ZT ~ 3.5: Fifteen Years of Progress and Things to Come

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Abstract

Fifteen years ago prospects for improving thermoelectric efficiency seemed slight and little evidence for ZT>1 was known [1]. Today, reports of ZT~2 are widely known, startup companies are forming based on this progress and enthusiasm for further advances is widespread. The enthusiasm is not without basis: at least one credible experimental study estimates ZT~3.5 [2] and at least one credible source estimates the bound may be as large as ZT~20 [3]. Thermoelectric applications have changed dramatically as well, with widespread commercial acceptance of products ranging from picnic baskets to automobile seats. Yet basic research advances have yet to impact applications, which thrive in spite of the basic R&D rather than because of it. This paper discusses some of the key developments, both technical and programmatic, which have driven recent progress and speculates on some developments which may be useful, possible and/or likely to drive progress forward.

Introduction

As an attempt to address the future this paper will first outline the status in 1992, summarize key developments since that time and then venture some speculations on future developments. No attempt is made to cover every possible aspect. Apologies in advance for omissions and errors.

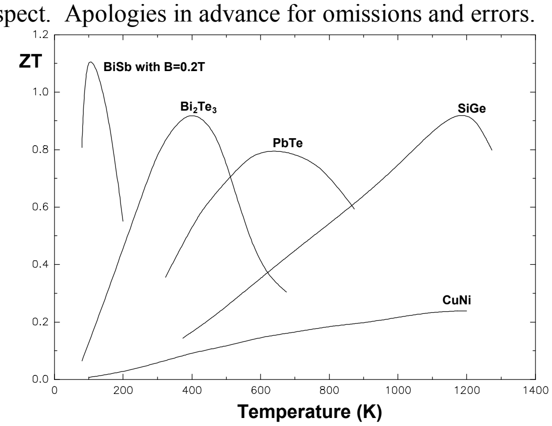

Figure 1: Thermoelectric figure of merit as a function of temperature for selected n-type alloys.

Thermoelectrics circa 1992

Three material families dominated thermoelectrics in 1992 (Figure 1) [1]: Bi_2Te_3-based materials for applications around room temperature, PbTe-based materials for use in an intermediate temperature range and SiGe for use at the highest temperatures, primarily in Radioisotope Thermoelectric Generators (RTGs) used to power spacecraft. Mature device technologies were available for each family of material and the general technology situation had changed little since the 1960s.

Counting BiSb, maximum ZT values near unity were known from about 100 K to about 1300 K. To be sure, occasional reports of higher ZT values existed but by and large these were not taken seriously and there was a real question of whether ZT~1 might represent a genuine physical barrier. Moreover, there was no serious discussion of how to achieve significant improvements.

A proposal to achieve, say, ZT~3 might easily be dismissed out of hand. No funding agency in the US was supporting basic R&D in thermoelectrics and, consequently, no US university groups were focused on the subject. To be sure, NASA and the space power portions of the US Department of Energy supported some work, but generally with the goal of incremental improvement in existing materials. Shortly, the NASA/USDoE support for space power thermoelectric R&D halted essentially completely for nearly a decade. Manuscripts submitted to journals were routinely rejected, often due to lack of qualified reviewers or simple lack of interest.

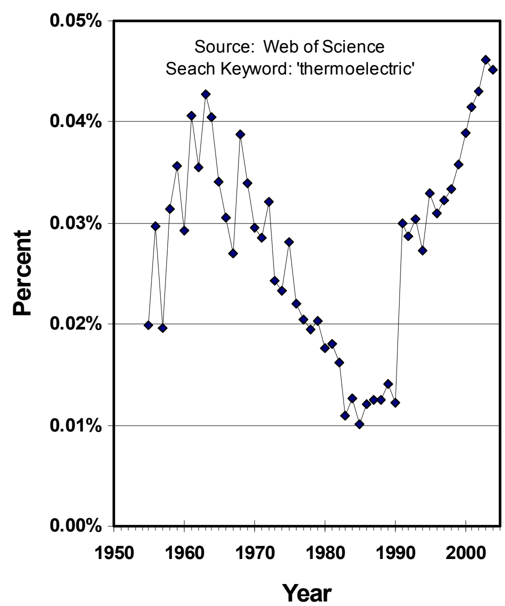

Figure 2: Publications in the Web of Science database with the keyword 'thermoelectric' as a percentage of all publications in the database from 1955-2003 [4].

