Special Double Issue Article

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Development of Inexpensive SiGe–$FeSi_2$ Thermoelectric Nanocomposites

Abstract: This investigation presents cost-effective fabrication of n-type silicon germanium–iron disilicide nanocomposites using low-cost low-purity germanium–germanium oxide powder. Moreover, the probability of the reduction of oxide powders during the synthesis process was studied. X-ray diffraction (XRD) analysis indicated reduction of germanium oxide impurity in the synthesized material. Scanning electron microscopy (SEM) along with the energy-dispersive spectrometer (EDS) showed a structure of dispersed iron disilicide particles embedded in silicon germanium matrix. The nanocomposite structure showed a moderate figure-of-merit, ZT, equal to 0.8. The ZT enhancement was related to both the nanocomposite structure and the reduction of germanium oxide.

Keywords: silicon germanium, iron disilicide, germanium oxide, nanocomposite, thermoelectric

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Introduction

Thermoelectric (TE) technology can be used for power generation, cooling or sensing and imaging applications, which has attracted much attention lately resulting in the development of new materials, devices and characterization techniques (Dresselhaus et al. 2007; Satyala, Norouzzadeh, and Vashaee 2014; Zhang et al. 2003; Fan et al. 2001; Tayebi, Zamanipour, and Vashaee 2014; Christofferson et al. 2001). TE materials are promising for production of clean energy due to their capability of converting heat into electricity. Therefore, they can play an important role in the global sustainable energy solution. In particular, power generation from automotive exhaust and industrial processes can convert enormous amounts of unused waste heat into electricity (Goldsmid 1964; Rowe 2005).

The efficiency of the TE material is directly related to the dimensionless figure of merit, ZT, defined as (Goldsmid 1964):

$$\mathrm{ZT} = \frac{S^2\sigma}{k}T \tag{1}$$

where S, σ, k and T are Seebeck coefficient, electrical conductivity, thermal conductivity and the absolute temperature at which the properties are measured, respectively.

Among different TE materials, several silicon- and germanium-based alloys have taken much attention due to their good TE properties. Nanostructuring approaches have been lately applied to such alloys in order to further enhance their TE properties (Satyala, Krasinski, and Vashaee 2014; Satyala and Vashaee 2012; Norouzzadeh, Myles, and Vashaee 2014). Among these alloys, silicon germanium (SiGe) has shown superior TE properties in the temperature range of 700–1,100°C (Vining 1991; Zamanipour et al. 2013; Zamanipour et al. 2012). The importance of the heavily doped SiGe alloys as a high temperature TE material was verified in 1960s (Abeles et al. 1962; Dismukes et al. 1964; Rowe 2005). In order to use them in TE devices, their synthesis, sintering process and properties have been extensively studied both theoretically and experimentally (Dismukes and Ekstrom

1965; Rowe and Bunce 1969; Vining 1991; Minnich et al. 2009; Nozariasbmarz et al. 2013).

In early 1990s, nanostructuring approach was proposed to increase ZT in TE materials via power factor enhancement and thermal conductivity reduction (Hicks and Dresselhaus 1993). The thermal conductivity reduction is due to increased phonon scatterings at the enhanced number of interfaces. The experimental results have proven advantages of this approach in different structures such as in quantum dot superlattice $PbSe_{0.98}Te_{0.02}$/PbTe (Venkatasubramanian et al. 2001; Harman et al. 2002). Bulk nanostructured SiGe has also been extensively studied. A significant enhancement in ZT was reported in p-type and n-type bulk nanostructured $Si_{0.8}Ge_{0.2}$ (Joshi et al. 2008; Zebarjadi et al. 2011; Mohebali et al. 2015; Nozariasbmarz et al. 2014). The ZT improvement was also demonstrated in SiGe/Si superlattice films (Lee, Cahill, and Venkatasubramanian 1997; Zhang et al. 2006; Zeng et al. 2002) and Si nanowires (Hochbaum et al. 2008; Boukai et al. 2008).

