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ABSTRACT BOOK

ORAL PRESENTATIONS

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IREC 10 Shaping Energy for a Sustainable Future

ECT'22

Session I - Auditorium

MATERIALS & PROCESSING I

ID: 04686
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Novel family of n-type thermoelectric oxides based on La0.15-xEuxSr0.775TiO3-?

Stephanie Mudd[1] , Rebecca Boston[1]

1) The University of Sheffield

We present a novel composition $La_{0.15-x}Eu_xSr_{0.775}TiO_{3-?}$ made using ionic liquid synthesis. Complex stoichiometry enabled by this synthesis method allowed the continuous reduction in the thermal conductivity with increasing Eu content of a previously shown, high power factor material. High power factor and low thermal conductivity are required to enhance the thermoelectric figure of merit, ZT, which are often mutually exclusive characteristics in intrinsic materials. High thermal conductivity is an issue in most oxides due to strong covalent bonds allowing the transport of heat energy via phonons, in an otherwise low cost, thermally stable and abundant material. What's more ionic liquid synthesis allows lower calcination temperatures (800 ºC compared to ~1200 ºC used in conventional solid state synthesis methods) which reduces the energy consumption of manufacture and presents the opportunity to preserve nano-structuring in the final material, which can also scatter phonons and reduce thermal conductivity further.

ID: 04998
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Reduction of Thermal Conductivity of Mg2Sn Single Crystal via Lattice-Defect Engineering through Chemical-Pressure Control

Kei Hayashi[1] , Wataru Saito[1] , Zhicheng Huang[1] , Kazuya Sugimoto[2] , Kenji Ohoyama[2] , Naohisa Happo[3] , Masahide Harada[4] , Kenichi Oikawa[4] , Yasuhiro Inamura[4] , Kouichi Hayashi[5] , Takamichi Miyazaki[6] , Yuzuru Miyazaki[1]

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Mg_2Sn is a potential thermoelectric (TE) material that can be used at mid-temperature range. Recently, we prepared Mg_2Sn single crystals (SCs) containing Mg vacancy (V_{Mg}) as a point defect, which exhibited lower *?* compared with an Mg_2Sn polycrystal (PC) [1]. Furthermore, we enhanced the TE performance of the Mg_2Sn SC with V_{Mg} by Sb-doping [2]. The maximum *zT* of the Sb-doped Mg_2Sn SC reached 0.72 at 650 K, which is higher than that of single-phase Sb-doped Mg_2Sn PCs. Although relatively high *zT* was obtained, the Sb-doping decreased the V_{Mg} fraction, i.e., decreased the effect of reducing *?*. In this study, we have performed the B-doping to the Mg_2Sn SC, expecting the increase of the V_{Mg} fraction accompanied by the increase of chemical pressure. The crystal structure of the B-doped Mg_2Sn SCs is investigated by using white neutron holography and single-crystal X-ray diffraction (SC-XRD). From the white neutron holography, the B atom is found to substitute for the Mg site. In addition, the presence of V_{Mg} around the B atom is clearly observed. These results indicate that the B-doping increases the V_{Mg} fraction, which is confirmed by the SC-XRD. The transmission electron microscopy observations revealed the presence of V_{Mg} regions as well as edge dislocations, similar with the cases of non-doped and Sb-doped Mg_2Sn SCs [1,2]. The presence of V_{Mg} and dislocations significantly reduces $?_{lat}$. The lowest $?_{lat}$ of 0.65 W/Km at 650 K is obtained for the B-doped Mg_2Sn SC with the B-doping concentration x = 0.50%. This study demonstrates that the V_{Mg} fraction can be controlled by the chemical pressure, and can aid in realizing a high TE performance for the Mg_2Sn SC.

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[2] W. Saito, K. Hayashi, et al., ACS Appl. Mater. Interfaces 12, 57888 (2020).

ID: 05044
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Tuning of thermoelectric performance of Al doped ZnO using in-situ O2 plasma treatment with integration into hot pipe networks

Vikesh Sethi[1] , Daniel Newbrook [1] , Ruomeng Huang [1] , Kees De Groot [1]

1) School of Electronics and Computer Science, University of Southampton

Increasing interest in the internet of things (IoT) and integrated wireless sensor networks (WSNs), has led to an unprecedented increase in IoT nodes, with over 10 billion active connections and predictions claiming over 25 billion active nodes by 2025. The intermittent operation and low energy consumption of IoT nodes, allows for energy harvesting technologies to offer a solution to the ever-growing energy consumption of IoT technologies. A prime candidate for powering WSNs due to the ability of solid-state conversion of heat to electricity are thermoelectric generators.

At present most high performing thermoelectric materials are tellurium based. Concerns over the natural abundance of tellurium along with material stability at high temperatures, makes chalcogenide-based materials ineffective for high temperature applications. A promising material due to its stability over a wide temperature range, high electrical conductivity and Seebeck (S), is Al doped ZnO (AZO). With the material benefiting further from the high natural abundance of constituent elements allowing for resource conscience energy harvesting.

In this work, we will present the deposition of highly uniform and conformal AZO thin films *via* atomic layer deposition (ALD). With this deposition method allowing for great control of composition to achieve an Al doping concentration of 4%, as well as excellent control of film thickness giving nm control. We present characterisation of the deposited films with a range of techniques including SEM, XPS and XRD. We go on further to report a full thermoelectric characterisation include Hall, Seebeck and thermal conductivity measurements. With the latter being conducted using our custom made ?-3? set up. Moreover, we present the ability to tune thermoelectric performance via an *in-situ* O_2 plasma treatment [2]. Additionally, we fabricate a lateral thermoelectric generator designed for integration into hot pipes systems tested experimentally and supported by simulations to reveal potential power densities for integrated systems.

