ACT3 & MNHTE 2018

Design and fabrication of high performance thermoelectric modules

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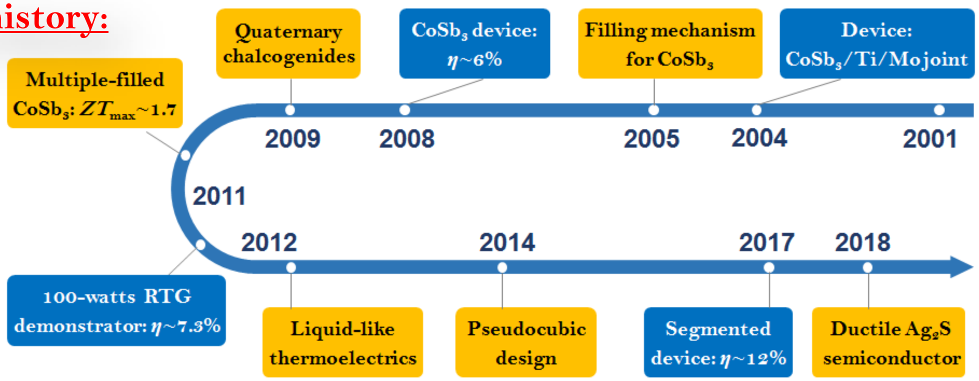
Outline

1. Brief introduction to SICCAS TE group
2. Current status of thermoelectric modules
3. Topologic design of TEG modules
4. Interface design of TEG modules
5. Performance & service behavior testing
6. Summary

Our goal : Full chain TE technology development

Our history:



Quaternary chalcogenides — 2009

$CoSb_3$ device: η~6% — 2008

Filling mechanism for $CoSb_3$ — 2005

Device: $CoSb_3$/Ti/Mojoint — 2004

2001

Multiple-filled $CoSb_3$: ZT_{max}~1.7 — 2011

100-watts RTG demonstrator: η~7.3%

Liquid-like thermoelectrics — 2012

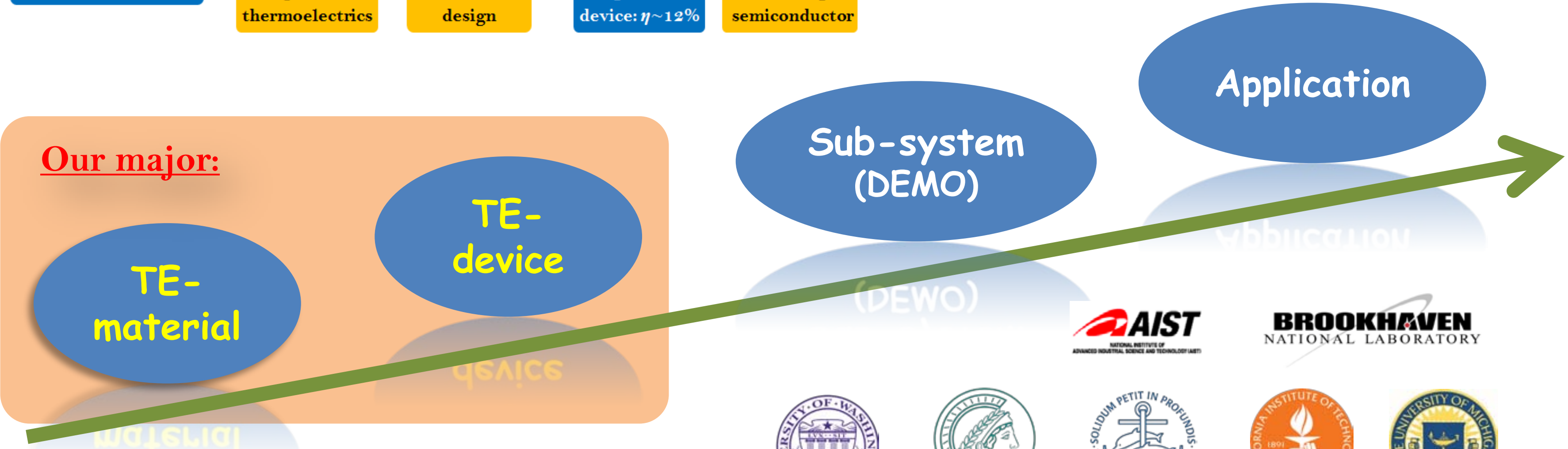
Pseudocubic design — 2014

Segmented device: η~12% — 2017

Ductile Ag_2S semiconductor — 2018

- ☐ 17+ years research history
- ☐ Focus on TE materials & devices
- ☐ Full chain technology development
- ☐ World-wide cooperation

Our major:



TE-material

TE-device

Sub-system (DEMO)

Application

SAIC 上汽集团 SAIC MOTOR

Materials Magic Hitachi Metals

SAMSUNG

FerroTec

工業技術研究院 Industrial Technology Research Institute

BAOSTEEL

CORNING Discovering Beyond Imagination

OAK RIDGE National Laboratory

AIST

BROOKHAVEN NATIONAL LABORATORY

UNIVERSITY OF WASHINGTON 1861

MAX-PLANCK-GESELLSCHAFT

UNIVERSITAS ARHUSIENSIS

CALIFORNIA INSTITUTE OF TECHNOLOGY

THE UNIVERSITY OF MICHIGAN 1817

TONGJI UNIVERSITY 1907

DONGHUA UNIVERSITY 1951

XI'AN JIAOTONG UNIVERSITY 1896

HIT 1920

TSINGHUA UNIVERSITY 1911

ZHEJIANG UNIVERSITY 1897

WUHAN UNIVERSITY OF TECHNOLOGY

SHANGHAI UNIVERSITY

SOUTH UNIVERSITY OF SCIENCE AND TECHNOLOGY OF CHINA 2011

22 Staffs：Multiple Disciplinary Team

Team Leader	Fundamental Research Leader	Engineering Research Leader
		
Prof. **Lidong Chen**	*Prof.* **Xun Shi**	*Prof.* **Shengqiang Bai**

Qin Yao	*Associate Prof.*	**Organic TE**
Pengfei Qiu	*Associate Prof.*	**Inorganic TE**
Ruiheng Liu	*Associate Prof.*	**TE application**
Sanyin Qu	*Research Assistant*	**Organic TE**
Tianran Wei	*Research Assistant*	**Inorganic TE**
Qihao Zhang	*Research Assistant*	**Device design**
Minghui Wang	*Secretary*	
Dudi Ren	*Technician*	
Jie Xiao	*Technician*	

Jiehua Wu	*Associate Prof.*	**Service behavior**
Xugui Xia	*Senior Engineer*	**Device integration**
Ting Wu	*Senior Engineer*	**Industrialization**
Junqiang Song	*Research Assistant*	**Industrialization**
Jincheng Liao	*Engineer*	**Device measurement**
Ming Gu	*Engineer*	**Service behavior**
Chao Wang	*Engineer*	**Device integration**
Tingting Yang	*Technician*	
Hongxiu Zhou	*Technician*	

~30 Ph. D. / Master candidates in TE group

Platform: Support TE design, fabrication, measurements

Total Lab area: ~1500 m² （in SICCAS Jiading campus）

- ☐ Synthesis & performances evaluations for TE materials: 4K ~ 1100K
- ☐ Fabrication & performances evaluations for TE modules: R.T. ~ 1200K
- ☐ Mass production of bulk materials (SKD, HH, SG, …): ~50 kg/year
- ☐ Mass production of Bi_2Te_3- & SKD-based TEG modules: ~ 10 kW/year
- ☐ Measurement standards for TE materials & modules: IEA-AMT member

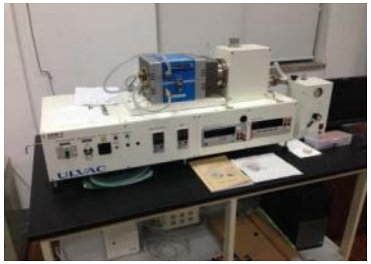
材料合成
(Materials preparation)

热处理
(Melting & annealing)

材料加工
(Materials & module)

材料切割
(Elements)

接触电阻测试
(Element testing)