Power factor can also be improved using composite structure which was proposed in 1991 (Bergman and Levy 1991) and later (Bergman and Fel 1999) when an effective-medium theory was developed to calculate the power factor of a composite of two different TE materials. Some experimental studies have shown power factor enhancement in $Si/Si_{0.8}Ge_{0.2}$ (Zebarjadi et al. 2011), InGaAs/ErAs (Zide et al. 2006; Bahk et al. 2011), Bi/Cu (Kanno et al. 2009) and Bi/Ag (Hermans and Jaworski 2008).

The theoretical model of nanocomposites for silicide-SiGe has shown promising TE properties (Mingo et al. 2009). However, few experiments have been made on developing nanocomposites. In our previous work, we studied $CrSi_2$ nano-inclusions embedded in $Si_{0.8}Ge_{0.2}$. We showed the enhancement of power factor in $Si_{0.8}Ge_{0.2}$–$CrSi_2$ nanocomposite resulting from the enhancement of charge carrier mobility (Zamanipour and Vashaee 2012).

In this study, a cost-effective fabrication of $Si_{80}Ge_{20}$–$FeSi_2$ composite has been investigated. Although we used low-purity components, the effect of composite structure resulted in high TE properties of the material. Time-consuming milling for mechanical alloying was replaced by induction melting to reach alloy of SiGe in much shorter time. Moreover, the effect of in situ reduction of oxide impurity during the process was studied.

Experimental procedure

Powders of Si (99% purity), Ge (70 %Ge and 30% GeO_2), P (99% purity) and Fe (99% purity) were weighted and mixed together according to the stoichiometric ratio of $(Si_{0.80}Ge_{0.20}P)_{0.95}(FeSi_2)_{0.05}$. Different samples were prepared according to the following three steps. In step I, the mixture of the powders was loaded into a boron nitride (BN) crucible and mounted in the centre of a graphite crucible in a sealed quartz tube. The assembly was placed into an induction furnace to reach to the melting point of the composite. A $4 \times 4 \times 20$ mm rod and a disk of diameter 6 mm were cut out with similar axis direction from the ingot for TE property measurements. In step II, the processed ingot was broken into small pieces (< 1 mm), loaded in a tungsten carbide (WC) cup and milled for 5 h with ball-to-powder ratio (BPR) of 2 at 950 rpm in a Fritsch-P7PL planetary ball mill to reduce the particle size. The powder was again loaded into the induction furnace following similar procedure and rods and disks with similar sizes were prepared for TE property measurement. In step III, the ingot was crashed again into small pieces and milled for another 5 h. In this step, carbon was added to the powder to help in situ reduction of the GeO_2 impurity. The milled powder was melted for the third time and similar rods and disks were cut from the ingot for TE characterizations. The final ingot was again milled for 5 h in similar configuration. The powders were loaded into a graphite die with an inner diameter of 6 mm and hot pressed at 1,150°C. Rods and disk of 6mm in diameter were cut from the hot-pressed samples and characterized for comparison with the induction-melted samples.

Table 1 shows the different process parameters.

Table 1: Conditions of the samples.

Sample ID	Composition	No. of induction melting	Milling time (h)	Hot press temperature (°C)	Soaking time (min)	Density (g/cm^3)
S1	$(Si_{80}Ge_{20}P)_{0.95}(FeSi_2)_{0.05}$	1	–	–	–	2.95
S2	$(Si_{80}Ge_{20}P)_{0.95}(FeSi_2)_{0.05}$	2	–	–	–	3.05
S3	$(Si_{80}Ge_{20}P)_{0.95}(FeSi_2)_{0.05}$	2	5	1,150	7	2.98
S4	$(Si_{80}Ge_{20}P)_{0.95}(FeSi_2)_{0.05}$-C	3	–	–	–	2.93
S5	$(Si_{80}Ge_{20}P)_{0.95}(FeSi_2)_{0.05}$-C	3	5	1,150	7	3.14

The phase identification and morphology of the samples were characterized using x-ray diffraction (XRD, Bruker AXS D8-Discover) and scanning electron microscopy (SEM, Hitachi S-4800) equipped with an Oxford Inst. energy-dispersive spectrometer (EDS). The electrical conductivity and Seebeck coefficient were simultaneously measured by a four-probe method using the commercially available Ulvac, ZEM-3 instrument in the range of 28–950°C. The thermal conductivity was measured using laser flash instrument (Netzsch's LFA 457 Micro-Flash). The accuracy of the data from the ZEM is within 5% and from the laser flash is typically within 10%.