ID: 05085
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

High Performance (ZT>1) Strontium Titanate based Oxide Nanocomposites for High Temperature Thermoelectric Power Generation

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Oxide thermoelectrics are attractive because of their environmentally benign nature, low fabrication cost and better stability at high temperature compared to other metal-based thermoelectric materials. However, the challenges in oxide thermoelectrics are manifold as figure of merit, ZT values of oxides suffer from lower electrical conductivity as well as relatively higher thermal conductivity compared to state-of-the-art materials like chalcogenides. Especially, achieving ZT>1 remain elusive for bulk oxide thermoelectrics. Recently, we have put forward a strategy of boosting the electron transport by manipulating semiconductor to metal transition temperature in donor doped $SrTiO_3$by synthesizing nanocomposites with graphene oxide (GO), graphite (G) and MXene. We could enhance electrical conductivity remarkably by making composites of La, Nb doped $SrTiO_3$. GO sheets embedded in the grain boundaries of polycrystalline ceramics facilitated electron transport without increasing much impact on its thermal conductivity, resulting in 50 times ZT enhancement due to GO incorporation. Furthermore, we have manipulated the metal-semiconductor transition in doped $SrTiO_3$by inducing enough strain as a result of graphite and Mxene inclusions and we reported the first experimental demonstration of ZT>1 in oxide thermoelectrics. Introduction of conductive graphite inclusions in Nb doped $SrTiO_3$(STN) matrix has led to a surge in electrical conductivity due to 21-times increase in weighted mobility of electrons resulting in high thermoelectric power factor ~5400 $\mu W/mK^2$, which is ~16-times higher than that of pure STN. Furthermore, we could restrain the increase in thermal conductivity by attaining enhanced Umklapp scattering along with phonon-glass-like temperature-independent phonon mean-free-path above Debye temperature. We have achieved the maximum ZT ~1.42 in STN+0.5wt% G composite. We have fabricated 4-legged n-type thermoelectric power generator demonstrating milliwatt-level power output, hitherto remained unattainable for oxide thermoelectrics. Our proposed way of designing rare-earth-free composites with graphite and MXene can potentially open up the possibility of fabricating novel thermoelectric generators.

ID: 05132
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Stress/Pressure-Stabilized Cubic Polymorph of Li3Sb with Improved Thermoelectric Performance

Mujde Yahyaoglu[1,2] , Thomas Soldi[3] , Melis Özen[1,2] , Christophe Candolfi[4] , G. Jeffrey Snyder[3] , Umut Aydemir[1]

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The solution to the current energy dilemma rests on a concerted e?ort to broaden our renewable energy resources and increase our energy e?ciency. Since almost two-thirds of the energy consumed worldwide is lost as waste heat, a new energy landscape would bene?t from the use of thermoelectric materials to produce electricity through waste heat recovery. Li_3Sb has two polymorphs crystallizing in a face-centered cubic cell (c-Li_3Sb; BiF_3 structure type; space group *Fmm*) and in a hexagonal unit cell (h-Li_3Sb; Na_3As structure type; space group $P6_3/mmc$). c-Li_3Sb was predicted to be a promising thermoelectric material based on recent first-principles studies; however, the experimental transport characteristics have remained unknown so far. Herein, we will report the successful preparation of the c-Li_3Sb by stress-induced mechanochemical synthesis (high-energy ball milling) along with its high-temperature thermoelectric properties. The hexagonal Li_3Sb (h-Li_3Sb) was revealed to be the stable phase at ambient conditions, while it starts unexpectedly transforming to the c-Li_3Sb by ball milling or under 60 MPa applied pressure at room temperature. Transport properties measurements performed on two polycrystalline specimens evidence that c-Li_3Sb behaves as a *p*-type degenerate semiconductor due to the formation of Li vacancies. In agreement with lattice dynamics calculations, c-Li_3Sb exhibits very low lattice thermal conductivity despite the lightweight of Li. A zT value of around 0.3 was obtained at 550 K. Modelling suggests that the hole concentration should be reduced through aliovalent substitutions or under Li-rich conditions for further optimization. Although the strong air sensitivity of Li_3Sb makes its use in thermoelectric applications challenging, this simple superionic binary provides an attractive experimental platform to elucidate the effect of stress/pressure on phase transition as well as Fermi surface complexity on thermoelectric properties.

ID: 05179
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

SrCuP and SrCuSb Zintl Phases as New Thermoelectrics screened by DFT

Adrien Moll[1] , Jean-Claude Crivello[1] , Jean-Marc Joubert[1] , Eric Alleno[1] , Céline Barreteau[1]

1) Institut de Chimie et des Matériaux Paris-Est

The experimental research of new thermoelectric materials with the support of screening calculations by DFT is an interesting approach to make the investigation of new candidates easier and more efficient. We recently reported a high-throughput calculation-based method for screening new materials[1,2]. The thermodynamic stability and the electronic properties of thousands of ternary intermetallic compounds were calculated to highlight new stable and non-metallic materials. Among the promising results, we focused on SrCuP and SrCuSb. The structural studies of these materials were only reported in 1978[3] and 1974[4] respectively, but their physical properties were never investigated.

A new synthesis method of SrCuP, based on mechanical alloying, will be presented. This process enables the easy synthesis of phosphorus-based materials, which are difficult to handle with conventional methods such as sealed quartz tube. The SrCuX (X=P or Sb) structures can be described within the Zintl concept, with large graphene-like $[CuX]^{2-}$ networks and Sr^{2+} intercalated layers. These materials are *p*-type with charge carriers concentrations around 10^{20} cm^{-3} and with notable high carriers mobilities and low effective masses. The thermal conductivities of both compounds are below 6 W/mK and with similar phonon contributions despite of the large mass differences between P and Sb. This effect is explained by DFT calculations. Combining these results, we obtain an *ZT* value of 0.2 and 0.1 at 600 K for SrCuP and SrCuSb without any extrinsic doping. This study opens opportunities for thermoelectric properties improvements of these new materials. It also experimentally supports our screening calculation approach for the research of new and non-metallic ternary intermetallic compounds.

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Session II - Parallel room

DEVICES & APPLICATIONS I

ID: 05049
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Implementation of devices self-powered by Fe2VAl-based thermoelectric generator for sensor applications in the scope of domestic hot water distribution

Valentin Marchal-Marchant[1] , Geoffrey Roy[1] , Camille van der Rest[1] , Mathieu Delcroix[1] , Olivier Poncelet[1] , Pascal J. Jacques[1]

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The fast emergence of the “Internet of Things” (IoT) leads to a massive dissemination of connected devices. Especially, in the case of domestic hot water, smart devices such as thermostat, connected gas sensors (humidity, CO2) or intelligent calorimeter can improve comfort and heating efficiency while decreasing energy consumption. The main drawback of these devices is their (lack of) autonomy mostly relying on batteries. This limits the system lifetime and its recyclability as well as increases maintenance operation needs. An alternative to batteries is the use of energy harvesting systems that can use heat exchange(s) inherent in a hot water circuit (up to 50°C hot source) to locally produce electricity using thermoelectricity. However, the extended growth of IoT field requires the use of sustainable thermoelectric materials. Full Heusler Fe_2VAl-based thermoelectric modules are very promising for such large-scale applications, besides off-the-shell thermoelectric modules.

A test bench has been developed to assess the capabilities of two designed and manufactured Fe_2VAl-based thermoelectric generators [1,2] operating respectively in two types of heat exchange environments (water-water and water-air). The generated electricity has been used by a DC-DC converter dedicated to low power energy harvesting system. The powered device integrated three sensors (humidity, CO2 and light), a supercapacitor for energy buffering and a Bluetooth communication module able to send the measurements to a smartphone app.