The situation in Europe was little better. A few university research groups, notably Rowe in Wales, the Scherrers in France and a few others, managed to eke out original research with limited funds. Together with groups from Japan and the former Soviet Union a small R&D community, newly organized as the *International*

Thermoelectric Society, gathered every two years for what had become the annual *International Conference on Thermoelectrics* (ICT) [4].

By 1992, the mood of the thermoelectric R&D community was subdued. Research publications had been in decline for years (Figure 2). Participants at ICT92 in Arlington, Texas USA noted the deteriorating prospects for funding and progress. But unknown to most of the then existing thermoelectric R&D community, change was coming, to be discussed below.

The business of thermoelectrics fared slightly better, perhaps, than the R&D community. The three largest thermoelectric manufacturers in the US, Marlow Industries, Melcor and Tellurex, each had carved out a business niche. In Japan, Komatsu and Ferrotec deserve note and in Europe Supercool must be mentioned. Organizations from the former Soviet Union were hardly known in the west and as yet Chinese manufacturers had not emerged.

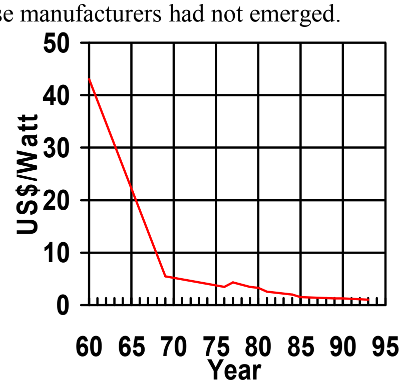

Figure 3: Price per Watt of cooling in 1993 US$ [5]

A key development in the early 1990s was the introduction of the first significant consumer product based on thermoelectrics: a thermoelectric picnic basket cooler introduced by Igloo. The idea was not entirely new but Win Eastman, working with Igloo, introduced this attractive and affordable consumer product providing a major boost to the thermoelectric cooler business. Thermoelectric 'industry' was by no means large, but it was stable and growing. Key to the advancement of industry, of course, was a steady reduction in the cost of manufacturing thermometric coolers as estimated by Buist and shown in Figure 3 [5].

Dr. Buser (Army Night Vision Lab) and Seeds of Change

The thermoelectric community, particularly the R&D community, was about to change and the source of change was not obvious. One man, Dr. Rudy Buser, more than any other single person is probably responsible for the main advances in thermoelectric R&D over the past 15 years. Perhaps the time was ripe for new ideas and certainly many people contributed but Dr. Buser got things going.

Dr. Buser was Director of the US Army Night Vision Laboratory and regularly funded Marlow Industries for thermoelectric device R&D. By 1990 Dr. Buser expressed his frustration to Raymond Marlow, the CEO. Year after year, Marlow would engineer an additional 'stage' to their cooler, or improve the radiation shields.

So Dr. Buser asked Mr. Marlow how to avoid the incremental progress and instead have just one big project to reach the sorts of cryogenic temperatures he really needed. Mr. Marlow, being an honest and capable engineer, said it was impossible. Dr. Buser simply asked "Why"?

When told the limitation was a fundamental property of the thermoelectric materials, Dr. Buser persisted: "Why"? And again, Marlow answered the only way a capable and honest engineer can: he didn't know. But he would find out.

This line of questioning led Ray Marlow to commit significant resources, particularly for a company the size of Marlow Industries, to examine how thermoelectrics could be improved. And not just a little, but a lot. Marlow hired consultants (including Dr. Julian Goldsmid and myself), formulated plans and traveled the US to meet with anyone who would listen to crazy ideas about how to improve thermoelectrics. Such was his confidence that on Feb. 19, 1993 Marlow signed a placemat with his hand-sketch of dramatic improvements in ZT (Figure 4).

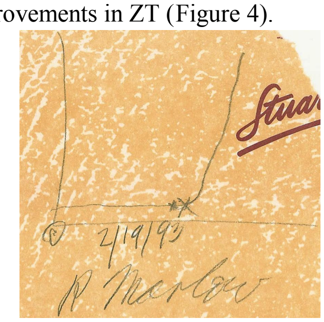

Figure 4: Raymond Marlow of Marlow Industries confidently predicted high ZT values in 1993.