Results and discussion

The XRD data of S4 and S5 are shown in Figure 1. The peaks for $Si_{0.80}Ge_{0.20}$ and $FeSi_2$ are observed in the spectra. It is shown that there are no measurable phases or any contamination in the composite structure.

SEM-EDS micrograph of the surface of a broken piece of S3 shows a uniform distribution of Si and Ge in the sample (Figure 2). It seems that $FeSi_2$ phase as an Fe-rich phase is uniformly dispersed in the microstructure, which is expected to increase the phonon scattering and decrease the thermal conductivity of the composite.

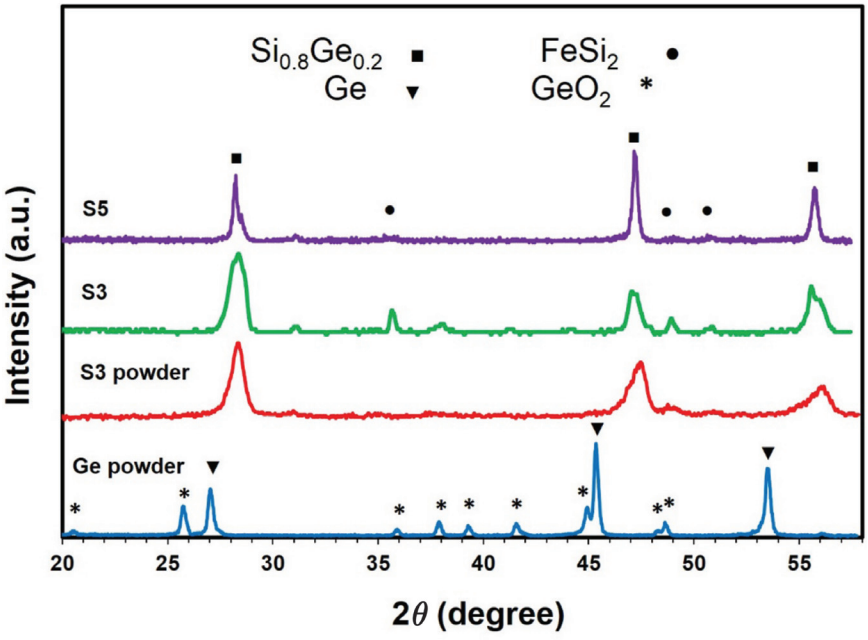

Figure 1: XRD patterns of the composites S4 and S5.

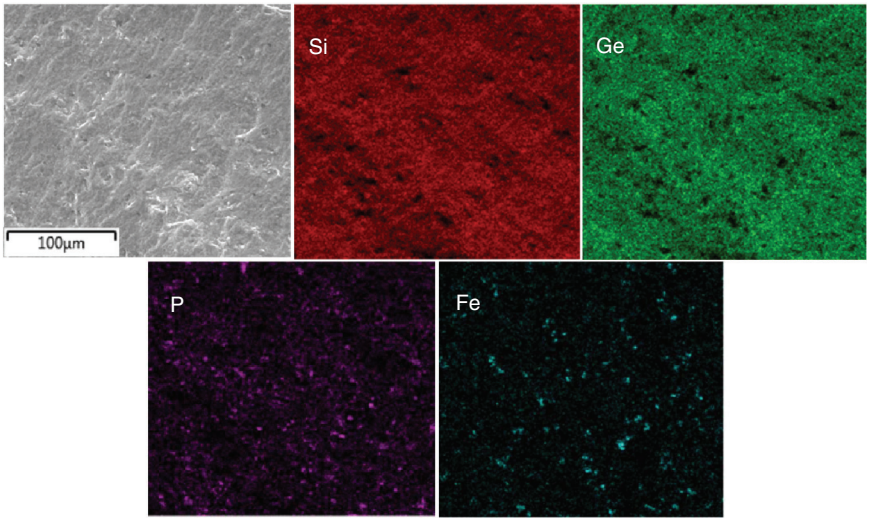

Figure 2: SEM-EDS of S3.