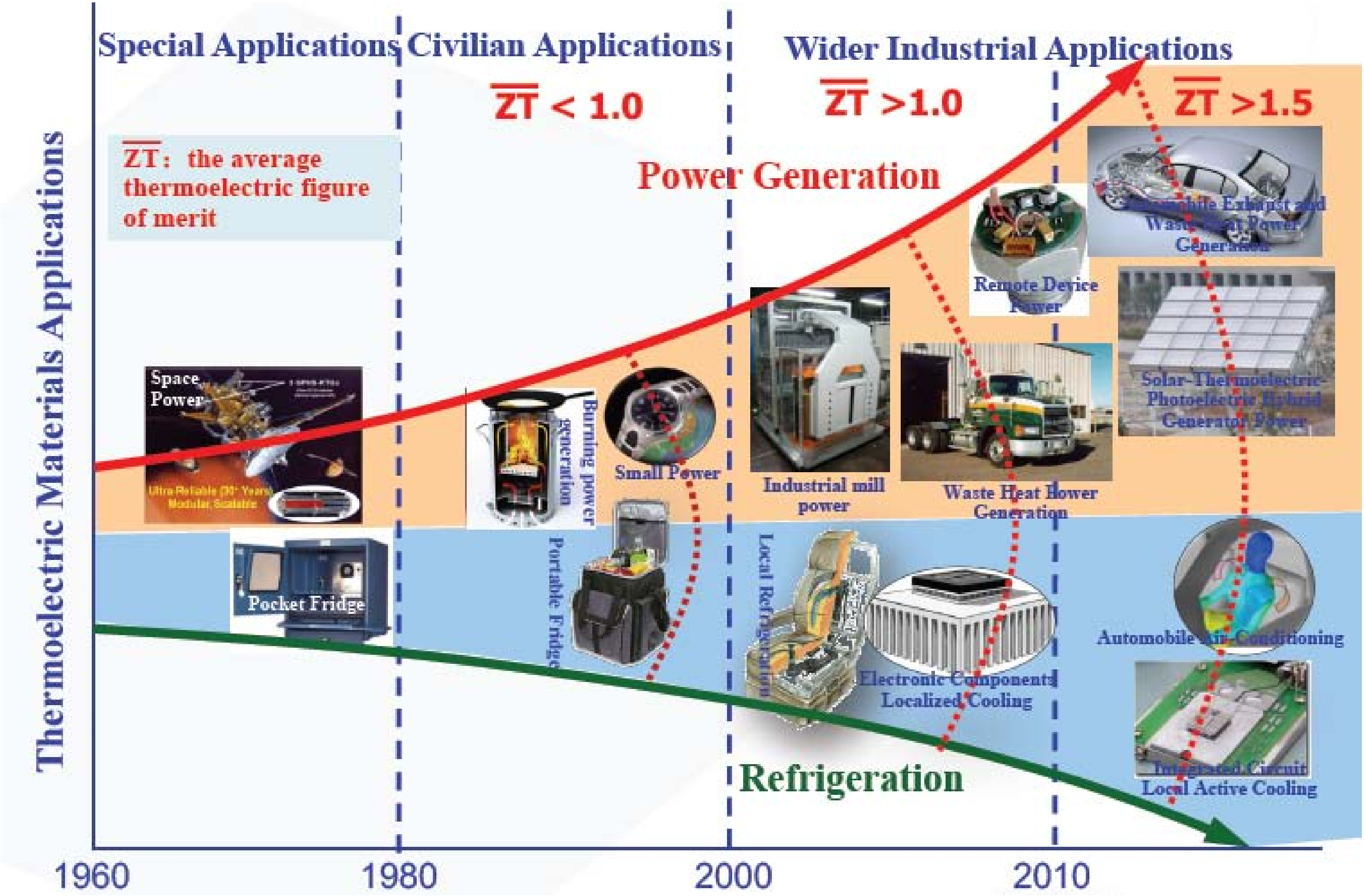


材料性能测试
(Materail testing)

器件性能测试
(Module testing)

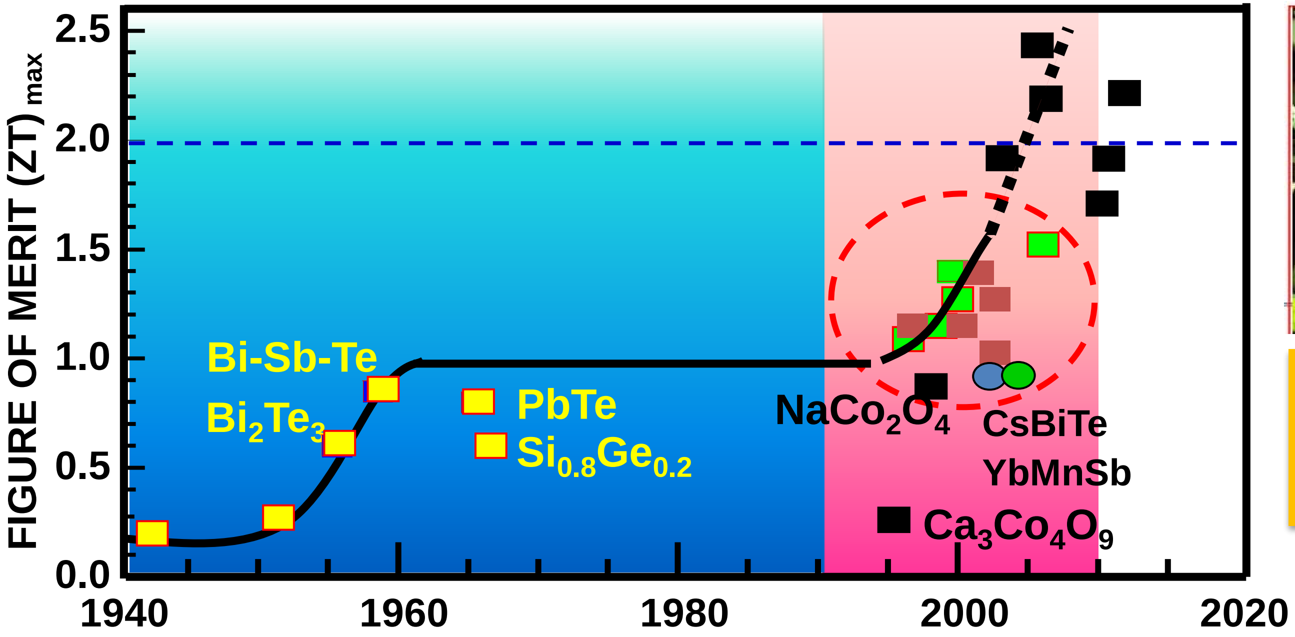
材料烧结
(Sintering)

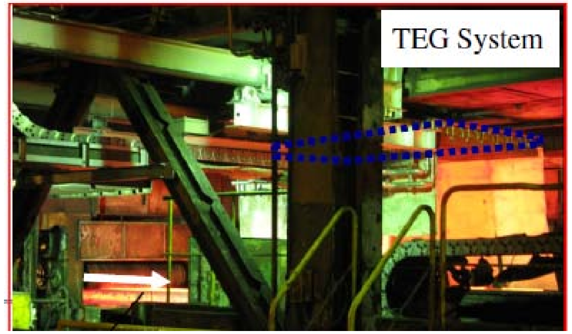
TE materials applications: Wider with *ZT* value improving



From Prof. Lidong Chen (SICCAS)