Dr. Buser did at least two other important things in the early 1990s. He commented in an Army newsletter that perhaps after all these years there must be some new ideas worth trying in thermoelectrics. Ted Harman at MIT's Lincoln Labs, who had not worked on thermoelectrics in years, saw Dr. Buser's comments and began thinking about using MBE (molecular beam epitaxy) to make thermoelectrics in ways not possible in the 1960s.

The other thing Dr. Buser did was to issue a Call for Proposals and hold a workshop on thermoelectrics. About 70 people, including 33 from the Night Vision Lab which sponsored and organized the event, attended the 1992 1st National Thermogenic Cooler Conference [6].

There never was another Thermogenic Conference, but many ideas were introduced for the first (or nearly first) time at this conference. Among the notable presentations were:

- Hicks & Dresselhaus, Quantum Wells [7]
- Venkatasubramanian, Superlattices[8]
- Harman, PbTeSe/BiSb MBE Superlattices [9]
- Slack, Skutterudites [10]

I discussed the ZT~1 'limit' (essentially identical to [1]) and contributed to Dr. Stuart Horn's overview [11], which outlined the national thermoelectric materials R&D program envisioned by the US Army Night Vision Lab. Many of the ideas presented at that Conference, and embodied in the proposals eventually submitted to the Army for funding, had not been presented publicly before and represented quite new directions for thermoelectric R&D.

Funding for this particular project never did materialize, but the effort demonstrated to other US funding agencies, notably the Defense Advanced Research Projects Agency (DARPA) and the Office of Naval Research (ONR), that Dr. Buser's insight was correct: after several decades of decline there were by 1992 many new ideas in thermoelectrics.

One other connection, albeit indirect, to Dr. Buser is worth mentioning. By now the two 1993 thermoelectric papers by Hicks and Dresselhaus on quantum wells and wires are well known [12, 13], but the essential results had already been presented at the 1992 Thermogenics Conference [7]. Dr. Mildred Dresselhaus had a longstanding interest in low-dimensional physics but had become aware of the possibilities for thermoelectrics only after a dinner in Belgium with Dr. Jean-Paul Issi and Dr. John Stockholm in the early 1990s. John Stockholm often tried to stir up interest in thermoelectrics, but may have been indirectly influenced by Dr. Buser via Ray Marlow, who had started talking about improving materials already in 1990.

Dr. Buser played the role of a catalyst, reigniting interest in thermoelectrics. For new ideas to actually emerge and genuine technical progress to be made, the scientific 'soil' must be already fertile. Perhaps eventually something else would set it off, but still one must acknowledge the timely contributions and stimuli which came from Dr. Buser.

Thermoelectrics circa 2007

This section will briefly summarize selected key developments since 1992. For more detailed reviews of thermoelectric materials R&D the reader is referred to several excellent books [14-16] and recent review articles [17, 18]. Thermoelectric materials R&D, thermoelectric power generation and thermoelectric products and business developments will be discussed in turn.

Thermoelectrics Materials R&D

Based in large part on the ideas presented at the 1992 Thermogenics Conference, DARPA and ONR began funding in the US a wide variety of basic thermoelectric R&D efforts. Just as importantly, they managed to sustain funding for a decade or more. Figure 2 illustrates the explosion of scientific publications on thermoelectrics since 1992, exceeding even the level of activity in the late 1950s.

The variety of materials investigated in this period is impressive indeed: skutterudites, Clathrates, Zintls, Zn-Sb, heavy fermions, opals (and inverse opals), organics, aerogels, Half Heuslers, cobalt oxides, quasicrystals, pentatellurides and on and on. Only a very few of these studies, however, approach the goals set by the funding agencies. The fruits of this labor, as measured by ZT, are shown in Figure 5.

The Hicks and Dresselhaus quantum well superlattice idea [12] has been described as an attempt to increase the Seebeck coefficient both due to quantum confinement effects (principally an enhanced density of states) and due to selection (by design) of the most favorable effective mass. The original calculation omitted a number of effects, including tunneling between quantum wells, reduction of degeneracy, increased electron-phonon scattering and thermal conduction of the barrier material each of which tend to reduce the theoretical improvement in ZT [19-21].

Essentially the same quantum well idea described by Hicks-Dresselhaus [12] was examined independently, and somewhat more completely, by Whitlow [19] of Daikin Industries in Japan. However, the possible enhancement in ZT predicted by Whitlow was more modest and Daikin never pursued the matter. In one of those curious coincidences that sometime happen in science, Whitlow was entirely unaware of the Hicks-Dresselhaus work and an internal, unpublished Daikin report indicating his main results was known to Daikin before the Hicks-Dresselhaus paper appeared [22].