Comparing TE properties of the samples S1 and S2, we can see ZT enhancement in S2 due to the formation of SiGe alloy (Figure 3). Thermal conductivity of S1 is high which indicates existence of non-alloyed silicon. In comparison to S1, both the electrical conductivity and the absolute value of the Seebeck coefficient are higher in S2.

Figure 4 shows the electrical conductivity, the Seebeck coefficient, the power factor times temperature (PFT), and the thermal conductivity of S3, S4 and S5 samples. Hot-pressed samples show better ZT in comparison to the induction-melted samples with similar compositions, which is mainly associated with their smaller grain size.

In S4 and S5, due to the 30% GeO_2 impurity in the initial powders, 30% carbon was added to the stoichiometric amount of Ge to study the effect of reduction of

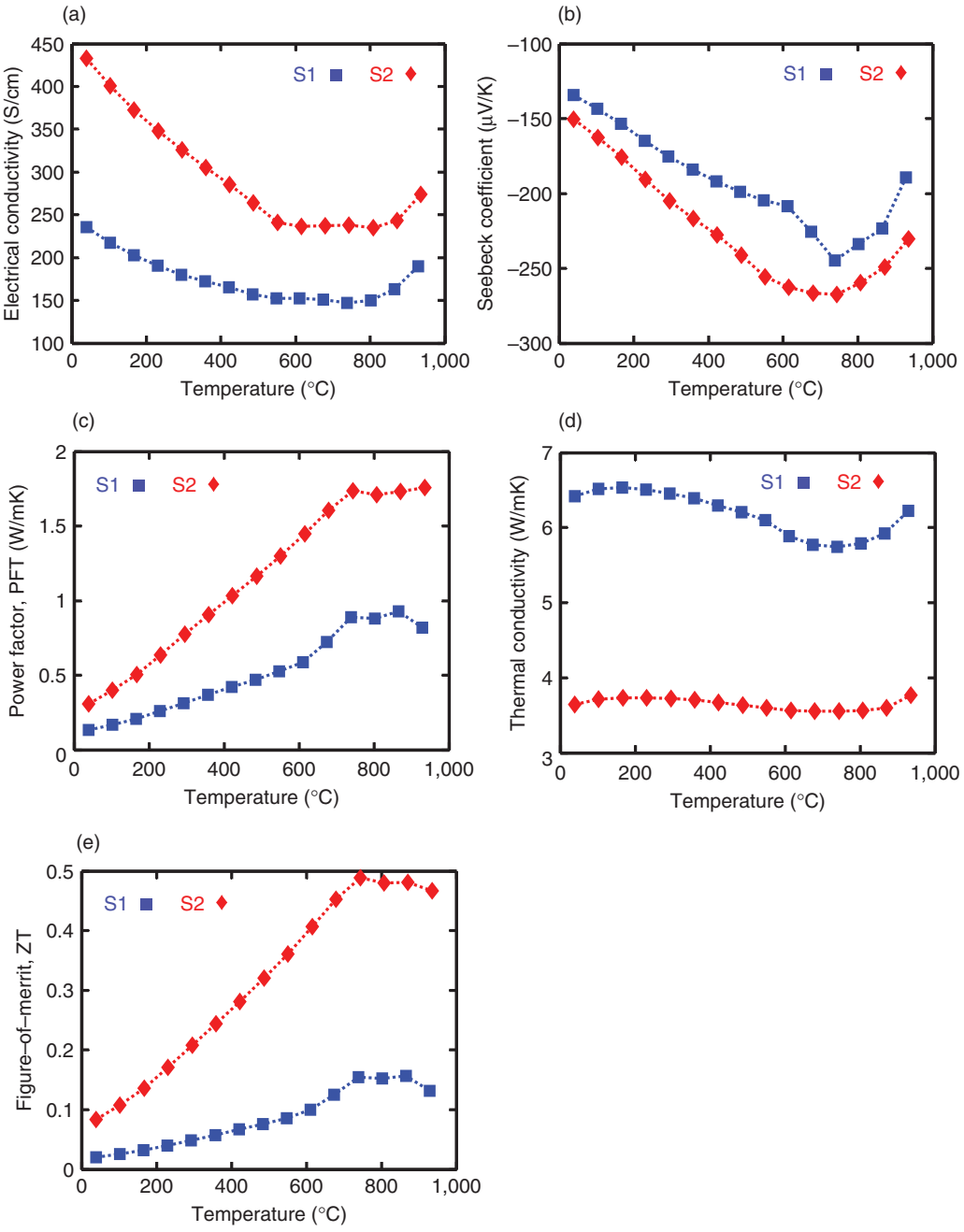

Figure 3: (a) Electrical conductivity, (b) Seebeck coefficient, (c) power factor times temperature, (d) thermal conductivity and (e) figure-of-merit of induction-melted composites.

