

New Directions for Nanoscale Thermoelectric Materials Research

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Abstract

Many of the recent advances in enhancing the thermoelectric figure of merit are linked to nanoscale phenomena with both bulk samples containing nanoscale constituents and nanoscale materials exhibiting enhanced thermoelectric performance in their own right. Prior theoretical and experimental proof of principle studies on isolated quantum well and quantum wire samples have now evolved into studies on bulk samples containing nanostructured constituents. In this review, nanostructural composites are shown to exhibit nanostructures and properties that show promise for thermoelectric applications. A review of some of the results obtained to date are presented.

1 INTRODUCTION

The field of thermoelectrics advanced rapidly in the 1950's when the basic science of thermoelectric materials became well established, the important role of heavily doped semiconductors as good thermoelectric materials became accepted, and the thermoelectric material bismuth telluride was discovered and developed for commercialization. Thus in the 1950's the thermoelectrics industry was launched. By that time it was already established that the effectiveness of a thermoelectric material could in an approximate way be described in terms of the dimensionless thermoelectric figure of merit, $ZT = S^2\sigma T/\kappa$, where S , σ , T and κ are, respectively, the Seebeck coefficient, the electrical conductivity, the temperature and the thermal conductivity. Over the following 3 decades 1960–1990, only incremental gains were made in increasing ZT , with Bi_2Te_3 remaining the best commercial material today, with $ZT \approx 1$. During this 3 decade period, the thermoelectrics field received little attention from the worldwide scientific research community. Nevertheless, the thermoelectrics industry grew slowly but steadily, by finding niche applications for space missions, laboratory equipment, and medical applications, where cost and energy efficiency were not as important as energy availability, reliability, and predictability.

In the early 1990s, the US Department of Defense became interested in the potential of thermoelectrics for new types of applications, encouraging the research community to re-examine research opportunities for advancing thermoelectric materials to the point they could be used more competitively for cooling and power conversion applications. This attempt was successful in stimulating the research community to once again become active in this field and to find new research directions that would have an impact on future developments and would lead to thermoelectric materials with better performance. As a result of this stimulation, two different research approaches were taken for developing the next generation of new thermoelectric materials: one using new families of advanced bulk thermoelectric materials [1], and the other using low dimensional materials systems [2–4]. During the 1990's these two approaches developed independently and mostly along different directions. More recently the two approaches seem to be coming together again as the most successful new bulk thermoelectric materials are host materials containing nanoscale inclusions that are prepared by chemical approaches [5]. In this article, recent advances in the field of low dimensional thermoelectrics are summarized and connections are made to new families of bulk thermoelectric materials that incorporate nanoscale building blocks.

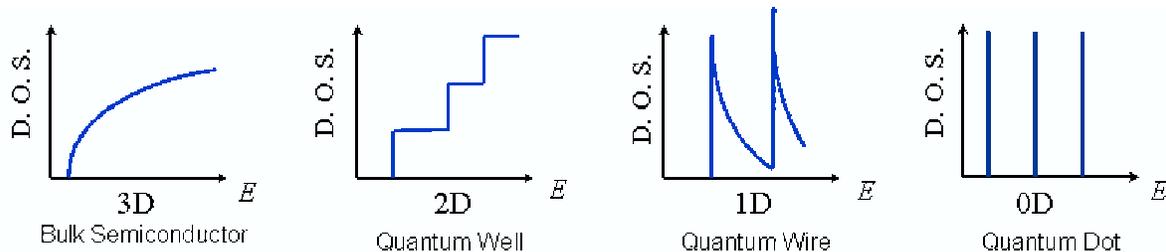


Figure 1: Electronic density of states for (a) a bulk 3D crystalline semiconductor, (b) a 2D quantum well, (c) a 1D nanowire or nanotube and (d) a 0D quantum dot. Materials systems with low dimensionality also exhibit physical phenomena, other than a high density of electronic states (DOS), that may be useful for enhancing thermoelectric performance (see text).