TE module: The bridge connect material and application

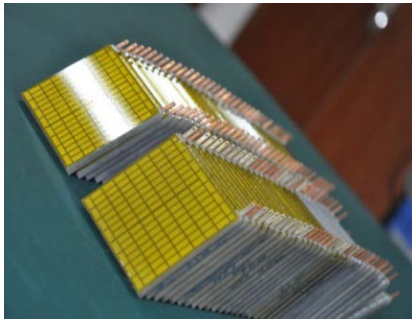




TEG system integration & industrial application

Developed TE transport theory & control strategy

High ZT value TE materials & new synthesis methods

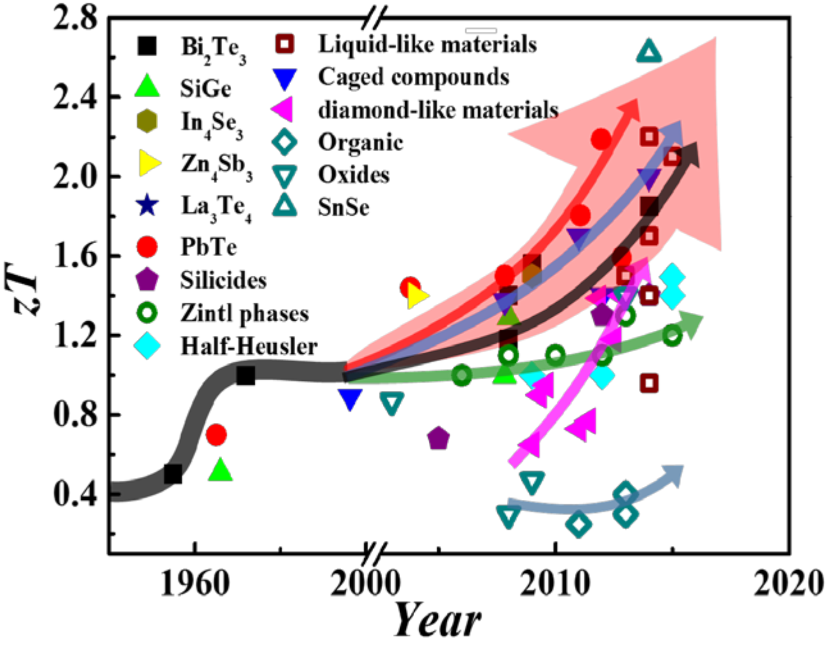


TE module design & fabrication

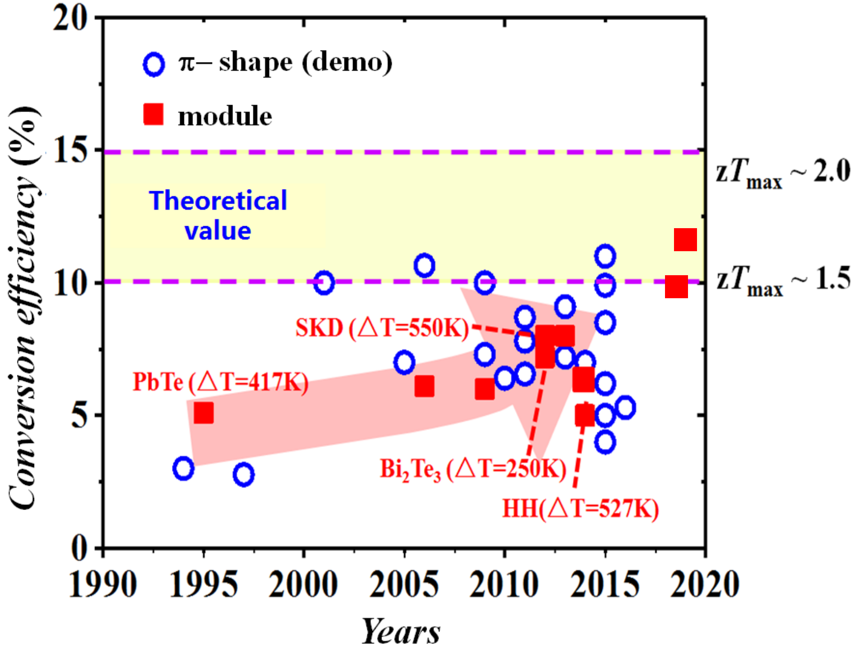
- **High efficiency**
- **High reliability**
- **Low cost**

Development history of TE materials & modules

- ◆ The continuous enhancement of TE materials performance enable the widespread practical applications. (Theoretical efficiency: 10 ~ 15%)
- ◆ Nevertheless, actual TE module development has remained stagnant with rather poor efficiencies. (Actual efficiency: 5%-8%)
- ◆ More than 70% researches focus on the validation of material performance (1 leg or π-shape).
- ◆ TE module full chain technology including "Design – Fabrication - Evaluation" **NOT** established.



TE materials



TE modules

Int. Mater. Rev., 2016(6):1-37.

Current status of TE modules for power generation

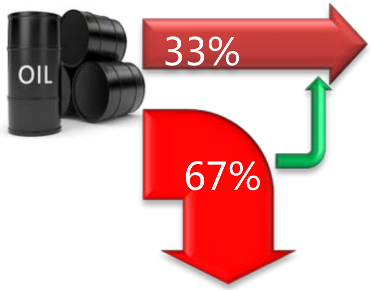
	Producer	TE materials	Efficiency	Source
Low-T.	KELK	Bi_2Te_3	7.2 %	JEMs-2014
	Marlow	Bi_2Te_3	5.03 %	Website
	Thermonamic	Bi_2Te_3	5 %	Website
	Hi-Z	Bi_2Te_3	4.5 %	Website
	Ferrotec	Bi_2Te_3	5 %	
	ITRI	Bi_2Te_3	5 %	
	SICCAS	Bi_2Te_3	6 %	
High-T.	Marlow	skutterudite	N/A	JEMs-2013
	Furukawa	skutterudite	8 %	JEMs-2012
	SICCAS	skutterudite	9.8 %	
	SICCAS	segmented	12 %	EES-2017
	AIST	segmented	11 %	
	Fraunhofer	Half-Heusler	5.4 %	
	GMZ	Half-Heusler	4.5 %	Website
	SICCAS	Half-Heusler	6.2 %	NC-2015

Low efficiency & high cost is the bottleneck of TEG in industrial application

Industrial application requirement:

- ◆ Conversion efficiency: >10%
- ◆ Service life: > 10 years

Energy saving & Emission reduction

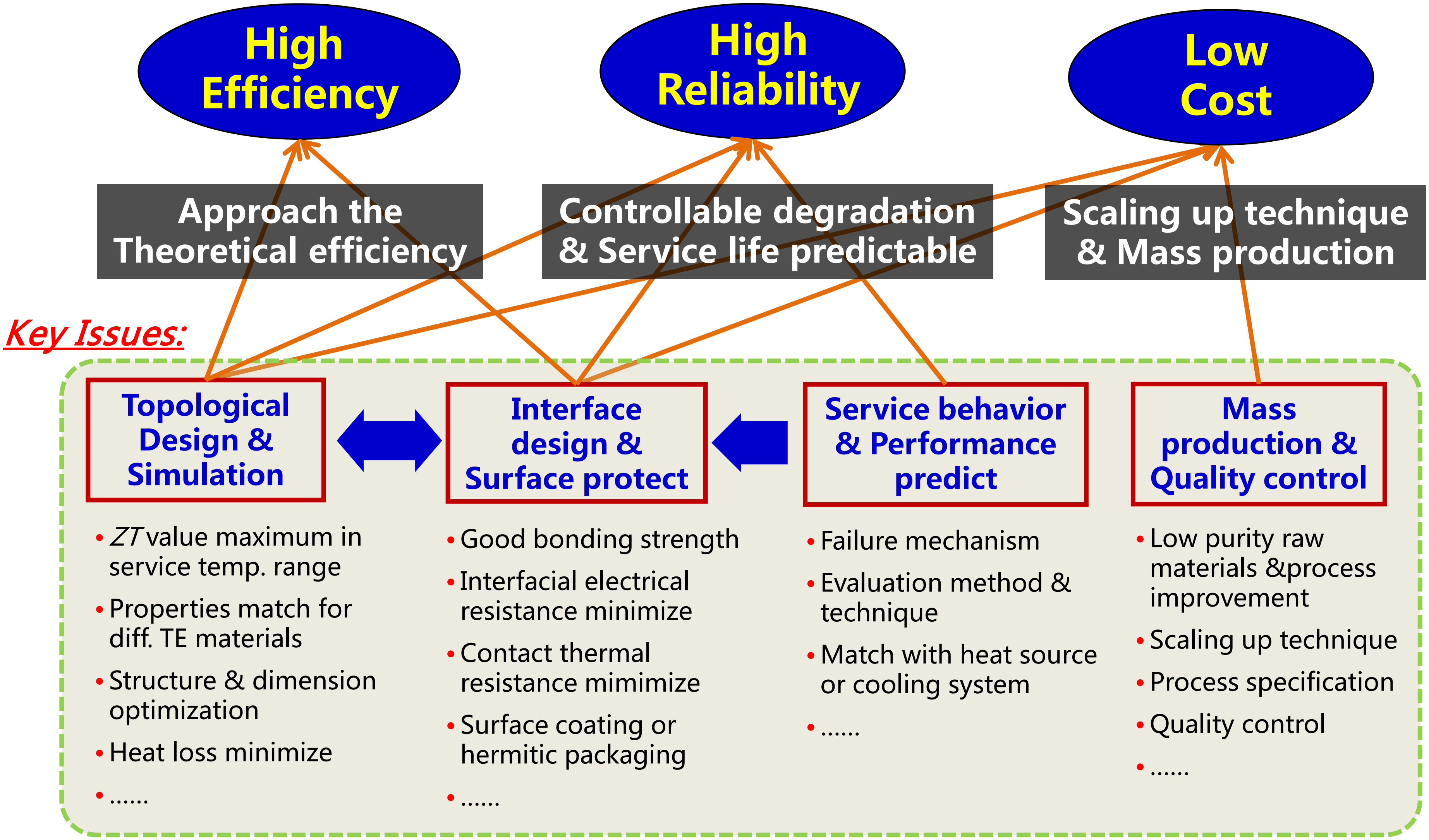


Exhaust heat recovery

Waste heat recovery

Air- conditioner

The targets of development of TE modules



High Efficiency

High Reliability

Low Cost

Approach the Theoretical efficiency

Controllable degradation & Service life predictable

Scaling up technique & Mass production

Key Issues:

Topological Design & Simulation

- *ZT* value maximum in service temp. range
- Properties match for diff. TE materials
- Structure & dimension optimization
- Heat loss minimize
- ……

Interface design & Surface protect

- Good bonding strength
- Interfacial electrical resistance minimize
- Contact thermal resistance mimimize
- Surface coating or hermitic packaging
- ……

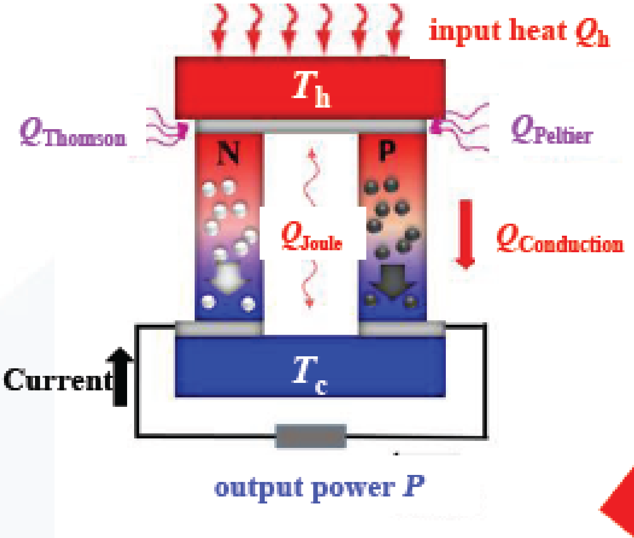
Service behavior & Performance predict

- Failure mechanism
- Evaluation method & technique
- Match with heat source or cooling system
- ……