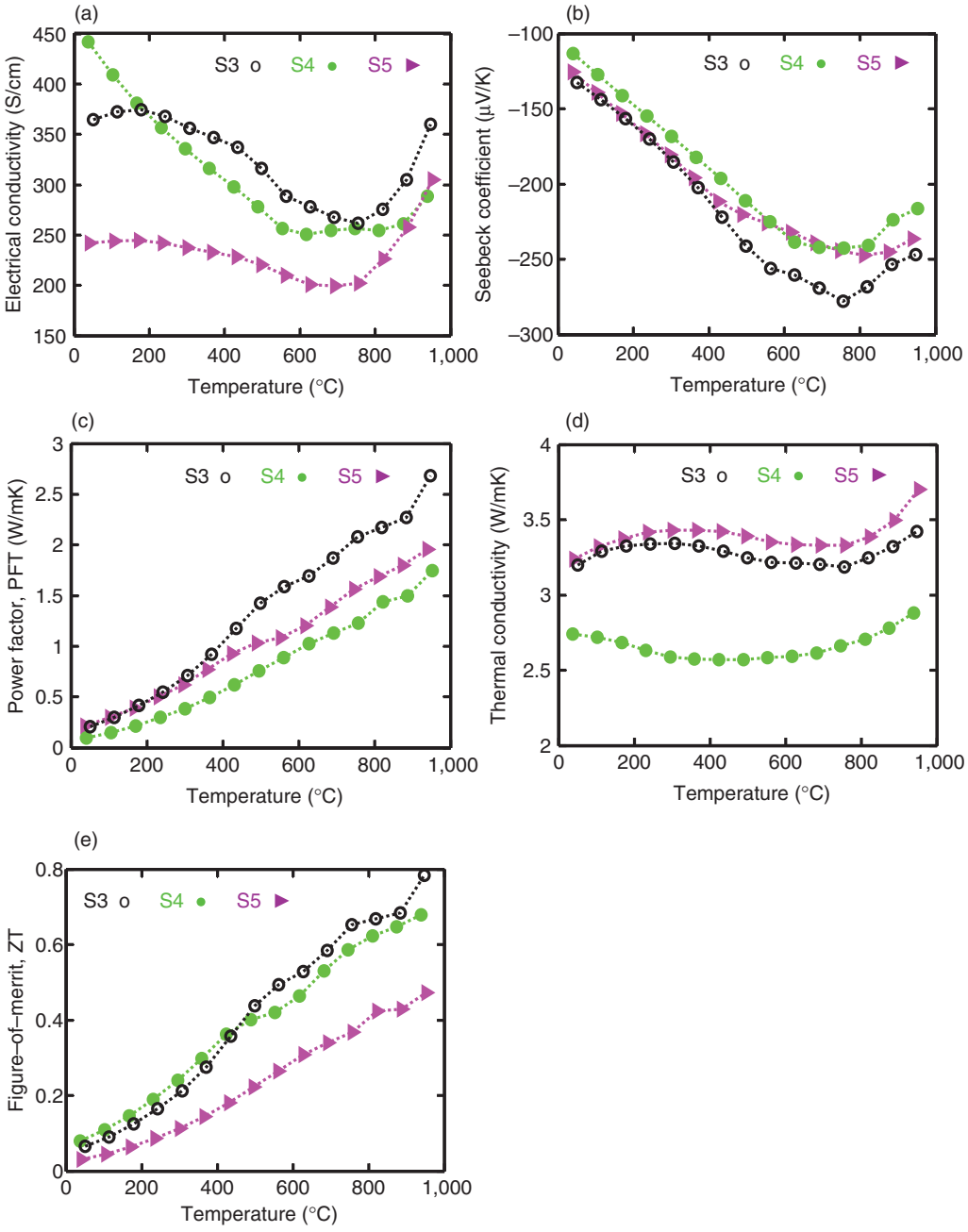

Figure 4: (a) Electrical conductivity, (b) Seebeck coefficient, (c) power factor times temperature, (d) thermal conductivity and (e) ZT of synthesized composites.

GeO_2 to Ge during the process of materials preparation. This result confirmed that addition of carbon during the synthesizing process of SiGe can reduce concentration of GeO_2 phase to achieve higher purity materials.

The reduction of electrical conductivity with temperature in all the samples is due to the decrease of carrier mobility originated from increasing acoustic phonon scattering with temperature. The electrical conductivity enhancement at temperature above 750°C was resulted from the increase of the intrinsic carrier concentration, which is also confirmed by the reduction of the absolute value of the Seebeck coefficient. At temperature above 200°C, S3 has the highest Seebeck coefficient and electrical conductivity. S4 has the lowest Seebeck coefficient and highest electrical conductivity at room temperature, which can be associated with a higher carrier concentration in this sample.

The composite structures show similar PFT at temperatures lower than 400°C. The highest PFT of 2.7 W/mK belongs to S3 which has fabricated via hot pressing procedure.

Due to the enhancement of phonon–phonon scattering with temperature, thermal conductivity of the TE materials generally decreases as the temperature is increased. At temperatures above 750°C, as a result of ambipolar thermal diffusion, thermal conductivity increased. However, nanostructured samples showed reduction in thermal conductivity due to the dominancy of the grain boundary scattering. This behaviour indicates that S3 and S5 have nanostructured morphology (Figure 4(d)).

Comparing the two crystalline samples of S2 versus S4, one can see that S2 has larger Seebeck coefficient; hence, lower carrier concentration. Since the electrical conductivities of S2 and S4 are in the same range, considering the lower carrier concentration in S2, the carrier mobility in S2 must be higher than in S4. Therefore, the addition of carbon has resulted in lower