2 Background

There are several ideas behind using low dimensional concepts for enhancing thermoelectric performance. Since the quantities S , σ and κ for conventional 3D crystalline systems are inter-related, it is very difficult to control these variables independently so that ZT could be increased. But if the dimensionality of the material is decreased, the new variable of length scale becomes available, and as the system size approaches nanometer length scales, it is possible to cause dramatic differences in the density of electronic states (see Fig. 1), allowing new opportunities to vary S , σ and κ independently when the length scale gives rise to quantum confinement effects as the number of atoms in any direction becomes small (e.g., less than $\sim 10^2$). In addition, as the dimensionality is decreased from 3D crystalline solids to 2D (quantum wells) to 1D (quantum wires) and finally to 0D (quantum dots), new physical phenomena are introduced and these phenomena may also create new opportunities to vary S , σ and κ independently. Furthermore, the introduction of many interfaces, which scatter phonons more effectively than electrons, or serve to filter out the low energy electrons at the interfacial energy barriers allows the development of nanostructured materials with enhanced ZT , suitable for thermoelectric applications. The field of low dimensional thermoelectricity started with the introduction of these two concepts and the validity of these concepts was tested in model periodic quantum well [6] and quantum wire [7] systems, both from a theoretical standpoint and by experimental demonstration of the proof-of-principle of these concepts [8].

Up until now, the main gains in increasing ZT for low dimensional systems are based on strategies to reduce the thermal conductivity [9], whether by increasing the effect of rattlers in the cages of skutterudite-like materials [1], or by increasing the presence of interfaces which scatter phonons more effectively than electrons [9]. But to increase ZT sufficiently to lead to commercialization of low dimensional thermoelectric materials, it may not be enough to only decrease the thermal conductivity, but it may also be necessary to increase the power factor $S^2\sigma$ at the same time, and this point of view has been adopted in this report.

Strategies to increase the power factor involve searching for bulk materials with a large maximum in the electronic density of states (DOS), such as are found in transition metals or heavy Fermion systems, and then placing the Fermi level at this maximum in the DOS. Another strategy involves finding ways to increase the number of carrier pockets contributing to transport through imposing suitable perturbations to the electronic structure [9]. These two strategies are appropriate for both bulk materials and for nanostructured materials, though nanostructured materials can also make use of the special physics that applies to low dimensional systems.

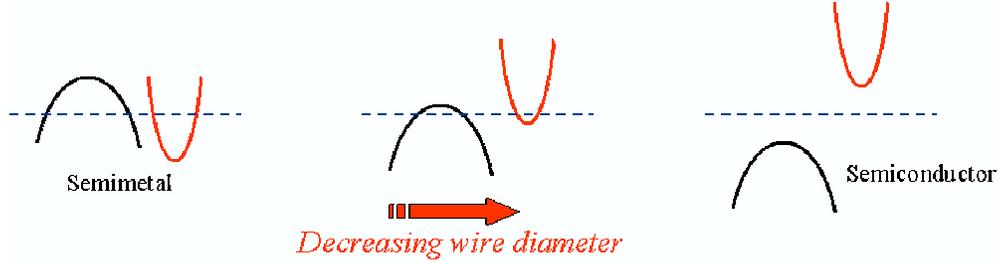


Figure 2: Schematic diagram of the bismuth nanowire semimetal–semiconductor electronic transition as the lowest conduction subband at the L -point moves up in energy, and the highest valence subband at the T point moves down in energy, as the nanowire diameter d_W decreases. (a) $d_W \gg 50$ nm, (b) $d_W \simeq 50$ nm, (c) $d_W \ll 50$ nm, the values being appropriate to pure bismuth with no antimony [2].

Another size-dependent property that can be exploited is the semimetal-semiconductor transition [10, 11]. For example, when the size of a semimetal nanowire decreases and there are relatively few quantum states normal to the axis of the nanowire, then the energy bands split into discrete levels that can be individually monitored. In this quantum regime, as the wire diameter decreases, the lowest conduction subband energy level moves up in energy while the highest valence subband energy level moves down in energy until the energy levels cross as the material makes a transition from a semimetal (with overlapping energy states for the lowest conduction band and the highest valence band) to a semiconductor with a band gap between the valence and conduction bands and the material has one dominant carrier type (see Fig. 2). Such a semimetal-semiconductor transition has been predicted [6], and observed experimentally [4], thus confirming theoretical predictions. Since alloying Bi with Sb changes the electronic structure of the bulk alloy [12], calculations show the dependence of the semimetal-semiconductor transition for a bismuth-antimony nanowire, on both wire diameter and antimony concentration [13]. These phenomena have also been confirmed experimentally [2] by changing both the wire diameter and Sb composition, thereby providing two variables for controlling and optimizing nanomaterials for enhanced thermoelectric performance.