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ID: 05113
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Development of a novel gas thermoelctric generator (TEG) accumulating electricity and heat with ~80 % total efficiency

Roni Shneck[1] , Zinovi Dashevsky[1]

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The objective of the the present work is to improve the efficiency of gas thermoelectric generators (TEGs). I unque design of multilayer TE unicouple is suggested. Two types of thermoelctric materials were selected: "low temperature" TE materials (for operating temperature range of 300-600 K) - *n*-and *p*-type materials, based on Bi2Te3 compound with optimal crystal orientation, prepared by hot extrusion and "middle temperature TE materials (for operating temperature range of 600-900 K) including *n*-type type TE material based on PbTe compound and *p* -type TE material based on GeTe compound, prepared by SPS technique. The TE efficiency ? ~ 15 % was measured for TE multiilayer unicouple at the temperature difference ?*T* = 550 K (*T*h = 900 K), which is practically maximal efficiency in TE modules of gas TEGs. We suggest a system combining TEG and gas boiler into one autonomous source of electrical energy and heat for domestic use. The prototype of such hybrid system can produce ~ 400-500 W of electrical energy and 1700-2000 W of heat energy for heating and hot water supply.

ID: 05160
Type: Oral Presentation
Topic: Thermoelectric devices and applications

SUCCESSFUL DEVELOPMENTS IN THERMOELECTRIC APPLICATIONS

David Astrain[1] , Antonio Rodriguez[1] , Álvaro Martínez[1] , Patricia Aranguren[1] , Miguel Araiz[1] , Leyre Catalán[1] , Alvaro Casi[1] , Patricia Alegría[1] , Iranzu Erro[1] , Nerea Ariz[1]

1) Public University of Navarre

A thermoelectric device is a solid-state thermal engine with the capability of directly transforming electricity into heat or performing the inverse process without any moving parts. Therefore, this technology is much more reliable, compact, noiseless, and with less cost of maintenance than common mechanical thermal engines. Nevertheless, this technology is much less efficient than others based on vapor systems. Consequently, there is a lack of commercial applications of thermoelectric generators (TEG) and coolers (TEC), limiting their development. Thus, it is very important not only to improve the efficiency, but also to develop new and interesting applications.

In this work, two successful thermoelectric applications, carried out by the Thermal and Fluid Engineering research group, are presented. The first one is a novel TEG, which is able to supply electric power to volcanic monitoring stations, making them completely autonomous. This TEG uses the heat from fumaroles, which emit steam gases at a temperature close to 80ºC, to produce electric power, thanks to an optimized passive heat exchanger with phase change. The result is a prototype placed in Teide volcano (Canary Islands, Spain), at an 3500 m of altitude working properly during the day and night and with extreme weather conditions, outperforming the photovoltaic panels, which need heavy batteries to work. Thanks to thermoelectricity, it has been able to register geological parameters, and emit them, every 4 minutes, to a distance of 15 km, for the first time in this volcano.

The second developed application shows an interesting way to find new possibilities to bring thermoelectricity to the market: hybrid devices. It has been experimentally proved that the inclusion of a Thermoelectric Subcooling System (TESC) at the exit of the gas-cooler of a vapor compression cooling facility, working with transcritical CO2, increases 20% the overall efficiency of the refrigeration plant.

ID: 05172
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Investigating novel and cost-effective thermoelectric systems for energy recovery in steel making processes

Matthew Phillips[1] , Ugo Chiarotti[2] , Valerio Moroli[2] , Frank Mintus[3] , Susanna Bosi[4] , Mitja Padovan[4] , Stefano Spagnul[4] , Daniele Gaspardo[5] , Matteo Chini[5] , Alberto Viotto[5] , Loris Bianco[5] , Tim Bause[6] , Piero Fritella[6] , Nils Katenbrink[7] , Gao Min[1]

1) School of Engineering, Cardiff University UK 2) Rina Consulting – Centro Sviluppo Materiali SpA, Rome Italy 3) VDEh-Betriebsforschungsinstitut GmbH, Germany 4) Ergolines lab s.r.l., Italy 5) Ferriere Nord S.p.A, Italy 6) ESF Elbe-Stahlwerke Feralpi GmbH, Riesa D, Germany 7) QuickOhm Kupper & Co GMBH, Germany

Energy recovery from steel making processes is a promising application for thermoelectric technology to reduce the environmental impact of steel production. The EU funded THERELEXPRO and InTEGrated projects seek to investigate the use of thermoelectric generators (TEGs) to recover thermal energy from furnace cooling water and radiation from steel products. The preliminary THERELEXPRO tested two prototypes within an operational steel plant and identified challenges to the implementation of TEGs, including space for retrofitting, incorporation into existing systems and the harsh operating environment. The InTEGrated project focuses on solving these challenges through novel TEG and system design. We present key outcomes of the THERELEXPRO project and discuss some initial concepts and approaches of InTEGrated project.

Acknowledgment:

THERELEXPRO (No. RFSR-2013-00029) and InTEGrated (No. 899248) are funded by EU-RFCS Programme and coordinated by RINA CONSULTING. Tanujdeep Singh and Juan Arribas-Ramirez are also acknowledged for their contributions to the THERELEXPRO project

ID: 05272
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Use of Waste Heat Recovery Units to transform European energy demanding industries into more sustainable.

Dr. Raúl Aragonés[1]

1) Alternative Energy Innovations SL.

The European Union lose the 21% of its energy needs in form of waste heat.

Alternative Energy Innovations, spinoff of the Microelectronics Department of the Autonomous University of Barcelona, has developed a family of waste-heat thermoelectric-based recovery units (WHRU) to help heavy industries to revalorize their waste heat creating negative carbon electricity.

In this presentation, there will be introduced the HEAT-R project granted by the European Commission LIFE Programme (LIFE16 ENV/ES/000344). We will present 3 different WHRU approaches in 5 energy demanding industries of Cement, Steel, Paper, Food and chemical sectors. Major results are obtained in the Chemical sectors allowing a total power generation of 500W /m2 at 254ºC.