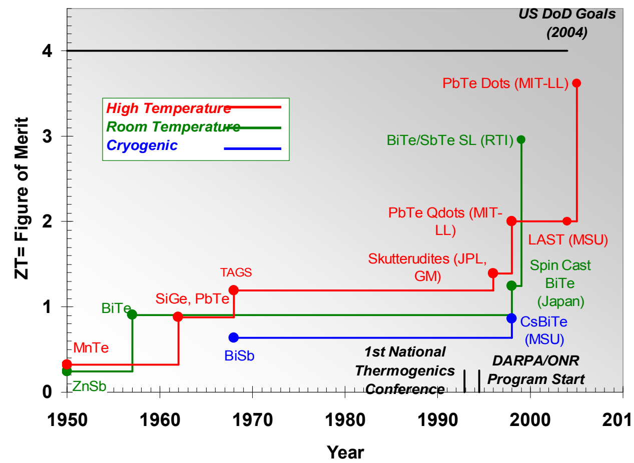

Figure 5: History of the thermoelectric figure of merit, selected results, inspired by Dubois [23].

Excellent experimental work by Harman on PbTe/Te superlattices did indeed confirm substantial enhancement of the Seebeck coefficient [24], but power factor (σS^2) and ZT enhancement [25] proved much more modest. This approach has, for the most part, been abandoned.

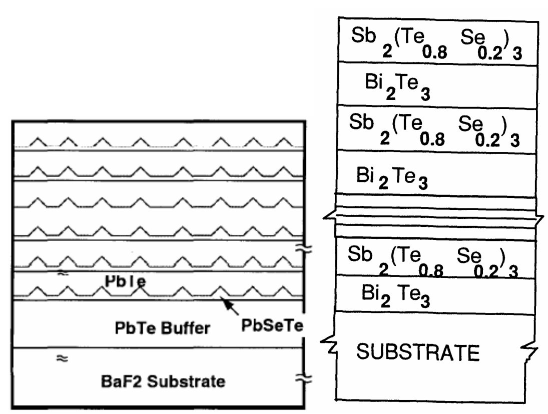

Figure 6: Harman (left, currents flow parallel to substrate) [26] and Venkatasubramanian (right, currents flow perpendicular to substrate) [8] superlattice concepts to improve ZT.

As a direct consequence of these studies, however, Harman (MIT Lincoln Labs) has demonstrated impressive ZT gains in PbTeSe quantum-dot superlattices [27], reaching ZT values of 3.5-3.6 [2]. This last report is the highest credible report of ZT in the literature. The high ZT appears to be primarily a result of phonon scattering associated with the quantum dots, but quantum confinement effects may also play a role in enhancing the Seebeck coefficient and compensating for some loss of carrier mobility.

Venkatasubramanian (Research Triangle Institute, RTI, and more recently Nextreme) [28] has reported ZT=2.4 for a Bi_2Te_3-Sb_2Te_3 superlattice. Figure 6 shows the original 1992 concept, although Se was not used in the high ZT study. Here, the idea from the beginning was to enhance ZT due to phonon scattering at the superlattice boundaries. Remarkably the electrical mobility appears not to be reduced by these same boundaries.

A final example would be the LAST (Lead-Antimony-Silver-Tellurium) materials with ZT~2 [29]. These materials are based on PbTe with addition of a small amount of $AgSbTe_2$ which alters both the crystal structure and the microstructure. The result is a significantly lower thermal conductivity and higher ZT compared to PbTe.

These three examples (PbTe quantum dots, Bi_2Te_3-Sb_2Te_3 superlattices and the LAST materials) share several features in common: none has been yet reproduced in other laboratories and predictive models are not yet available.

Thermoelectric Power Generation

Unlike cooling applications, where growing markets have helped drive down costs, thermoelectric power generation applications markets remain quite small. Costs, therefore, remain relatively high for thermoelectric power generation devices. Four areas of thermoelectric power generation deserve some brief mention here: remote power, space power, waste heat and solar thermoelectrics. Companies such as Global Thermoelectric and FerroTec (which acquired Teledyne's remote power product line in 2003) continue to provide fossil-fuel fired thermoelectric power generators for locations where grid power is unavailable, or simply unreliable. The remote power business appears stable, but little promise for major growth.