Another previously known concept that was recently introduced to enhance the power factor is the concept of energy filtering of carriers by the introduction of appropriate barriers which restrict the energy of carriers entering into a material at an interface, so that the mean energy of the carriers substantially exceeds those at the Fermi level E_F , thereby enhancing the thermopower which depends of the excess energy ($E - E_F$) of carriers in the sample [14, 15]. Using the energy filtering approach, barriers are introduced in such a way that the reduction in the electrical conductivity σ is more than compensated by the increase in the Seebeck coefficient S through the energy filtering process, thereby resulting in an increase in power factor $S^2\sigma$. All of these concepts and strategies are currently being exploited in improving the performance of nanostructured materials for thermoelectric applications. Both fundamental and applications-oriented studies are now currently being pursued to advance the field.

3 Nanocomposite Thermoelectric Materials.

At the present time a number of research groups are developing nanocomposite materials with a potential for practical applications. The goals for designing materials for such applications are to introduce many interfaces that are specially chosen to: (1) reduce the thermal conductivity more than the electrical conductivity, and (2) to increase the Seebeck coefficient more (for example,

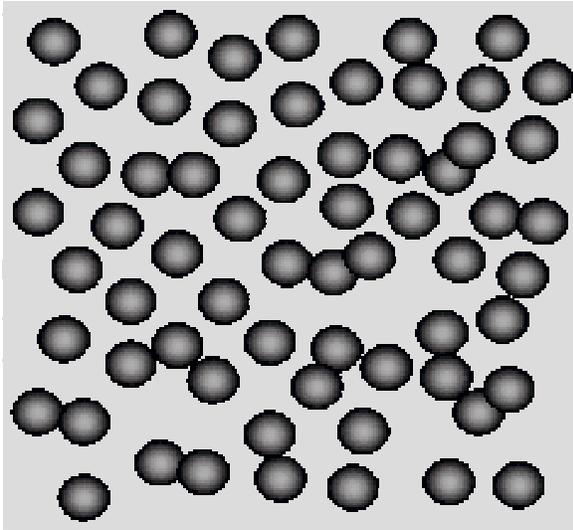


Figure 3: Conceptualization of a nanocomposite material with nanoparticles embedded in a host material, as for example Si nanoparticles (melting point 1687K) in a germanium host material (melting point 1211K).



Figure 4: Types of particle distributions that have been used in model calculations[16].

by carrier energy filtering or by quantum confinement) than decreasing the electrical conductivity, thereby yielding an increase in power factor, with both goals helping to increase ZT . Nanocomposites offer a promising technique for the preparation of bulk samples which are more easily handled from a properties measurement/materials characterization point of view, can be assembled into a variety of desired shapes for device applications, and can be scaled up directly for commercial applications.

In this report we show preliminary results to verify that a random assemblage of nanoparticles in a host material (see Fig.3 for the conceptualization of a nanocomposite) of bulk length scale (several mm or about half an inch in size) can yield enhanced thermoelectric performance relative to the alloy with the same composition of constituents, that the thermal conductivity can be reduced over a wide temperature range, and that the power factor can be increased at the same time by increasing S more than σ is decreased. Conceptual advances are presented for designing effective nanocomposite materials with enhanced thermoelectric performance along with explicit experimental results shown for Si-Ge nanocomposite materials under development for NASA-related radio-isotope power conversion applications. A variety of materials synthesis processes and approaches have been suggested by various research groups [17–19], involving different materials systems and processing methods, utilizing many common fundamental concepts, but differing in detail in their execution. The objective of this overview is to report on one such approach which has potential for practical applications.

