ma Yi, Lan Yucheng, Minnich Austin, Yu Bo, Yan Xiao, Wang Dezhi, Muto Andrew, Vashaee Daryooshe, Chen Xiaoyuan, Liu Junming, Dresselhaus Mildred S., Chen Gang, Ren Zhifeng (2008). High-thermoelectric performance of nanostructured bismuth antimony telluride bulk alloys, *Journal of Alloys and Compounds*, 634 - 638.
 4. Seo J., Park K., Lee D. and Lee C. (1998). Microstructure and thermoelectric properties of P-TYPE $Bi_{0.5}Sb_{0.5}Te_{0.5}$ compounds fabricated by hot pressing and hot extrusion. *Scripta Materialia*, 38 (3), 477 - 484.
 5. Ha Heon Phil, Oh Young Joo, Hyun Dow Bin and Yoon Eui Pak (2002). Thermoelectric properties of n-type bismuth telluride based alloys prepared by hot pressing and zone melting method. *Int. J. Soc. Mater. Eng. Resour.*, 10(2), Sept (2002).
 6. Kim Taek-Soo Kim, Kim Ik-Soo, Kim Taek-Kyung, Hong Soon-Jik, Chun Byong-Sun (2002). Thermoelectric properties of p-type 25 % Bi_2Te_3 +75 % Sb_2Te_3 alloys manufactured by rapid solidification and hot pressing. *Materials Science and Engineering*, B90, P. 42 - 46.
 7. Shi Jianxu, Chen Hualing, Jia Shuhai, Wang Wanjuan (2018). Rapid and low-cost fabrication of thermoelectric composite using low-pressure cold pressing and thermocuring methods. *Materials Letters*, 299 - 302.
 8. Kavei G., Ahmadi K and Seyyedi A. (2011). Hot pressing effect on $(Bi_{0.25}Sb_{0.75})_2Te_3$ mechanical and thermoelectric properties. *Bull. Mater. Sci.*, 34 (7), 1591 - 1597.
 9. Fan Xi'an, Rong Zhenzhou, Yang Fan, Cai Xinzhi, Han Xuewu, Li Guangqiang (2015). Effect of process parameters of microwave activated hot pressing on the microstructure and thermoelectric properties of Bi-Te-based alloys. *Journal of Alloys and Compounds*, 282 - 287.
 10. Shtern Yu.I. (2008). Issledovaniie elektofizicheskikh svoystv i opredeleniie mekhanizmov teplo- i elektroprovodnosti v termoelektricheskikh materialakh na osnove Bi_2Te_3 [Study of electrophysical properties and determination of the mechanisms of thermal and electrical conductivity in Bi_2Te_3 -based thermoelectric materials]. *National Research University of Electronic Technology (MIET)* [in Russian].
 11. Simkin A.V., Biriukov A.V., Repnikov N.I., Khovailo V.V. (2015). Termoelektricheskaya effektivnost nizkotemperaturnykh generatornykh materialov i vozmozhnosti yego povysheniia [Thermoelectric figure of merit of low-temperature generator materials and possibilities of its improvement]. *Bulletin of Cheliabinsk State University*, 7. Physics. Issue 20, 21 - 29.
 12. Ben-Yehuda O., Shuker R., Gelbstein Y., Dashevsky Z. and Dariel M. P. (2007). Highly textured Bi_2Te_3 -based materials for thermoelectric energy conversion. *Journal of Applied Physics*, 101, 113707.
 13. Navratil J., Sary Z. and Plechzek T. (1996). Thermoelectric properties of P-type antimony bismuth telluride alloys prepared by cold pressing. *Materials Research Bulletin*, 31 (12), 1559 - 1566.

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