ID: 05334
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Development Status of Skutterudite-based Thermoelectric Technology for Integration into a Potential Skutterudite-Multi Mission Radioisotope Thermoelectric Generator

Thierry Caillat[1]

1) Jet Propulsion Laboratory/California Institute of Technology

The flight-proven Multi-Mission Radioisotope Thermoelectric Generator (MMRTG) is currently powering NASA's Mars Curiosity and Perseverance rovers and selected for NASA's Dragonfly mission. The MMRTG design includes sixteen thermoelectric converter modules, each composed of 48 couples of PbTe/TAGS (Lead-Telluride/Tellurium-Antimony-Germanium-Silver) packaged in fibrous insulation and is operated under inert gas in a hermetically sealed environment. The skutterudite (SKD)-MMRTG design in development would be nearly identical except for upgraded 48-couple modules using skutterudite-based couples. While preserving the majority of the flight-qualified MMRTG design, fabrication tooling and flight support equipment, the new skutterudite-based couples would provide a substantial improvement in power output over time. A team composed of Aerojet Rocketdyne (AR), Teledyne Energy Systems, Inc. (TESI), and NASA's Jet Propulsion Laboratory (JPL) team has been maturing the SKD technology for integration into a potential SKD-MMRTG. From JPL-transferred skutterudite technology, TESI has developed manufacturing capabilities for skutterudite-based couples as well as 48-couple modules, a building block of the SKD-MMRTG. Life assessment testing of SKD couples is underway at JPL and TESI, with some couples achieving more than three years of continuous time-on-test. Testing of SKD couples and a 48-couple module has shown that their power output is in good agreement with the predicted values. Physics-based life performance prediction models developed at JPL, and a newly developed one at TESI, utilize the couple test data results and analysis to forecast the generator power output. The current best-estimate power predictions for the SKD-MMRTG meets specification requirements and offers up to about 37% more power at End-Of-Design-Life (EODL) than the MMRTG at equivalent operating conditions. The SKD-MMRTG would provide a substantial improvement in power output in the out years, making it ideal for many long-duration, deep-space missions.

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Session III - Auditorium

MATERIALS & PROCESSING II

ID: 05047
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Beyond rattling: tetrahedrites as thermoelectric materials with low thermal conductivity

Paz Vaqueiro[1]

1) University of Reading

Ternary and quaternary copper sulfide minerals are attracting much interest owing to their abundance, low cost and promising thermoelectric properties. Many of these materials exhibit values of thermal conductivity well below those found for liquids, whilst retaining good electrical properties. Some of these materials, such as $Cu_{2-x}S$, exhibit ionic conductivity. It has been proposed that copper-ion mobility leads to a “liquid-like” state of the copper sublattice disturbing the propagation of phonons. There is a however a growing number of copper-based chalcogenides with intrinsically low thermal conductivities, which do not exhibit ionic conductivity. Here, our recent work on tetrahedrites, which are a family of copper sulfide minerals, will be presented, and the origin of their low thermal conductivity will be discussed.

For tetrahedrites, which are excellent *p*-type thermoelectric materials, their low thermal conductivity has been attributed to rattling vibrations of the trigonal-planar copper cations. Our work has shown that copper rattling in tetrahedrite is a direct consequence of a tetragonal-to-cubic phase transition at 90 K, which results in a sharp increase, by approximately 200%, of the atomic displacement parameters of the trigonal-planar copper cations. This phase transition is consequence of a Jahn-Teller electronic instability. In the cubic phase, the trigonal-planar copper cations form regular octahedral Cu_6^{7+} clusters, which are electronically degenerate, and undergo a Jahn-Teller distortion at 90 K. Below 90 K, the formation of “molecular-like” Cu_5^{7+} clusters suppresses copper rattling vibrations due to the strengthening of direct copper-copper interactions. Our recent quasielastic neutron scattering measurements on stoichiometric and copper-rich tetrahedrites, $Cu_{12}Sb_4S_{13}$ and $Cu_{14}Sb_4S_{13}$, combined with molecular dynamics simulations, reveal that above 200 K copper diffusion occurs. In copper-rich tetrahedrites at high temperatures, long-range copper diffusion occurs, in agreement with previous observations of superionic conduction. However, in stoichiometric tetrahedrite, copper diffusion is confined to the cages containing the Cu_6^{7+} clusters.

ID: 05062
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

3D Bi2Te3 cross-linked nanowire network metamaterials for enhanced thermoelectric efficiency

Alejandra Ruiz de Clavijo[1] , Olga Caballero[1] , Cristina V. Manzano[1] , Albert Beardo[2] , F. Xavier Álvarez[2] , Marisol Martín-González[1]

1) CSIC 2) Universidad Autonoma de Barcelona

n addition, the Seebeck coefficient is twice that of nanowires and nanofilms and 50% higher than that of single crystals. As an educated guess, this increase has been explained as a non-equilibrium effect induced by the geometry of the structure on the phonon distribution function, resulting in increased phonon resistance. Although other explanations can be also possible. These thermoelectric metamaterials have higher performance and are fabricated in large areas by low-cost processes, making them suitable for mass production.

3D interconnected nanowire scaffolds have been shown to improve thermoelectric efficiency compared to 1D nanowires of similar diameter and films grown under similar electrodeposition conditions. The Bi2Te3 3D nanonetwork reduces thermal conductivity while maintaining the high electrical conductivity of the film. The decrease in ?T is modeled using the hydrodynamic heat transport equation and can be understood as a thermal viscosity effect due to the 3D nanostructure interconnections.

3D interconnected nanowire scaffolds have been shown to improve thermoelectric efficiency compared to 1D nanowires of similar diameters and films grown under similar electrodeposition conditions. The Bi2Te3 3D nanonetwork reduces thermal conductivity while maintaining the high electrical conductivity of the film. The decrease in ?T is modeled using the hydrodynamic heat transport equation and can be understood as a thermal viscosity effect due to the 3D nanostructure. In addition, the Seebeck coefficient is twice that of nanowires and nanofilms and 50% higher than that of single crystals. As an educated guess, this increase has been explained as a non-equilibrium effect induced by the geometry of the structure on the phonon distribution function, resulting in increased phonon resistance. Although other explanations can be also possible. These thermoelectric metamaterials have higher performance and are fabricated in large areas by low-cost processes, making them suitable for mass production.

ID: 05087
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Thermoelectric properties of high-entropy-type metal chalcogenide compounds

Aichi Yamashita[1] , Mizuguchi Yoshikazu[1]

1) Tokyo Metropolitan Univ.

High-entropy-alloys (HEAs) have attracted much attention in the fields of materials science and engineering because of their tunable properties as structural materials, such as excellent mechanical performance under extreme conditions [1]. HEAs are the alloys with 5 or more constituent elements and a 5–35% occupancy of each element, resulting a high configurational mixing entropy value ($D S_{mix}$) above 1.5 [1]. Introduction of large disturbances and strains due to the multiple elements solid solution is the core of excellent mechanical performances. Even in thermoelectric materials, it is expected that the lattice thermal conductivity (k_L) will be reduced by introducing such large disturbances. In 2015, Shafeie et al. reported a HEA $Al_xCoCrFeNi$-based thermoelectric material [2]. On the other hand, because it is an alloy, the thermoelectric figure of merit (ZT) remains low due to its low Seebeck coefficient and high electron thermal conductivity.