Since nanoparticles of the constituent materials Si, Ge and $\text{Si}_{1-x}\text{Ge}_x$ alloys are not generally available in the sizes, compositions and doping levels desired, initial effort went into the synthesis of suitable nanoparticles for this project. The nanoparticles were prepared using wet chemistry by the Boston College group, ball milling by the JPL group and by inert gas condensation at the University of Illinois, Champaign/Urbana. Nanoparticles of silicon were fabricated along with Ge particles, both of nm and micron size. These particles were either hot pressed using a plasma

pressure compaction (P²C) method at Boston College or hot pressed (HP) in argon at 1333 K at JPL. These processes yielded dense, mechanically strong, bulk nanocomposites of near theoretical density. Disks a half inch in diameter were thus prepared, and other shapes were also prepared. Many different conditions of compaction and levels of n and p type doping were used to study the dependence of the nanocomposite materials on the processing conditions and materials parameters. Different sample shapes were used for different materials characterization measurements. The optimization of the processing conditions for maximizing the figure of merit (along with providing good in-service properties) are still at an early stage of development. Each set of samples, grown with deliberately chosen processing parameters, was then characterized by x-ray, SEM and TEM techniques to verify and characterize the nanoparticle integrity, meaning that the nanoparticle inclusions remained in the nanocomposite after all the processing steps were completed. The x-ray and TEM characterization results show that small nanoparticles of 5–10 nm size are retained after our nanoparticle compaction (P²C) procedures were carried out. These results confirm that nanoparticle inclusions remain after processing when the P²C steps were carried out in the 1050–1100°C range. Materials science studies of the effect of porosity on the transport properties show that the electrical conductivity of the nanocomposite changes by orders of magnitude when the sample density changes by only a few percent. These results highlight the great importance of achieving close to theoretical materials density for the nanocomposite, especially for the compaction of particles of nanosize relative to the compaction of particles of micron size. The importance of achieving a high density of the nanocomposite material for thermoelectric applications is likely to be a general result for nanocomposite thermoelectric materials for a large variety of choices for the constituent species, compaction methods and doping levels.

Model calculations are routinely used to guide the choice of processing parameters and approaches to be taken for doping and other process-sensitive considerations. Two approaches to model calculations have been taken: (1) Solution of the Boltzmann transport equation for a unit cell containing aligned nanoparticles with periodic boundary conditions imposed on the heat flow direction, with a fixed temperature difference across each unit cell in the model nanocomposite. For this model calculation the interface reflectivity and the relaxation time are used as input parameters. (2) A Monte Carlo method is also used for the modeling calculations, particularly for the case of random particle size, orientation and distribution [16]. Checks between the two calculational approaches have been made, and the types of particle distributions that have been considered in the model calculations are shown in Fig. 4. The model predictions for the thermal conductivity of nanocomposites have been published [20] and the modeling simulation of electron transport are presently under development using similar approaches.

The model calculations show that the thermal conductivity for nanocomposites can fall below bulk values for particle sizes in the 10 nm range for alloy compositions of Si_{1-x}Ge_x in the range $0.2 < x < 0.8$. It is interesting to point out the fundamental differences between bulk alloy samples and nanostructured samples of the same composition, as shown in Fig. 5 where the thermal conductivity along the wire direction vs volumetric fraction of Si is plotted for different nanostructures in comparison to the bulk alloy of the same composition. The calculations in Fig. 5 and in Fig. 6 show that superlattices of 10 nm diameter Si nanowires embedded in a Ge matrix and the corresponding nanowire composite of the same nanowire diameters and Si_{1-x}Ge_x stoichiometry had a lower thermal conductivity than their corresponding bulk alloys. For bulk alloys or nanostructured composites of large size (~ 500 nm), Fig. 5 shows that the lattice thermal conductivity increases with increasing Si concentration, reflecting the higher bulk thermal conductivity and higher sound velocity of Si relative to Ge [20]. However, for nanoparticle sizes 50 nm or less, the mean free path is limited by the particle size, so that the thermal conductivity κ becomes more sensitive to the velocity of sound and specific heat rather than to the mean free path for scattering. In this regime,