Mass production & Quality control

- Low purity raw materials &process improvement
- Scaling up technique
- Process specification
- Quality control
- ……

The principle of TE device structure design



$$\eta_{theory} = \frac{P}{Q_h} = \frac{P}{Q_{Conduction} + Q_{Peltier} - Q_{Joule} - Q_{Thomson}} = \ldots\ldots$$

$$= \frac{T_h - T_c}{T_h} \cdot \frac{\frac{\boxed{m}}{m+1}}{1 + \boxed{\frac{KR}{S^2}} \cdot \frac{m+1}{T_h} - \frac{1}{2} \cdot \frac{T_h - T_c}{T_h} \cdot \frac{1}{m+1}}$$

$\boxed{m} \rightarrow \frac{\text{internal resistance}}{\text{load resistance}}$

working condition

internal factor

$$\eta = \frac{T_H - T_C}{T_H} \frac{\sqrt{1+ZT} - 1}{\sqrt{1+ZT} + T_C / T_H}$$

Simplified mode

➤To minimize *KR/S*²

$$\frac{KR}{S^2} = \frac{\kappa_n(T)\rho_n(T) + \kappa_n(T)\rho_p(T)\xi + \frac{\kappa_p(T)\rho_n(T)}{\xi} + \kappa_p(T)\rho_p(T)}{\left(\alpha_p(T) - \alpha_n(T)\right)^2}$$

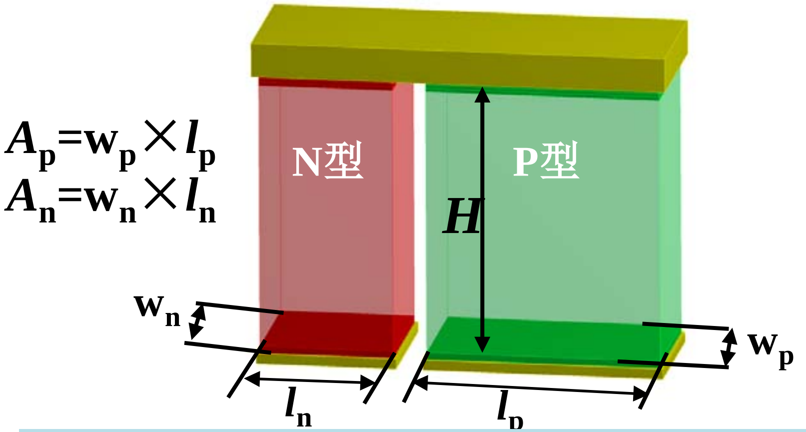
◆ *κ*, *ρ* *and* *α* are temperature-dependent

◆ Geometry factor *ξ*: related to the height & cross-sectional area of TE leg

◆ *ξ* can not be simply optimized when *κ*, *ρ* *and* *α* are temperature-dependent

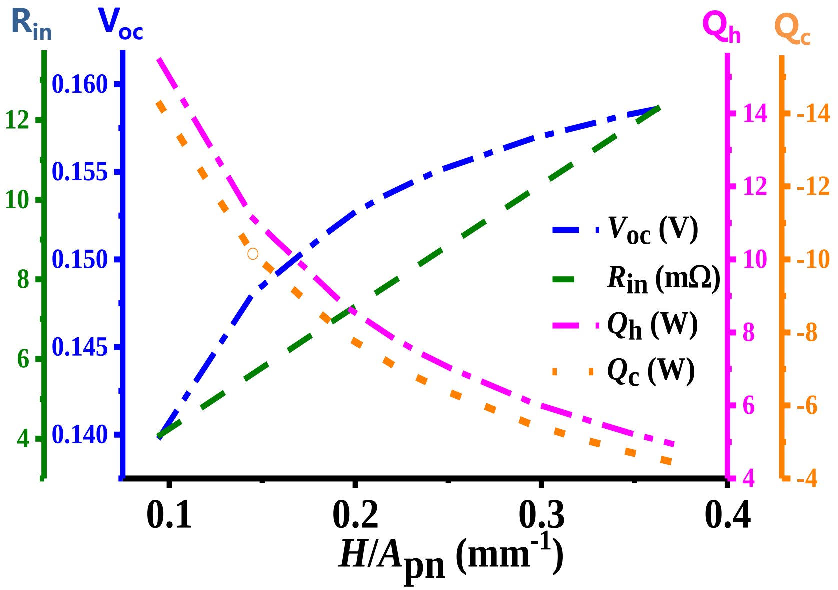
Topologic design: A useful tool to realize high efficiency

Structure & Dimension affect on Performance



$A_p = w_p \times l_p$

$A_n = w_n \times l_n$



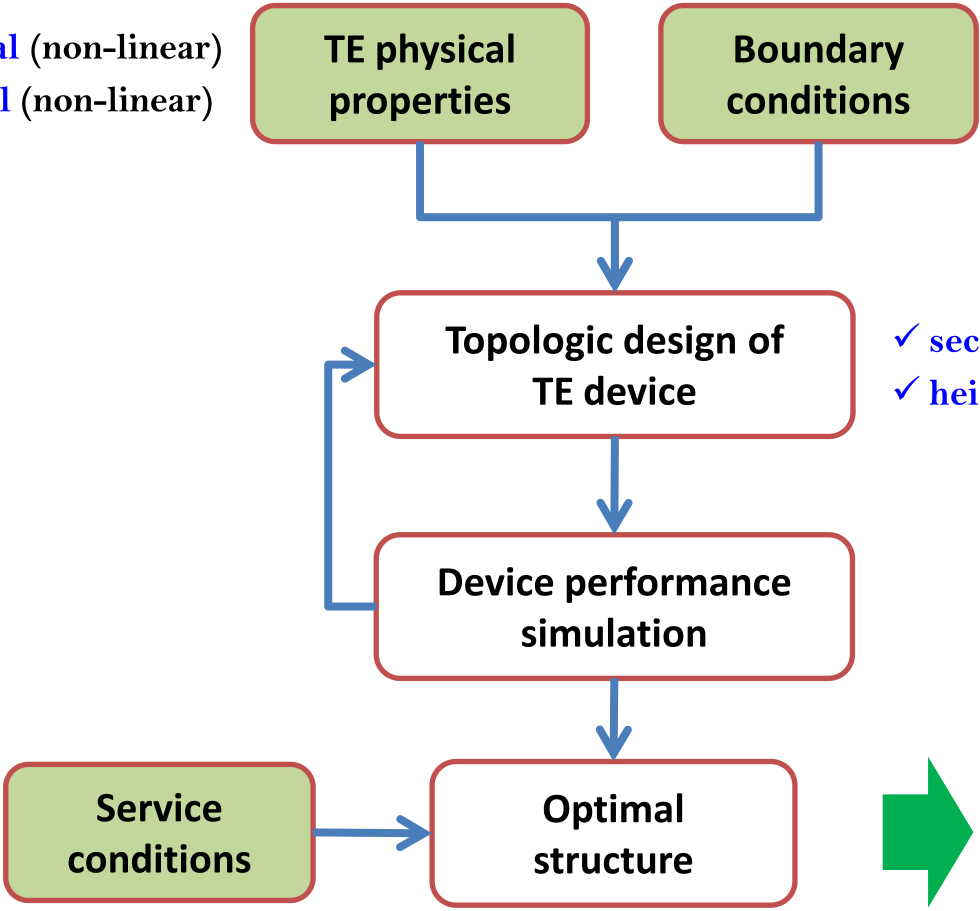
Structure factor: height, area, geometry, filling fraction

Traditional design approach:

Model	Principle	Solution method	Influence factors					Device conversion efficiency		
			effective △T	(α,σ,κ) *vs* T	structure factor	electric loss	thermal loss	predicted	measured	deviation
Energy balance	Global thermal equilibrium equation	algebra	✗	averaging	A_P/A_n	✗	✗	11.9%	8.4%	42%
One-dimensional	Local energy balance differential equation	analytical	✗	T-dependent	A_P/A_n	✗	✗	10.8%		29%

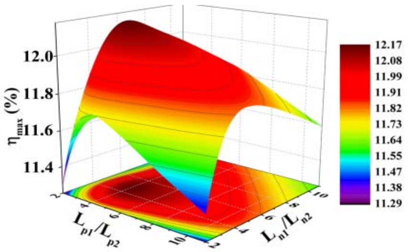
Design flow of device topologic optimization

✓ Electrical (non-linear)
✓ Thermal (non-linear)



✓ Contact resistance (affect P)
✓ Thermal Resistance (affect △T)
✓ Convection & Radiation (affect Q)

✓ section geometry & dimension
✓ height/area ratio



Goal

◆ "Structure – performance" relationship ("TE device genetic map")

◆ Full-parameter design based on service conditions

Three-dimensional Numerical modeling

Finite element method :