Radioisotope Thermoelectric Generators (RTGs) have been a genuinely enabling technology for deep space exploration and have been recently reviewed by Bennett [30]. SiGe RTGs have accumulated over a billion unicouple device-hours in space without a single failure and the latest RTG powers the 2006 launch of the New Horizons mission to the Pluto-Charon system. Significantly, the US has lost the capability to build new SiGe unicouples, although 2 or 3 SiGe RTGs may be built from existing unicouple stock.

NEDO in Japan has been investing in thermoelectric waste heat technology at least since 1997. Driven by environmental concerns as well as the desire to utilize industrial sources of waste heat, the current 5 year project is investing US$21M to demonstrate practical systems with 15% efficiency [31]. Commercial applications are expected to reduce CO_2 emissions 141 million tons by 2030.

Lewis and Crabtree have recently discussed thermoelectrics in combination with concentrated solar heat as a possible clean source of electricity [32]. More importantly, in 2006 the US DoE initiated a funding opportunity (DE-FG02-06ER06-15) for thermoelectrics as part of a broader effort to utilize solar power.

Thermoelectric Products and Business Developments

Since 1992 the business environment has changed in several ways. China, and to a lesser extent the Former Soviet Union, has emerged as a major supplier of low cost cooling modules. Chinese manufacturers include Fuxin Electronics, Hui Mao, HiCool, Hangzhou Jianhua Semiconductor Cooler, Hebei IT Shanghai, Taicang TE Cooler, and Taihuaxing Trading/Thermonamic Electronics. Fuxin alone reported annual sales over US$50M. Former Soviet Union manufacturers include our host for this conference, Thermion, as well as Altec (associated with the Institute of Thermoelectricity), Kryotherm, Nord, Osterm, RIF Corp., RMT, Thermix, and ADV-Engineering. Certainly this list is not exhaustive, and further information on each company can be found simply using Google.

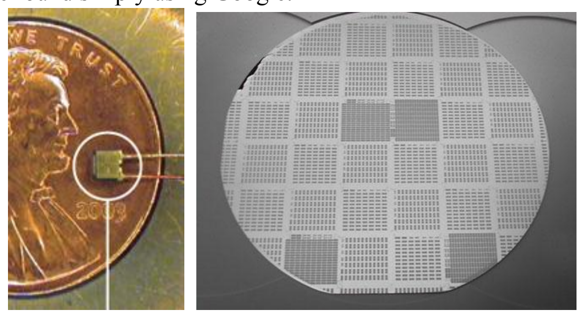

Figure 7: Nextreme (left) thin-film TE cooler and MicroPelt (right) Bi_2Te_3 thin-film TE wafer.

Table 1: Investments In Thermoelectric Companies

Year	Company	Investor	Investment
2006	MicroPelt	Fraunhofer/Infinion	N/A
2005	Nord	FerroTec	N/A
2005	Melcor	Laird	$20M
2005	Nextreme	RTI/Startup	$8M
2004	Marlow	II-VI Inc.	$31M
2003	Nanocoolers	Startup	$8.5M
2003	Teledyne (Telan)	FerroTec	N/A
1992	Melcor	Fedders	$14.9M
*N/A = Not Available			

Table 1 illustrates investor interest in thermoelectric companies both as acquisition targets and as startup vehicles. Startups MicroPelt (Germany) and Nextreme (US) are each pursuing thin-film technologies to produce next generation thermoelectric devices (Figure 7). MicroPelt has developed a sputtering method based on Bi_2Te_3 materials to produce wafers and TE coolers compatible with modern semiconductor industry mass-production methods. Nextreme technology is based on the Bi_2Te_3-Sb_2Te_3 superlattice developed by Venkatasubrmanian [28]. Both companies offer prototypes of their products and appear to be nearing general production. Neither company's devices are yet more efficient than conventional TE devices, but provide other benefits including size, speed and heat pumping capacity (Watts/cm^2).

Possibly the most significant commercial development in recent years has been the success of Amerigon (US) in placing thermoelectric coolers/heaters in automobile passenger seats. Amerigon shipped 718,000 units in 2006 has placed over 2 million of their Climate Control Seat^{TM} options (Figure 8) in 20 vehicle models sold worldwide. Just as importantly, Amerigon revenues have grown substantially (Figure 9) and the company is projecting the world market to grow to US$1 billion by 2010.