carrier mobility in S4. Comparing the thermal conductivities of S2 and S4, one can conclude that carbon has also reduced the thermal conductivity in sample S4. This may be associated with the higher concentration of Ge in S4 due to the reduction of GeO_2 by carbon. Overall, the reduction in thermal conductivity is more than the reduction in electrical conductivity resulting in higher ZT of sample S4 compared with that of sample S2.

Comparing the two nanostructured samples of S3 versus S5, one can see that both samples have similar Seebeck coefficient indicating that the samples have similar carrier concentration. Considering that S5 has smaller electrical conductivity, we can make similar conclusion as in the case of crystalline samples that the addition of carbon has reduced the carrier mobility. The thermal conductivity of the two samples is close with slightly smaller value for S3. This may be associated with the smaller density of sample S3. It may also be concluded that nanostructuring has dominated the effect of GeO_2 reduction in sample S5.

The dimensionless figure of merit of S3, S4 and S5 versus temperature is shown in Figure 4(e). It indicates that up to 400°C, S4 has the highest ZT; however, at temperature above 400°C, S3 has the highest ZT and its maximum is 0.78 at 950°C.

Conclusions

Nanocomposites of n-type SiGe–$FeSi_2$ were synthesized using low-purity germanium (70%Ge–30%GeO_2). The method proved a cost-effective way for fabrication of TE materials using inexpensive low-purity raw materials. The effect of carbon addition on reduction of oxides was studied. The synthesized materials were characterized by XRD and SEM-EDS and their TE properties were measured and compared. EDS analysis showed dispersed iron disilicide particles embedded in SiGe matrix. The composite structure showed a moderate figure-of-merit, ZT, of 0.8 which is comparable to that of high-purity crystalline SiGe. The high ZT despite the low purity of the raw materials was related to the composite structure and the reduction of the germanium oxide during the process.

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