So far, we have developed a new high-entropy superconductors that extends the concept of HEA from a simple alloy to a compound [3]. Among them, we successfully synthesized the new high-entropy-type metal chalcogenide superconductor by mixing the multi-site with multiple elements simultaneously, resulting the highest $D S_{mix}$ vaule of 2.00 [4]. We also applied this new concept for metal chalcogenide (PbSe-based) thermoelectric material [5]. Synchrotron X-ray diffraction patterns show a uniformity of sample. The very low k_L value of 0.5 W/mK and relatively high ZT value of 0.54 were observed at 723 K. We will talk details of the development of metal chalcogenide based high-entropy thermoelectric materials.

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ID: 05099
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Unexpected Complexity in Substituted Chalcopyrites

Anthony Powell[1] , Sahil Tippireddy[1] , Paz Vaqueiro[1] , Ricardo Grau Crespo[1] , Vikram Vikram[1]

1) University of Reading

Copper chalcogenides have attracted considerable interest as candidate materials for thermoelectric devices for energy harvesting in the mid-range of temperatures. The binary copper chalcogenides exhibit exceptionally low thermal conductivities, which has been attributed to the entry of the copper sub-lattice into a liquid like state, leading to the description phonon liquid electron crystal (PLEC). However, Cu-ion mobility results in degradation under the potential created by a temperature gradient.

In an effort to address this problem we are investigating materials containing the same tetrahedral CuQ_4 (Q = chalcogen) building unit present in the binary phases, together with additional cations in an effort to supress Cu-ion mobility. This includes preparation of a wide range of materials derived from chalcopyrite ($CuFeS_2$): a rare example of an n-type sulphide. We have sought to exploit chemical substitution, with high-valence cations including Sn^{4+}, Ge^{4+} and Cr^{3+}, at both the copper and iron sites in an effort to effect significant enhancements in charge-carrier concentration. This has resulted in improved thermoelectric properties. Figures of merit of ZT » 0.3 at 673 K have been achieved in Sn- and Cr-substituted phases, while values approach ZT = 0.4 on Ge-substitution. However, the increase in carrier concentration is less than expected on the basis of formal electron-counting. Application of a suite of state-of-the-art characterization and computational techniques reveals this is associated with a greater degree of complexity than anticipated for straightforward cation substitution. In addition to formation of micro-precipitates of substituent-rich phases; charge-localization through the formation of small polarons has been observed, together with changes in local structure. These results suggest an unexpected richness to composition-structure-property relations in *n*-type chalcopyrites.

ID: 05233
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Interplay of electron structure features and entropy improving thermoelectric properties in Cu7P(SxSe1?x)6 p-type argyrodites

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Chemical disorder appears to be driving factor of crystal structure phase transition but may also open new paths to influence electron transport properties by intentional modification of Fermi surface parameters of electrons or holes. $Cu_7P(S_xSe_{1?x})_6$. argyrodites with multi-atom disorder and complex crystallographic phase diagram offer opportunities to study in more details correlations between configurational entropy, phase stabilisation and multi-valley character of electronic band structure, promoting high thermoelectric properties [1]. In order to have an insight into the subtle changes of electronic states in disordered systems, the Korringa-Kohn-Rostoker method combined with the coherent potential approximation (CPA) was used for *ab initio* electronic structure calculations [2]. This approach allows to treat self-consistenly complex substitutional and calculate density of states and electronic dispersion curves E(k) with complex energy in disordered materials [3]. In this work, we present markedly different effect of such kind of disorder on valence bands (development of multivalley band character in the Brillouin zone center) and conduction bands (progressive gap opening), being likely responsible for enhancement of thermoelectric properties in p-type $Cu_7P(S_xSe_{1?x})_6$. We also show that the promotion of the crystal symmetry, caused by an increase of configuration entropy, forms a multivalley character of the valence band near the Fermi level. As a result of the significant increase of the charge carrier concentration without changing the Fermi level position, the remarkable enhancement of the effective mass was observed for high-symmetry ?-Cu7PS3Se3 (SG F-43m) in comparison to low-symmetry ?-Cu7PSe6 or ?-Cu7PS6 (SG P213) [1].

Acknowledgments The research was funded by the Foundation for Polish Science (TEAM-TECH/2016-2/14 Grant “New approach for the development of efficient materials for direct conversion of heat into electricity”), co-financed by the European Union under the European Regional Development Fund.

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Session IV - Parallel room

DEVICES & APPLICATIONS II

ID: 05213
Type: Oral Presentation
Topic: Thermoelectric devices and applications

3D printing of inorganic thermoelectric materials and devices

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Heat is omnipresent in natural and artificial environments, more than 60% of which is dissipated. Thermoelectric (TE) power generation can provide a unique solution to convert this dissipated, wasted heat into useful energy, that is, electricity. Generally, TE conversion efficiency depends on the material properties and design of the module structure. However, despite the development of highly efficient TE materials, module engineering is rather less advanced and is still fabricated by the traditional multi-step process of materials synthesis, dicing, and assembly, restricting the available design of modules to that of planar structures. At this moment, three-dimensional (3D) printing technology can maximize the flexibility in the design and fabrication of TE modules into more efficient structures. Furthermore, the printing process can significantly reduce the processing cost for the fabrication of TE modules owing to lower energy input and a simplified assembly process. Herein, I present the development of the 3D printing process applied to a range of different TE materials of Bi_2Te_3, BiSbTe, PbTe, and Cu_2Se-based inorganic alloys. The particle-based inks were formulated with the tailored surfaces of particles, achieving the suitable viscoelasticity of the TE inks to the extrusion-based 3D printing. The geometrically designed 3D-printed TE materials were assembled into power generating systems, in which high efficiencies of energy conversion were achieved by the optimization of heat transfer.

ID: 05146
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Magnesium-silicide-based thermoelectric generators: prototypes, analysis and challenges

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Magnesium silicide based solid solutions Mg_2X (X = Si, Ge, Sn) are among the most promising thermoelectric (TE) materials for the temperature range of 500 K to 800 K, where a large fraction of the reusable heat is available. Very good TE properties have been demonstrated repeatedly, especially for the n-type material. This, combined with a high material availability, low cost of raw materials and environmental compatibility, makes these materials preferred candidates for large scale applications.