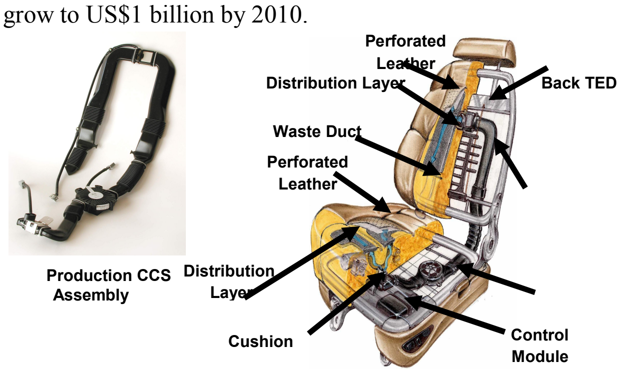

Figure 8: Amerigon Climate Control Seat^{TM} for cooling and heating of car seats.

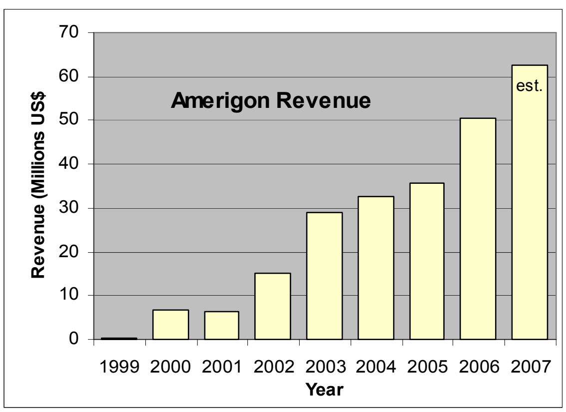

Figure 9: Amerigon revenue 1999-2007

Things To Come

Thermoelectric R&D has made admirable progress in these past 15 years, with laboratory ZT values increasing several fold. The business of thermoelectrics has also grown significantly. Startup companies Nextreme and MicroPelt are each preparing to introduce next-generation thermoelectric technology with unique characteristics. Amerigon, utilizing creative engineering, is placing large numbers of devices in cars. Indeed, much has happened in 15 years. Yet none of the thermoelectric devices available today, even at those at the prototype stage, are actually more efficient than the TE devices of 15 years past. So far as actual applications are concerned, it is almost as if the last 15 years of basic thermoelectric materials R&D did not happen.

A clear challenge for the thermoelectric materials R&D community will be to understand exactly why the reported high ZT values actually occur and to capture the essential underlying physics in materials that can be manufactured cost effectively. As part of this effort it would be desirable to see: independent verification(s) of high ZT; attempts to combine effects (such as combining quantum confinement effects with phonon scattering effects); extend the temperature range of high ZT; establish both n- and p-type materials with high ZT; and most importantly, develop predictive models. There is much basic research yet to do.

With companies like MicroPelt and Nextreme pursuing thin-film thermoelectric devices, particularly coupled with the growing markets being established by Amerigon, it seems reasonable to expect further size and cost reductions through mass production and utilization of modern semiconductor manufacturing technologies. Lower costs, and also the unique characteristics of thin-film thermoelectrics, will inevitably enable new products.

Based on the recent acquisition and merger activities we may see consolidation among the TE cooler module manufactures, particularly among the large number of companies in China, Russia and Ukraine. As economies of scale become increasingly important, small-scale suppliers may need to partner with larger organizations to compete.

The greatest challenge facing thermoelectricity in coming years will no doubt be associated with climate change. Climate change is the 'elephant in the living room'. Everyone likes elephants, but no one wants one in their living room. Energy-related R&D budgets can be expected to become increasingly dominated by climate change considerations. Already NEDO in Japan [31] and the Department of Energy in the US [32] have thermoelectric programs driven, at least in part, by climate change.

At ICT2007 the *International Thermoelectric Society* adopted as their primary goal:

> **"To promote an understanding of the role thermoelectric technology may play in environmental impact and mitigating global climate change."**

If thermoelectricity can contribute to the climate change problem, it is imperative that we do so. Alternatively, if it can be determined that thermoelectricity has little to offer then we must not advocate thermoelectric technology. Instead, resources must re-directed to technologies that may help. The stakes are simply too high for anything less.

Acknowledgments

The author wishes to thank V. Semenyuk and Thermion Company for their generous travel support.

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