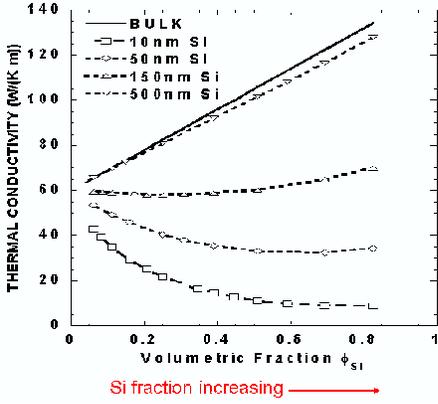


Figure 5: The thermal conductivity vs increasing Si fraction for Si nanowires in Ge host material to constitute the $\text{Si}_x\text{Ge}_{1-x}$ nanocomposite. For comparison, results for a bulk alloy with the same chemical composition as the nanocomposite material [20]. Transport is here along the wire direction.

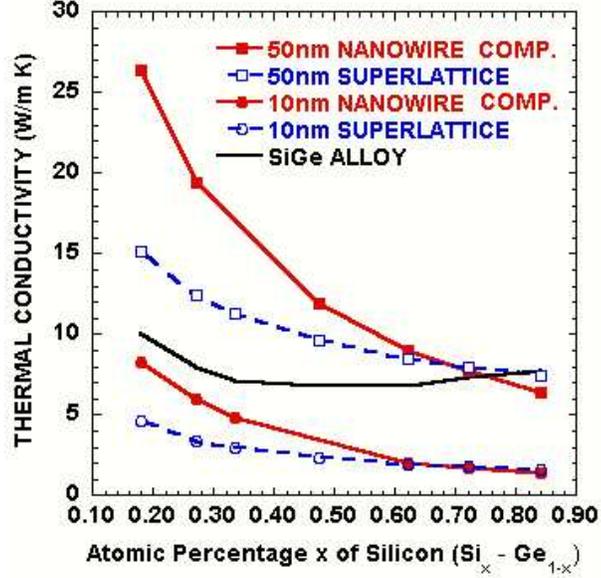


Figure 6: Thermal conductivity of a SiGe composite containing Si nanowires in a Ge host material. Transport is here normal to the wire direction [16, 21].

κ decreases with increasing volumetric fraction of Si (see Fig. 5), in contrast to the behavior of the 3D alloy samples with similar chemical composition. Monte Carlo simulations were made for many different mean particle sizes, size distributions and degrees of randomness and the results show that the thermal conductivity depends sensitively on the interface density (interface area per unit volume), following a universal curve as shown in Fig. 7. Provided that the interface area per unit volume is above 0.08 nm^{-1} , the thermal conductivity of the nanocomposite is lower than that of the bulk alloy. These results strongly indicate that ordered structures are not necessary to achieve a low thermal conductivity, thus providing a check on the use of self-assembled nanocomposite materials, such as in Figs. 3 and 4, for enhancing thermoelectric performance.

A comparison between preliminary experimental results and modeling calculations based on the $\text{Si}_{1-x}\text{Ge}_x$ nanocomposite materials (see Fig. 8) show a low value for both the experimental thermal conductivity over a wide temperature range on a *p*-type $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{0.01}$ sample that was ball milled for 96 hours and the calculated values based on the above modeling results for 10 nm Si nanoparticles embedded in a Ge host material. Preliminary results on a sample prepared by the P^2C method and measured only up to 500 K show a higher thermal conductivity than that for the ball milled sample, but nevertheless with an interesting reduction in thermal conductivity through the processing and preparation steps.

Experimental transport results given in Fig. 9 for a sample (SGMA04) similar to the one used in Fig. 8 (SGMA05) show that, due to strong interface scattering, nanocomposites generally have a higher resistivity and a higher Seebeck coefficient than bulk thermoelectric materials. The comparison sample shown in Fig. 9 is for an advanced bulk thermoelectric material that has been developed by NASA for radio isotope conversion applications, but not yet deployed. Since the increase in S for the nanostructured material prepared by ball milling (likely due to energy filtering effects) is substantially larger than the decrease in electrical conductivity σ , the power factor is increased in actual experimental nanocomposites samples (see Fig. 10), consistent with predictions from model calculations [22, 23]. Also shown in Fig. 10 is a comparison between the thermal