① Thermoelectric coupling constitutive equation :

$$\mathbf{q} = \alpha T \mathbf{J} - \lambda \nabla T$$

$$\mathbf{J} = -\sigma(\nabla V + \alpha \nabla T)$$

② Energy conservation equation & Charge continuity equation under **steady state**

$$\nabla \cdot \mathbf{q} = \dot{q}$$

$$\nabla \cdot \mathbf{J} = 0$$

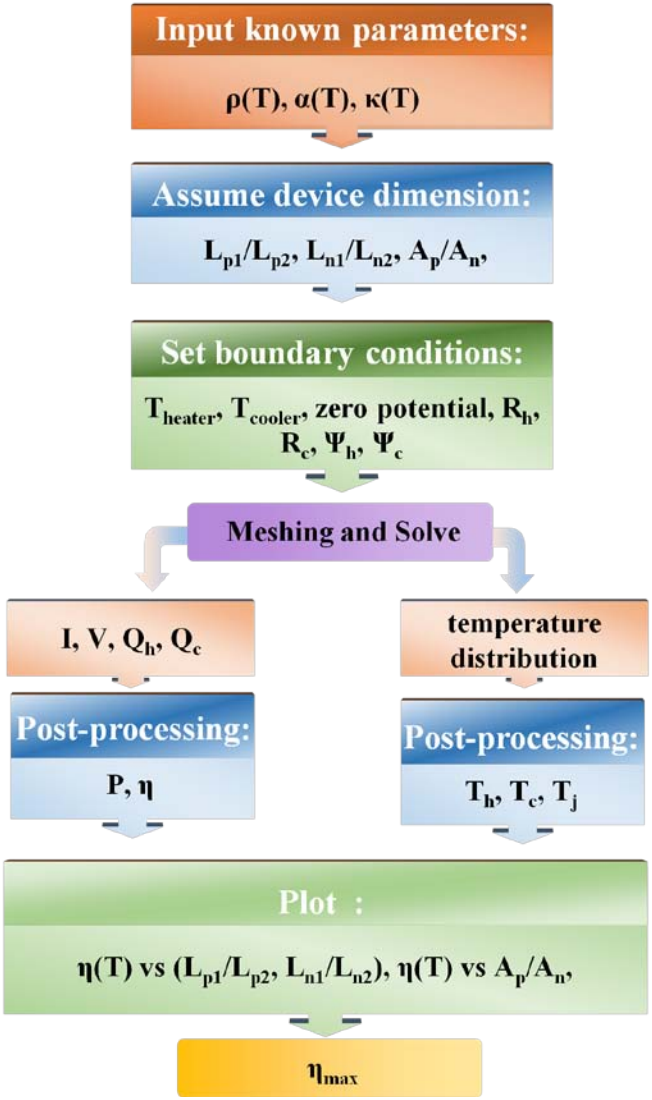
③ Governing equation describing the temperature, potential distribution & thermal-electrical coupling

$$\nabla(\lambda \nabla T) + \frac{J^2}{\sigma} - T\mathbf{J} \cdot \left[\left(\frac{\partial \alpha}{\partial T}\right) \nabla T + (\nabla \alpha)_T\right] = 0$$

$$\nabla \cdot ([\sigma]\nabla \varphi + [\sigma][\alpha]\nabla T) = 0$$

④ Obtain the finite element equation by variational principle

$$\begin{bmatrix} K^T & 0 \\ K^{T\varphi} & K^{\varphi} \end{bmatrix} \begin{Bmatrix} T_e \\ \varphi_e \end{Bmatrix} = \begin{Bmatrix} Q^L \\ I^L \end{Bmatrix}$$



Input known parameters:

$\rho(T)$, $\alpha(T)$, $\kappa(T)$

Assume device dimension:

L_{p1}/L_{p2}, L_{n1}/L_{n2}, A_p/A_n,

Set boundary conditions:

T_{heater}, T_{cooler}, zero potential, R_h, R_c, Ψ_h, Ψ_c

Meshing and Solve

I, V, Q_h, Q_c

Post-processing:

P, η

temperature distribution

Post-processing:

T_h, T_c, T_j

Plot :

$\eta(T)$ vs (L_{p1}/L_{p2}, L_{n1}/L_{n2}), $\eta(T)$ vs A_p/A_n,

η_{max}

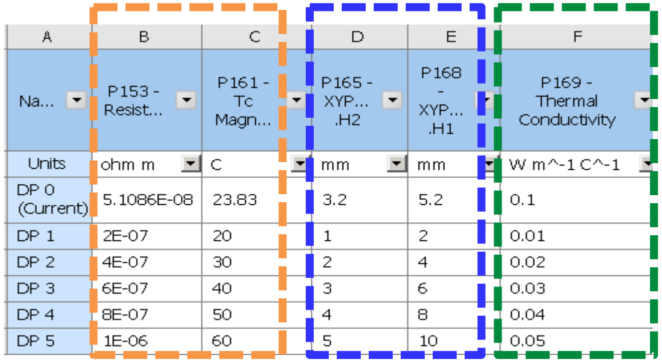
Flow chart

Three-dimensional Numerical modeling

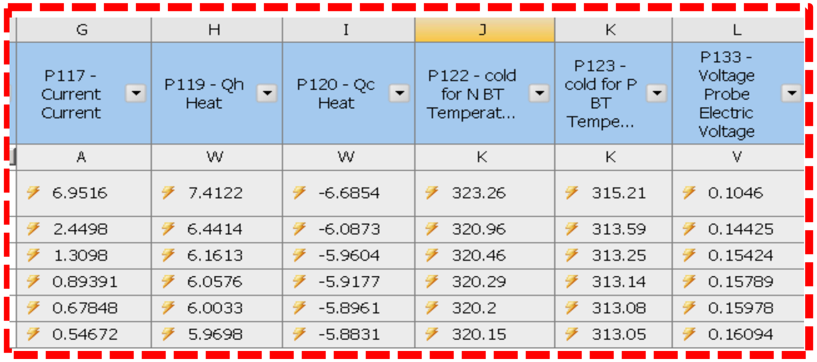
Input parameters

Output performance

Software interface



A	B	C	D	E	F
Na...	P153 - Resist...	P161 - Tc Magn...	P165 - XYP... .H2	P168 - XYP... .H1	P169 - Thermal Conductivity
Units	ohm m	C	mm	mm	W m^-1 C^-1
DP 0 (Current)	5.1086E-08	23.83	3.2	5.2	0.1
DP 1	2E-07	20	1	2	0.01
DP 2	4E-07	30	2	4	0.02
DP 3	6E-07	40	3	6	0.03
DP 4	8E-07	50	4	8	0.04
DP 5	1E-06	60	5	10	0.05



G	H	I	J	K	L
P117 - Current Current	P119 - Qh Heat	P120 - Qc Heat	P122 - cold for N BT Temperat...	P123 - cold for P BT Tempe...	P133 - Voltage Probe Electric Voltage
A	W	W	K	K	V
⚡ 6.9516	⚡ 7.4122	⚡ -6.6854	⚡ 323.26	⚡ 315.21	⚡ 0.1046
⚡ 2.4498	⚡ 6.4414	⚡ -6.0873	⚡ 320.96	⚡ 313.59	⚡ 0.14425
⚡ 1.3098	⚡ 6.1613	⚡ -5.9604	⚡ 320.46	⚡ 313.25	⚡ 0.15424
⚡ 0.89391	⚡ 6.0576	⚡ -5.9177	⚡ 320.29	⚡ 313.14	⚡ 0.15789
⚡ 0.67848	⚡ 6.0033	⚡ -5.8961	⚡ 320.2	⚡ 313.08	⚡ 0.15978
⚡ 0.54672	⚡ 5.9698	⚡ -5.8831	⚡ 320.15	⚡ 313.05	⚡ 0.16094

TE materials properties

σ
κ
S

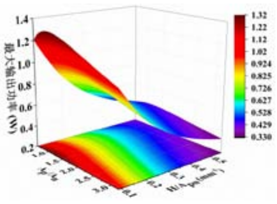
Structure factors

- H
- L_n
- L_p
- W_n
- W_p
- Gap
- ……

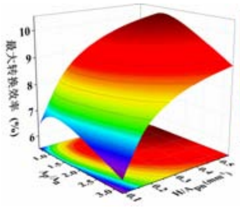
Boundary conditions

- T_h
- T_c
- R_{load}
- $\rho_{interface}$
- $W_{contact}$
- Convection
- Radiation …