We have fabricated TEG prototypes using previously optimized n- and p-type $Mg_2Si_{1-x}Sn_x$ powder-compacted material. These show efficiencies approaching 4% for and and a power density of 0.9 W/cm² with respect to the TE material area, the first efficiency result for a full Mg_2X-based TEG and among the best for TEG from sustainable and available materials. Measured module parameters (open circuit voltage, optimum current, power output, heat flow, inner resistance) are compared with predictions from a constant property model based on measured material and interface properties. This analysis allows to identify and assess potential loss and degradation mechanisms, including material changes due to thermal load and TE material/ electrode interaction, like crack development or charge carrier compensation. Approaches to overcome these challenges ranging from material coating to electrode design and modifications of the module assembly process are analyzed for their effectiveness. This will pave the way for long-term stable modules exceeding 5% conversion efficiency. Finally, results for an upscaled Mg_2X-based TEG fabrication by an industrial-ready process will be discussed.

ID: 04943
Type: Oral Presentation
Topic: Thermoelectric devices and applications

High power 2.5D integrated thermoelectric generators combined with microchannels technology

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We have developed high power integrated thermoelectric generators (µTEGs). These µTEGs are CMOS compatible, i.e. based on polycristalline SiGe materials. These µTEGs have been processed directly on a silicon interposer. Even if poly-SiGe exhibits low thermoelectric performances at room temperature, the specific design and proposed architecture enable µTEGs to deliver up to 680 µW for a temperature difference at 15.5 K. To reach such high power, an original 2.5D structure has been developed and µchannels technology has been associated, below the µTEG, to dissipate heat coming from the hot side. µTEGs have been tested in real environment, located below a hot test chip. Such µTEG performances overtake those from similar state-of-the-art CMOS compatible devices, and pave the way for a potential use in different applications such as sensors power supply or battery charger.

ID: 05244
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Heat sink implementation on micro-thermoelectric generators (µTEGs) for power enhancement

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Internet of Things (IoT) is raising as one of the keystones of our society. The ability to gather information from our environment enhances the decision-taking processes and is crucial for our prosperity. One of the main needs of IoT is the self-sustainable supply of energy to the sensors. Among the available environmental sources, heat can be harvested by means of thermoelectric devices.

Earlier, the group developed all-Si based µTEGs with planar architecture. This work reports on a procedure to reliably place a heat sink on top of the devices, and on how this has improved the power output to match IoT demands (10-100 µW/cm^2). The µTEG is based on Si (or SiGe) nanowires (NWs) acting as the thermoelectric material connecting a Si bulk and a thermally isolated suspended platform. The thermal resistance from the suspended platform to the ambient limits the power generated by the µTEG. A heat sink enhances this heat exchange, increasing the ?T across the NWs. Its integration on top of the fragile suspended platforms has only been possible by means of a µTEG-to-heat sink adapter, together with the development of a custom pick and place system.

The µTEG with the heat sink shows an increase in the generated voltage of up to six times with respect to that of a bare µTEG, leading to output powers well within the range of IoT needs. When placing the µTEG on top of a heat source above 200 ºC, it generates slightly more than 10 µW/cm^2 and more than 80 µW/cm^2 when in still air convection and when in an airflow of 1.3 m/s (equivalent to a natural breeze), respectively. These results demonstrate the validity of the µTEGs with heat sink as power supplies for IoT applications at the microscale.

ID: 05009
Type: Oral Presentation
Topic: Thermoelectric devices and applications

On-chip silicon-based thermoelectric generator

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The idea of a thermoelectric generator (TEG) completely integrated on-chip and able to power up an electronic circuit offers an intriguing challenge, especially within the Internet of things framework. The choice of silicon nanostructures as core components of these TEGs has been widely investigated [1,2]. Dense arrays of silicon nanostructures give the advantages of a reduced thermal conductivity combined with a great amount of current delivered to an electrical load, thanks to a cross section wider than the one of single nanowires. We present a TEG, on silicon on insulator (SOI) substrate, composed by two complementary-doped arrays of hundreds of parallel nanobeams defined by means of high-resolution lithography and deep reactive ion etching. The two arrays are connected through a central silicon body that acts as a hot source. A right and a left side collector complete the structure and act as cold sink. The use of SOI wafer guarantees the possibility to suspend the nanostructures, through a buried silicon oxide etching, and hence to avoid heat losses through the substrate. Warming up the central body the heat-flux is canalized through the nanobeams, and both the cold temperature and the Seebeck voltage can be measured on the collector pads. The results will be presented and discussed.

This project has received funding from the EU-â€•H2020 research and innovation programme under grant agreement No 654360 having benefitted from the access provided by CSIC (IMB-CNM) Cerdanyola del Valles, Spain within the framework of the NFFA-â€•Europe Transnational Access Activity. We thank Miguel Zabala, Roser Mas and the staff of the SBCNM for the kind support.

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Session V - Auditorium

MATERIALS & PROCESSING III

ID: 05214
Type: Indiferrent
Topic: Emerging topics

Machine-Learning Guided Prospection Of Efficient Thermoelectric Chalcogenides

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Chalcogenide materials like Bi_2Te_3 or GeTe are well-known for their good thermoelectric properties either at room or high temperatures, respectively [1]. Recently, it has been emphasized that the peculiar nature of the metavalent bonding mechanism seems to be related to their relatively good performance [2]. Yet, numerous compounds with further improved thermoelectric properties remain to be discovered for supporting a multi-modal approach to a sustainable energetic future. To this end, the rational design of thermoelectric materials can be drastically accelerated thanks to machine learning techniques by comparison to the classic and time-consuming trial-and-error methodology. In this sense, efficient and insightful studies assisted by machine learning can be conducted by encompassing experimental process and expert knowledge. One of the main routes to improve bulk thermoelectric properties is chemical doping either by substituting elements or by creating vacancies in the crystal structure. In this framework, we have investigated new chemical compositions of doped CuS, SnTe and GeTe supported by machine learning and further characterized in the lab. Traditional as well as cutting-edge deep learning techniques were considered along with Bayesian optimization for predicting thermoelectric properties of novel compounds as well as proposing candidates for improved performance. Moreover, this opens the possibility to bring novel insights on the underlying physical mechanisms in these very interesting metavalent materials. Here, we will present the preliminary results of this study on these slightly-doped binary chalcogenides.

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ID: 05264
Type: Oral Presentation
Topic: Transport phenomena

Interplay between magnetism and thermopower in the CuCr1-xTi1+xS4 thiospinels

Sylvie Hebert[1] , Oleg Lebedev[1] , Antoine Maignan[1]

1) Laboratoire CRISMAT, Normandie Université, CNRS, Ensicaen et Unicaen

The thermoelectric properties can depend on the magnetic field and on the magnetic properties of the materials as shown for example in the case of metallic and ferromagnetic nanowires [1] or in the case of chalcogenides such as $CuCrTiS_4$ [2]. For this compound, a large magnetothermopower of -26% had been evidenced at low temperature (20K), together with a large and negative magnetoresistance of -95% at 5K. Several mechanisms can contribute to these magnetotransport effects, and it has been shown that the paramagnetic fluctuations play a key role to generate this magnetothermopower. The $CuCr_{1-x}Ti_{1+x}S_4$ thiospinel family actually presents a complex magnetic phase diagram around x =0, with a modification of the magnetic fluctuations [3]. We will show how the transport properties are affected by this complex phase diagram, and how the thermopower and magnetothermopower sign by itself are modified [4].