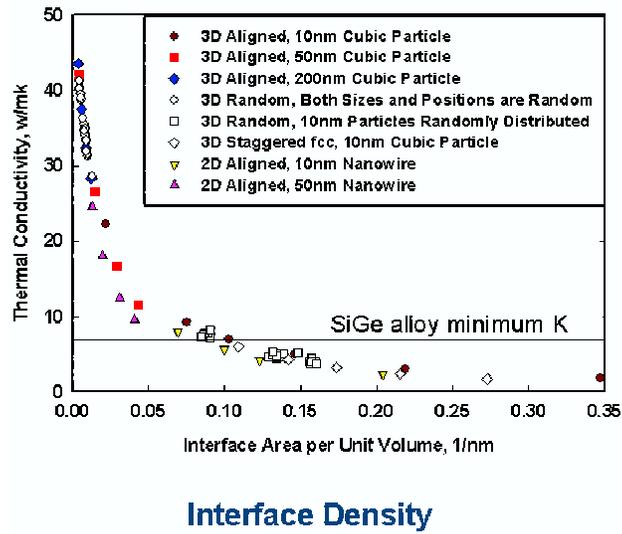


Figure 7: Universal curve for the thermal conductivity of Si nanoparticles and nanowires in a SiGe host material showing that nanoparticle and nanowire composites can give rise to very low thermal conductivity values, much lower than for an alloy of the same $\text{Si}_{1-x}\text{Ge}_x$ composition[16].

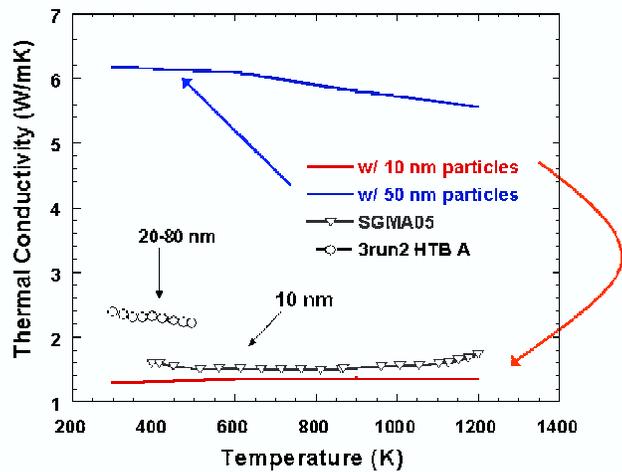


Figure 8: Comparison between model calculations for the temperature dependence of the thermal conductivity for nanocomposite of 10 and 50 nm Si nanoparticles in a germanium host and experimental measurements on two nanocomposite samples (see text).

Sample SGMA04 is a $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{1.5}$ p-type sample ball milled for 96 hours.

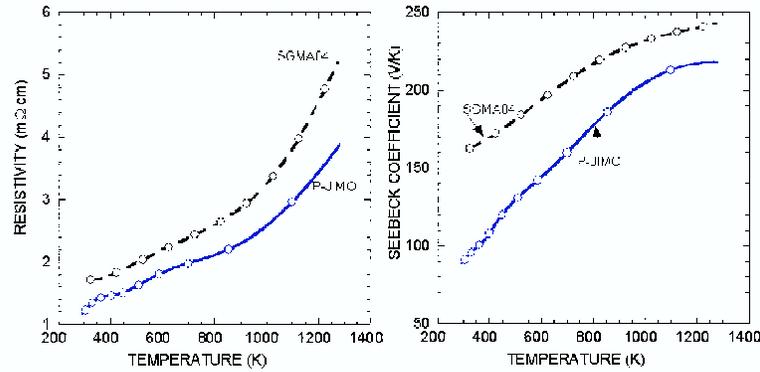


Figure 9: Comparison between the temperature dependent resistivity and Seebeck coefficient of a p -type $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{0.01}$ nanocomposite material obtained by ball milling for 96 hours in comparison to a bulk/advanced SiGe alloy material developed for thermoelectric applications showing that the power factor can be increased and the thermal conductivity reduced at the same time in the nanocomposite material [22]. Transport is along the nanowire direction.

conductivity for a nanocomposite and an advanced bulk thermoelectric material showing that it is possible for a nanocomposite material to increase its power factor and to decrease its thermal conductivity at the same time. Such behavior only occurs for nanostructure systems.