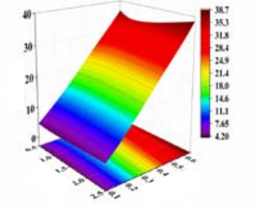
P vs. Structure



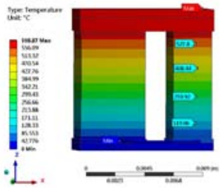
η vs. Structure



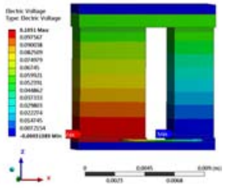
Rin vs. structure



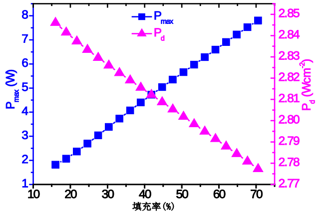
Temp. distribution



Voltage distribution

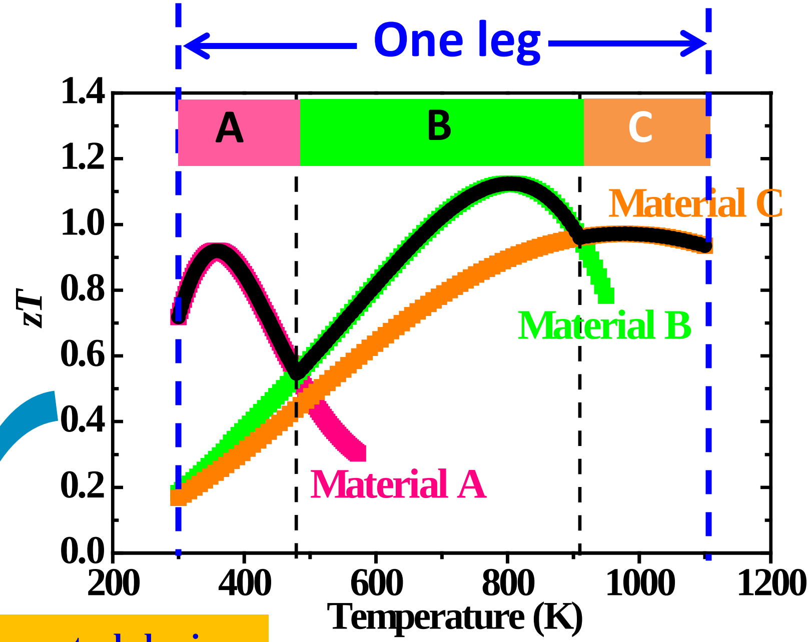


P/PD vs. filling fraction



✓ Relationship of "Performance – Structure" could be found out

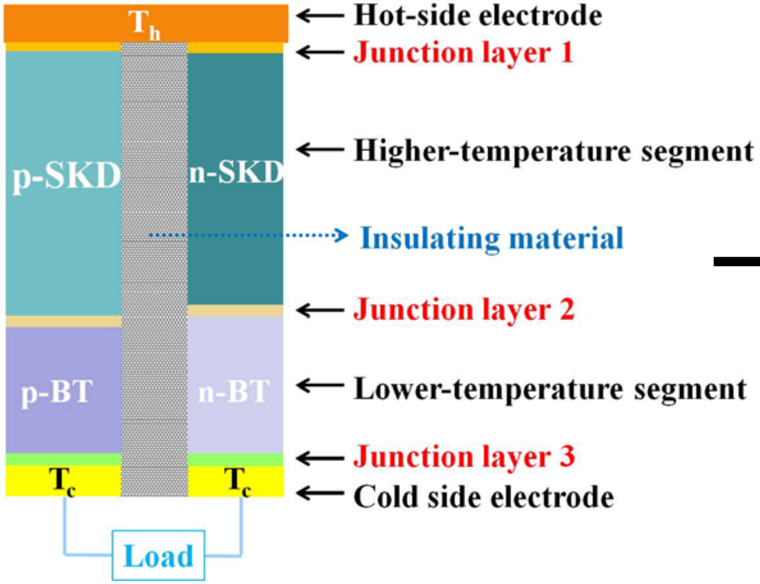
Segment: A strategy to realize high efficiency



$$\eta_{\max} = \frac{T_H - T_C}{T_H}\,\frac{\sqrt{1+zT}-1}{\sqrt{1+zT}+T_C/T_H}$$

- A single TE material shows peak *zT* under certain temperature range;
- To build segmented TE device can make each material operate in the temperature range where its efficiency is maximized;
- Enhance *zT & ΔT* simultaneously.

Segmented device



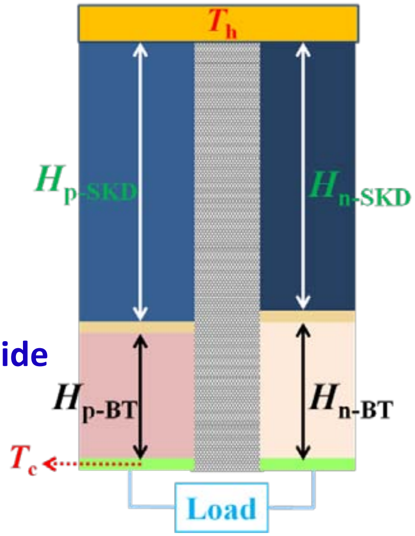
Increased interfaces lead to more energy losses

Complex structure brings more difficulty for topological design

Leg geometry optimization of segmented module

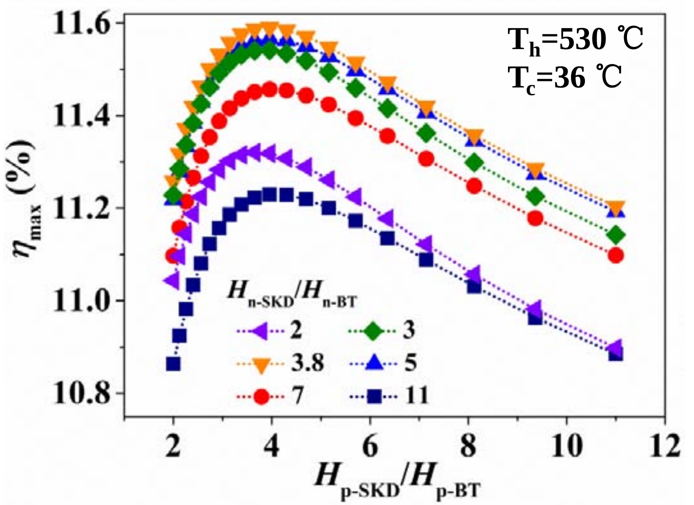
Skutterudite

Bismuth telluride

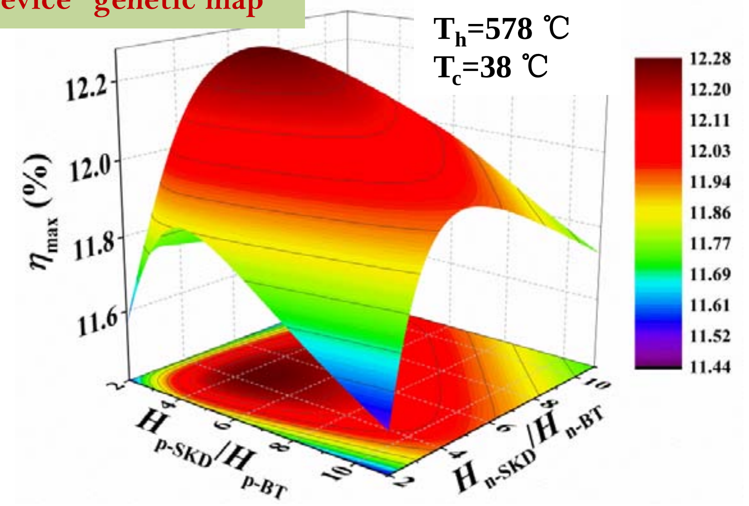


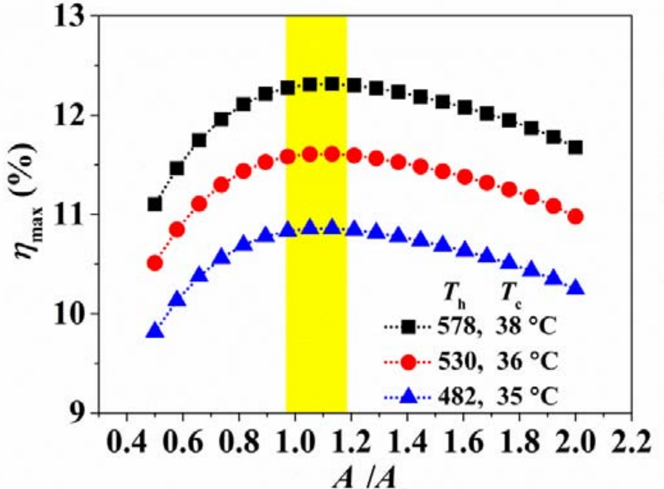
$H_{n\text{-SKD}}/H_{n\text{-BT}}$ is fixed

p- and *n*-type legs are mutually constrained due to the reallocation of the heat flow.