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[2] : David Berthebaud, Oleg I. Lebedev, Antoine Maignan, and Sylvie Hébert, J. Appl. Phys. 124, 063905 (2018).
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ID: 05122
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Origins of low thermal conductivity and high thermoelectric performance in Cl-doped Bi2Te3-xSex alloys

Taras Parashchuk[1] , Rafal Knura[1] , Oleksandr Cherniushok[1] , Krzysztof T. Wojciechowski[1]

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Bi_2Te_3-based alloys are the main materials for the construction of low- and medium-temperature thermoelectric modules. In this work, the microstructure and thermoelectric properties of Cl-doped $Bi_2Te_{3-x}Se_x$ alloys were systematically investigated considering the high anisotropy inherent in bismuth telluride-based materials. The prepared samples pose highly-oriented microstructure morphology, which results in very different thermal transport properties in two pressing directions. To accurately separate the lattice, electronic and bipolar components of the thermal conductivity over the entire temperature range, we employed a two-band Kane model to the Cl-doped $Bi_2Te_{3-x}Se_x$ alloys. It was established that Cl atoms act as electron donors, which tune the carrier concentration and effectively suppress the minority carrier transport in $Bi_2Te_{3-x}Se_x$ alloys. The estimated value of the lattice thermal conductivity was found to be as low as 0.15 $Wm^{-1}K^{-1}$ for $Bi_2Te_{3-x-y}Se_xCl_y$ at 673 K in parallel to the pressing direction, which is among the lowest values reported for crystalline materials. The large reduction of the lattice thermal conductivity in both pressing directions for the investigated $Bi_2Te_{3-x}Se_x$ alloys is connected with the different polarities of the Bi-(Te/Se)1 and Bi-(Te/Se)2 bonds, while the lone-pair (Te/Se) interactions are mainly responsible for the extremely low lattice thermal conductivity in the parallel direction. As a result of improved power factor, suppressed bipolar conduction, and ultralow lattice thermal conductivity, a maximum *ZT* of 1.0 at 473 K has been realized in Cl-doped $Bi_2Te_{3-x-y}Se_xCl_y$-based sample.

Acknowledgments
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ID: 05139
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Enhanced stability in nanostructured p-type Pb0.993-xNaxGe0.007Te for high efficiency thermoelectric modules

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Among the materials for thermoelectric modules, which can convert (waste) heat directly into electricity, PbTe has been established as one of the prime candidates for medium-to-high temperature range applications. Recently, maximum conversion efficiencies $?_{max}$ ~ 8.5 % and $?_{max}$ ~ 12% in single-stage and cascaded modules, respectively, made from nanostructured and Na-rich (4%) PbTe ($Pb_{0.953}Na_{0.04}Ge_{0.007}Te$) have been demonstrated for temperature difference $?T$ ~590 K. [1] Although 4% Na content in p-type PbTe has been shown to be beneficial for the thermoelectric performance of the materials by promoting convergence of the light and heavy valence bands at high temperature, Na-rich precipitates found at room temperature affect material stability negatively. [2] In this study, a significant improvement of the material stability of p-type $Pb_{0.993-x}Na_xGe_{0.007}Te$ was achieved by reducing the Na content from x = 0.04 used in previous studies to x = 0.02 while still maintaining high thermoelectric performance with zT_{peak} ~ 2.2 at 813 K. No Na-rich precipitates in x = 0.02 sample were found at room temperature The improved material stability is demonstrated by the significantly increased onset temperature for plastic deformation for x = 0.02 sample (T_{onset} ~ 690 K) compared to x = 0.04 sample (T_{onset} ~ 500 K) when heating bar-shaped samples in 3-point bending geometry with constant applied force. These results, as well as finite-element method simulations and measurements of the power generation characteristics of single legs and modules prepared using p-type $Pb_{0.973}Na_{0.02}Ge_{0.007}Te$ will be discussed in detail.

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Acknowledgment

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ID: 04997
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Do we really know our old materials? Bi2Se3 as a prototype of promising thermoelectric materials?

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Thermoelectric (TE) materials studied in the second half of the 20th century proved to be the best choice for applications, while new materials remained on the sidelines. There are many reasons why this is the case, and we may think that after 70 years of research, there is almost nothing to improve on TE materials. But that may not be entirely true. Let's take a closer look at one of the leading TE materials, Bi2Se3. Do we really know how it works? We don't think so at all. It seems that its family in particular still hides many secrets that can help deepen our understanding of thermoelectricity. Namely, Bi2Se3 attracted a lot of attention during the second half of the 20th century as a TE material that has been studied almost exclusively in this field for almost 40 years. Later, in connection with TE, doping with magnetic ions led to the formation of dilute magnetic semiconductors, which provided an impetus for further research in the field of spintronics. At the beginning of the millennium, Bi2Se3 became a prominent 3D topological insulator with pronounced surface conditions and even a superconductor or a real 2D conductor. This provoked further in-depth examination of its properties. Is it a coincidence? Our goal is to analyze all recent findings along with our new results in terms of thermoelectricity. We want to show that Bi2Se3 is to some extent unknown. Although it is advisable to look for new materials, re-examining old materials using advanced techniques may provide a useful guide to thermoelectricity.

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Session VI - Parallel room

DEVICES & APPLICATIONS III

ID: 05177
Type: Oral Presentation
Topic: Thermoelectric materials and materials processing

Tuning Electronic and Ionic Transport by Carbon-based Additives in Polymer Electrolytes for Thermoelectric Applications

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Ionic transport constitutes a key process in organic-based electrochemical energy storage media. As such, much work on the electrochemical device performance is devoted to cyclability, energy density and conductivity. Vice versa, these ionic transport characteristics offer intriguing possibilities for the utilization of these materials in thermoelectrics (TE) to recuperate waste heat into electrical power.
In this work we present our results on the electrical and thermoelectrical characterization of a methacrylate based solution processable solid polymer electrolyte. By means of impedance spectroscopy over a broad frequency regime from 100 mHz up to 500 kHz and in a technologically relevant temperature range between 263 K and 363 K we investigate the dynamics of charge carriers in the solidified electrolyte. The observed high ionic conductivity of about 10^{-3} S m^{-1} at room temperature distinguishes this material for lithium battery applications [1]. In combination with highly sensitive thermovoltage measurements, we demonstrate that the electronic and ionic transport properties can be efficiently varied by the ratio between Lithium-salt and carbon-based additives, in this case carbon nanotubes. By this approach we are able to increase the power factor by several orders of magnitude. Even more, we can reverse the sign of the occurring thermovoltage, which allows for different TE operational modes depending on ambient temperature. Different transport mechanisms such as Vogel-Fulcher-Tammann and Arrhenius behavior are identified.
In combination with the high electrical conductivity achieved on macroscopic length scales, high thermovoltages of about 2 mV K^{-1} allow for high output powers while the polymeric matrix maintains the temperature gradient which in turn is a prerequisite for application in thermoelectric generators (TEG). A proof-of-concept all organic TEG verifies the functionality of our approach and, thereby, substantiates the potential of mixed ionic and electronic materials for future TE applications.
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ID: 05240
Type: Oral Presentation
Topic: Thermoelectric devices and applications