Figure 11 shows the temperature dependence of ZT for many types of materials, showing that the best thermoelectric performance is achieved in the $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{1.6}$ doped with 1.6% boron doping and ball milled for 96 hours while the sample with 2.0% boron doping, and similar otherwise, showed the second best thermoelectric performance. The ZT values for these nanocomposites are about 25% higher than the materials now employed by NASA (RTG SiGe) and about 10% higher than the advanced bulk thermoelectric materials under development at NASA (JIMO and SP-100). Although the measurements on the nanocomposites prepared by the P²C method have only been measured up to about 500K, they also seem promising. Since the nanocomposite materials are still at an early stage of development, especially related to optimizing their processing conditions and doping levels and doping species, further improvements in ZT are expected.

In summary, this work shows that randomly distributed nanostructures in SiGe nanocomposite materials can lead to a reduction in the thermal conductivity below that of the alloy at the same overall chemical stoichiometry. Even though the electrical resistivity may increase as a result of the introduction of nanoparticles, the increase in the Seebeck coefficient can be significantly larger, so that the power factor can increase as a result of nanostructuring. Furthermore, on the same sample and chemical stoichiometry both an increase in the power factor and a decrease in the thermal conductivity can occur at the same time relative to the alloy sample, thereby resulting in an increase in ZT from both processes. These findings identify promising research directions for nanocomposite materials produced either by materials processing strategies as in this work or by chemical means as shown for other materials systems.

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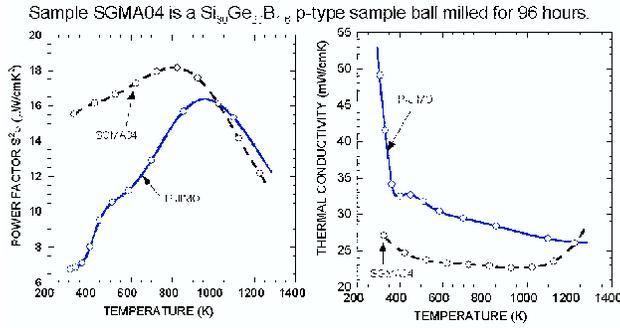


Figure 10: Comparison between the temperature dependent power factor and the thermal conductivity for a p -type $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{0.01}$ nanocomposite material produced by ball milling for 96 hours and a bulk SiGe alloy material developed by NASA for thermoelectric applications showing that the power factor can be increased and the thermal conductivity reduced at the same time in the nanocomposite material.

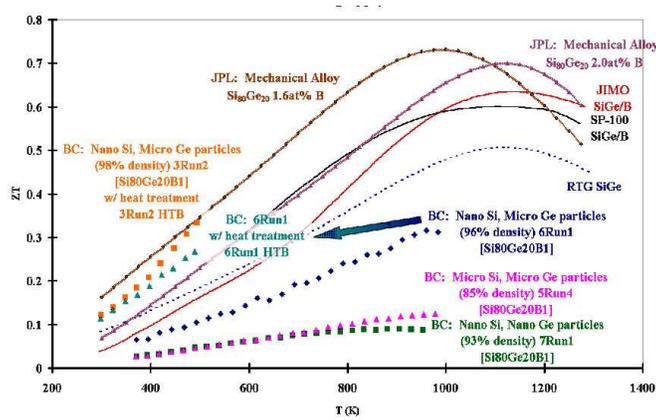


Figure 11: Plot of ZT versus temperature for the JPL prepared mechanical alloy of $\text{Si}_{0.80}\text{Ge}_{0.20}\text{B}_{1.6}$ as compared to the RTG SiGe bulk materials now used in NASA flights and to both a variety of JIMO test samples and a few of the nanocomposites obtained from the hot pressing (using the P^2C method) of nanoparticles prepared by the wet chemistry method.

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