From sustainable thermoelectric materials to devices: a contribution

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Thermoelectric materials currently used in commercial devices either possess toxic, rare or expensive elements, with much research being made to discover newer, cheaper and more environmentally friendly alternatives to replace them. These efforts already led to the identification of novel thermoelectric materials, like skutterudites, Half-Heusler and clathrates. More recently, metallic sulphides, antimonides and silicides have also been optimized or recognized for having good potential for thermoelectricity. From these, we highlight the potential application of more sustainable materials such as tetrahedrites ($Cu_{12}Sb_4S_{13}$) and MgAgSb, as p-type semiconductors, and Mg_2X (X=Si, Sn), as n-type semiconductor, that can work in the range of 350 K to 623 K. These materials are mostly composed of cheap and non-toxic elements that exist in high natural abundance, and show both p- and n-type behaviour with high thermoelectric figures of merit. However, to build a thermoelectric device is not enough to have “green” and inexpensive p- and n-type materials with good thermoelectric properties. It is also fundamental to have a high thermal and chemical stability, especially when in contact with other substances. Moreover, low cost and efficient production processes are also needed in order to envisage widespread or large-scale applications. In this communication, an overview of the research made in our group to produce and optimize the thermoelectric properties of tetrahedrite, MgAgSb and Mg_2X, as well as the investigation made on their thermal stability and chemical reactivity, will be presented. Particularly, the development of cheap and fast production processes, and the study of oxidation, reactivity with electrodes, diffusion barriers and electrical contacts quality will be briefly shown.

ID: 05125
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Fabrication and characterization of an environmentally friendly Mg2(Si,Sn) thermoelectric generator

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Magnesium silicide-stannide $Mg_2(Si,Sn)$ solid solutions are high-performance thermoelectric (TE) materials with the advantage of being composed of light, cheap and abundant elements. Therefore, they are especially attractive for the conversion of waste heat into electricity in fields like automotive or aerospace. While the materials properties have been well optimized, information on the fabrication and performance analysis of $Mg_2(Si,Sn)$-based TEG is quite scarce.

We have fabricated and measured such a TEG prototype, and obtained efficiencies approaching 4% for ?*T* = 375 K and a power density of 0.9 W/cm² with respect to the cross section of the TE legs. This measured efficiency is ~30% lower than what would be expected based on a constant property model including contact resistances, a phenomenon usually observed in TEGs.

In order to identify the origin of the discrepancy two further two-pairs TEG prototypes were built and a new measurement design was applied, where additional voltage probes are connected to the bridges between the legs. This allows for monitoring each leg's electrical resistance independently. One origin of the observed losses was successfully identified, as it was repeatedly found that one or both n-type legs have a significantly higher resistance than predicted, while the resistances of the p-type legs match their expected values. This is attributed to crack formation and propagation during temperature cycling. Indeed, cracks were observed in micrographs of the n-type legs close to the metallization layer. They are probably caused by mechanical stress induced by differential thermal expansion. However, the fact that this is systematically not observed in p-type legs, which have similar mechanical properties but a wider base, indicates that one strategy to avoid cracking could be to increase the cross-section of the n-type legs. Other improvement strategies are suggested and a final TEG is built and its performance is assessed and presented.

ID: 05142
Type: Oral Presentation
Topic: Thermoelectric devices and applications

Molecular Heat Engines

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Theory predicts that heat to charge conversion is so efficient in single molecules, that they hold unexplored potential as building blocks for thermoelectric energy harvesting [1]. Empirical verification of these predictions is, however, still to come, because of the considerable experimental challenges. In this talk, I will present our recent progress on experimental, thermoelectric studies of single molecules. We unravel the spin entropy variations associated with electron flow through molecules, extracting fundamental thermodynamic information about the molecule and its electronic ground and excited states (see Figure 1) [2]. We use a novel measurement protocol that simultaneously probes the DC and AC conductances and the thermocurrent flow as a function of bias and gate voltage. We show that the thermocurrent maps allow direct insight into the interplay of spin and vibrational degrees of freedom in single molecule spintronics [3]. These results open the path to the use of thermocurrent for the study of the internal processes during transport in single magnetic molecules. Moreover, they offer a fundamental tool necessary for the fundamental study of spin flip and decoherence, or Kondo correlations [4] in single molecule systems out of equilibrium.

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Micro Thermoelectric Devices: from thermal management to powering the internet of things.

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Micro-thermoelectric devices (µTEDs) have a high potential for future applications in the biomedical field, powering internet-of-things devices, and thermal management. For such applications, µTEDs need to have a robust packaging so that the devices can be brought in direct thermal contact with the target heat sink and source. The packaging technology developed for macroscopic modules needs improvement as it cannot be applied to µTEDs due to a large thermal resistance between the capping material and the device which deteriorates its performance. Here, we develop µTEDs using optimized geometry and contact resistance combined with a novel packaging technique that is fully compatible with on-chip integration.

We developed a process for the fabrication of µTEDs with vertically free-standing leg pairs without a top plate. The fabricated µTEDs were embedded in a photoresist for using the device in applications. The fabrication of the µTED is based on the photolithographic patterning process in combination with electrochemical deposition of $Bi_2(Te_xSe_{1-x})_3$ and Te as n-type and p-type thermoelectric materials respectively. Using the optimized geometry and contact resistance, the maximum net cooling temperature and the cycling reliability were enhanced. The geometrically optimized µTED with low contact resistance showed a maximum cooling of around 10.8K at an applied electrical current of 235mA, a rapid response time of 700µs and survived over 100 million cooling cycles in our reliability studies [1]. The fabricated µTED can be used to scavenge waste heat to provide solid-state electricity for powering electronics and have potential applications in wearable electronics, and wireless sensors. These embedded, optimized, stable and easily scalable µTEDs open new avenues for widespread applications in biomedical applications, powering internet-of-things devices, and local heat management.

Reference

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