TE device "genetic map"





Synchronously optimize two legs for optimum ratio

Optimize cross-sectional area ratio

Multi-parameter optimization of segmented module

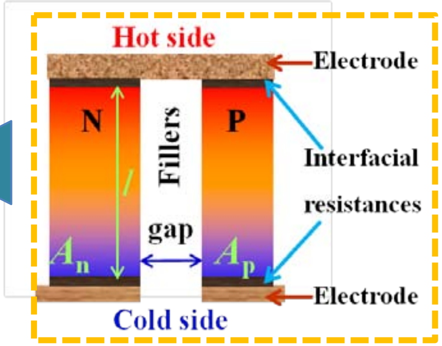
Thermal loss analysis

Additional boundary conditions:

gap, κ_F

Plot :

$\eta(T)$ vs gap&κ_F



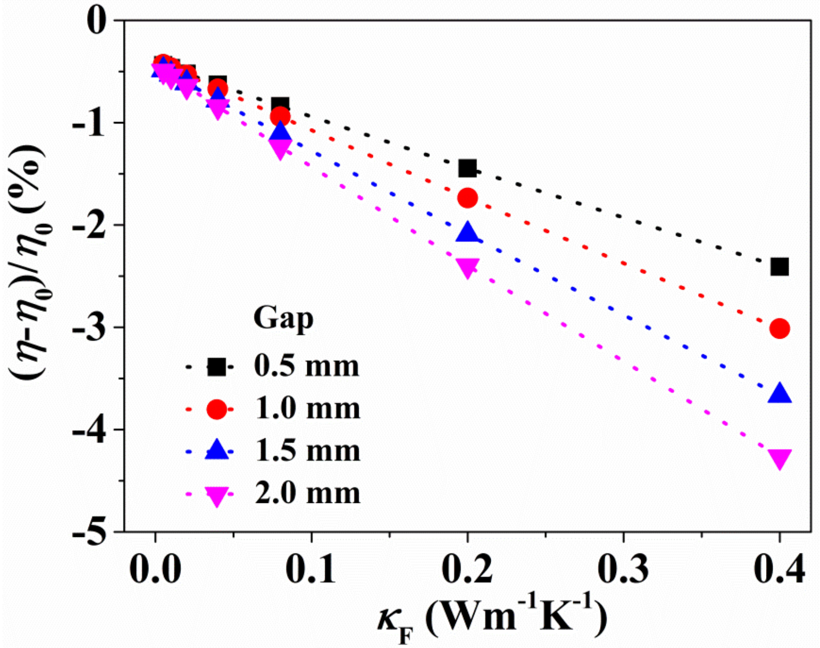
Electrical loss analysis

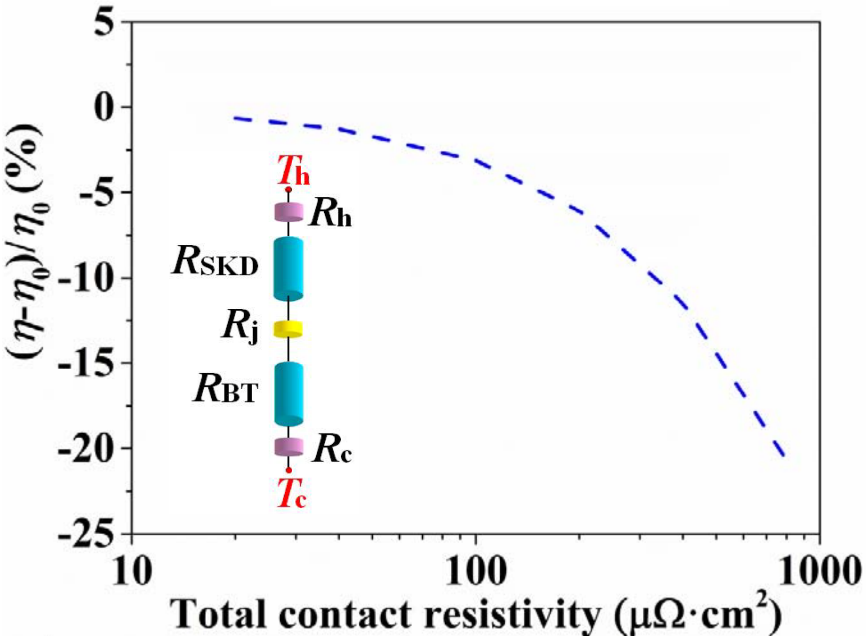
Additional boundary conditions:

ρ_c

Plot :

$\eta(T)$ vs ρ_c



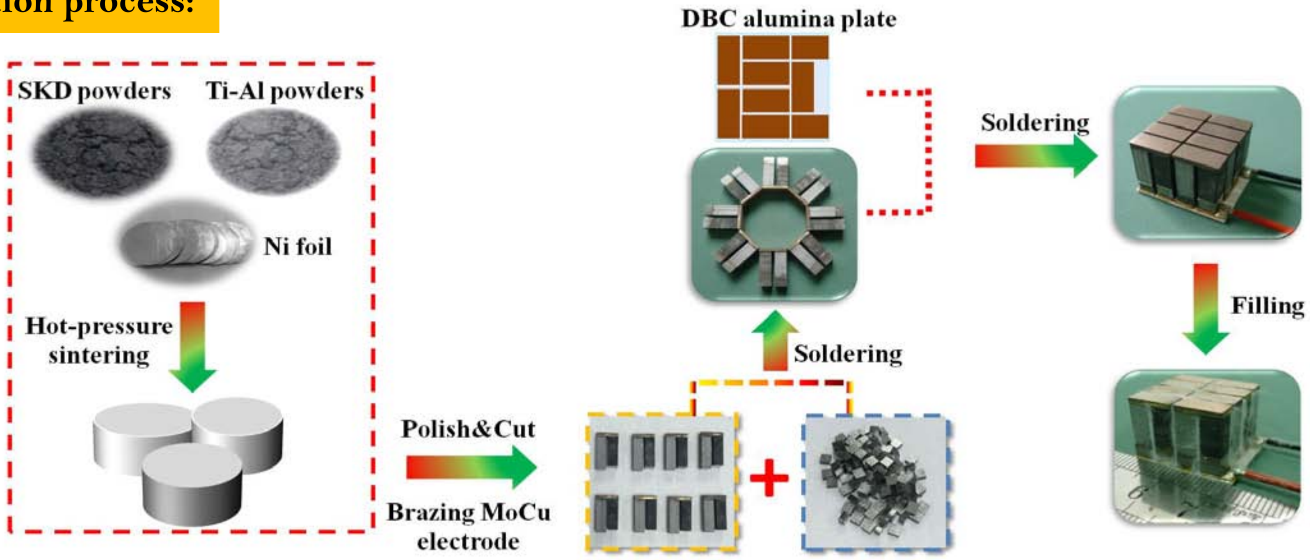


- *Efficiency loss increases with the increasing thermal conductivity of fillers & gap distance*

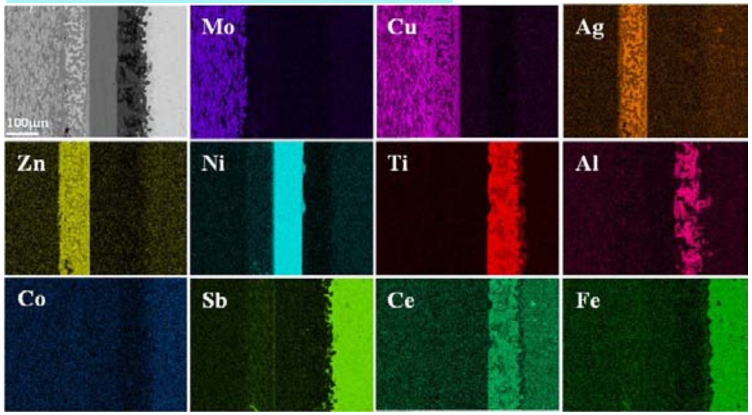
- *The increased total ρ_c leads to more losses in the efficiency*

Segmented BT/SKD module integration

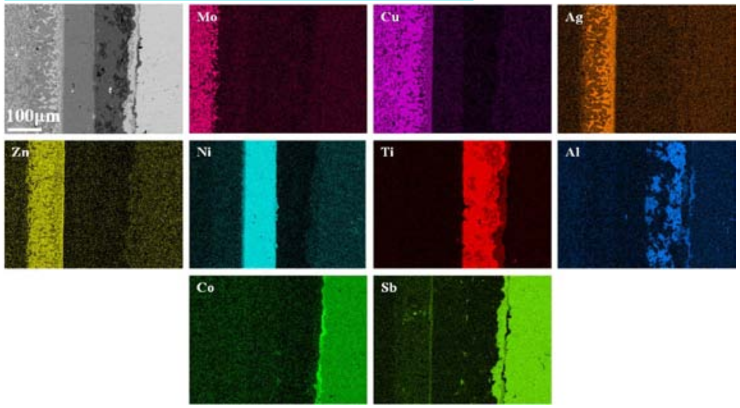
Overall fabrication process:



p-SKD/MoCu junction

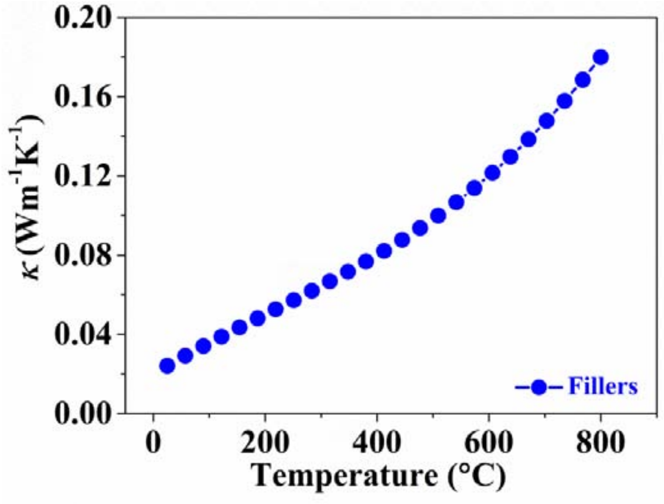


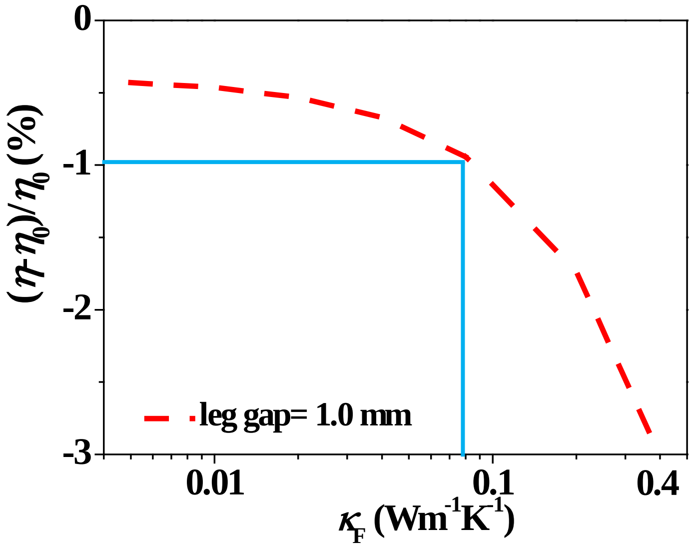
n-SKD/MoCu junction



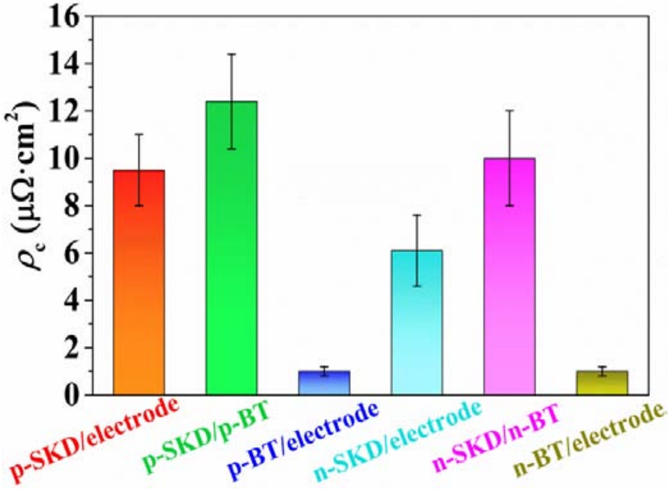
Microstructure and composition shows excellent interface bonding, no crack & no diffusion

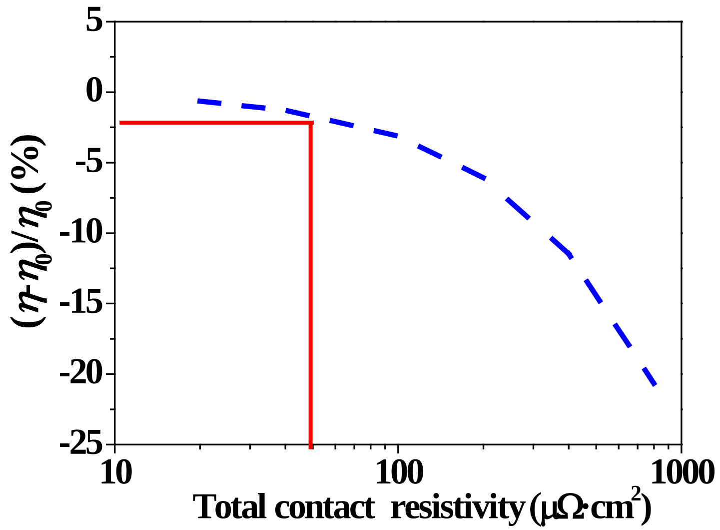
Energy loss characterization





- Glass fibers with the average κ of 0.09 $\mathrm{Wm^{-1}K^{-1}}$ cause a merely 1 % reduction in η_{max}

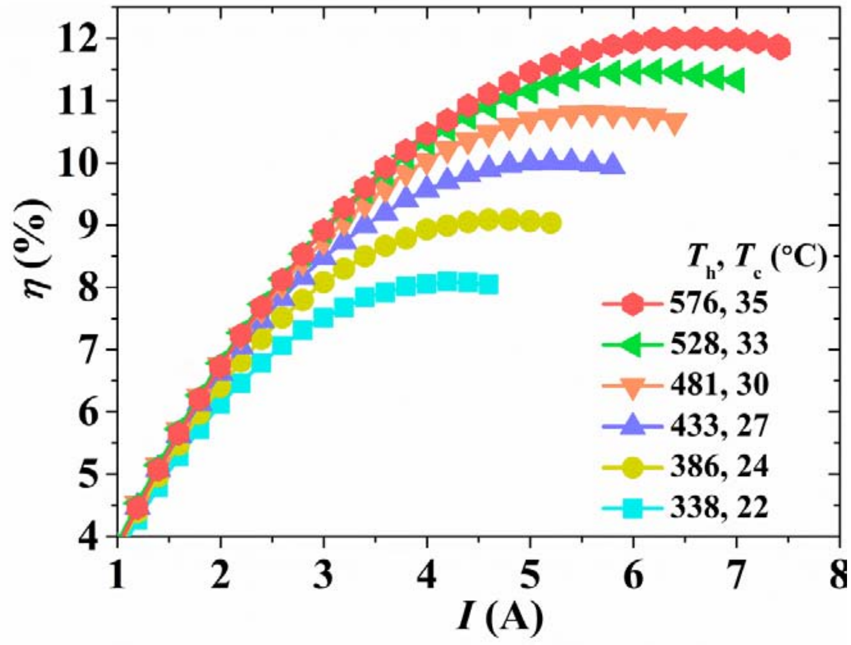


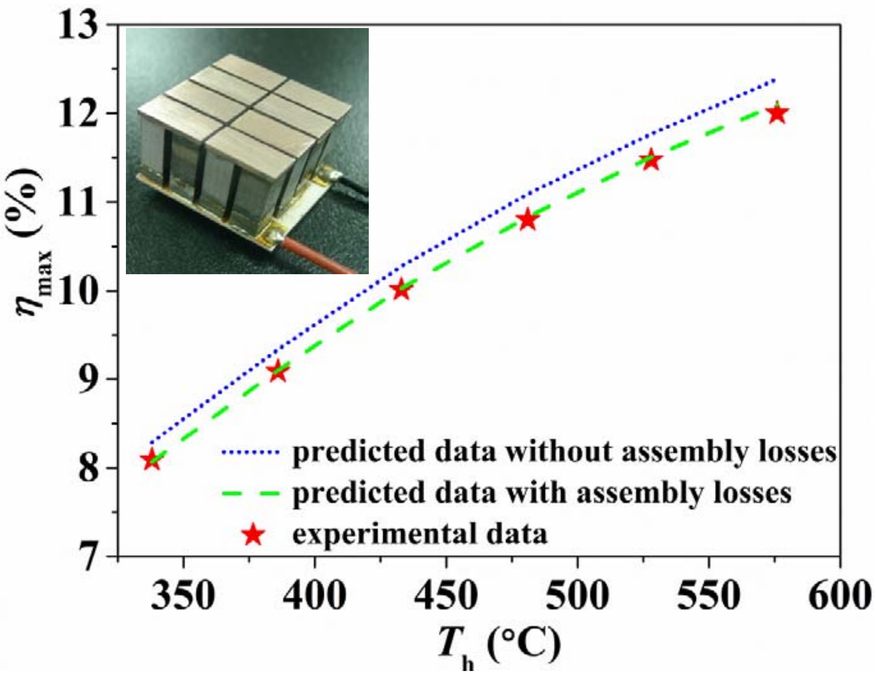


- Composition of each interface was optimized. ρ_c ~40 μΩ·cm² causes <2% loss in η_{max}

Characterization of the segmented BT/SKD module

Thermoelectric conversion efficiency:

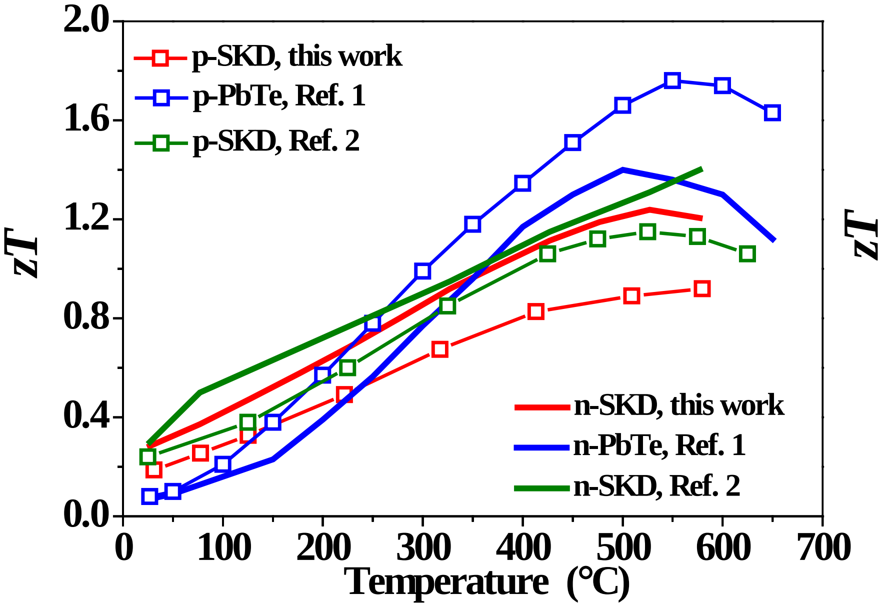


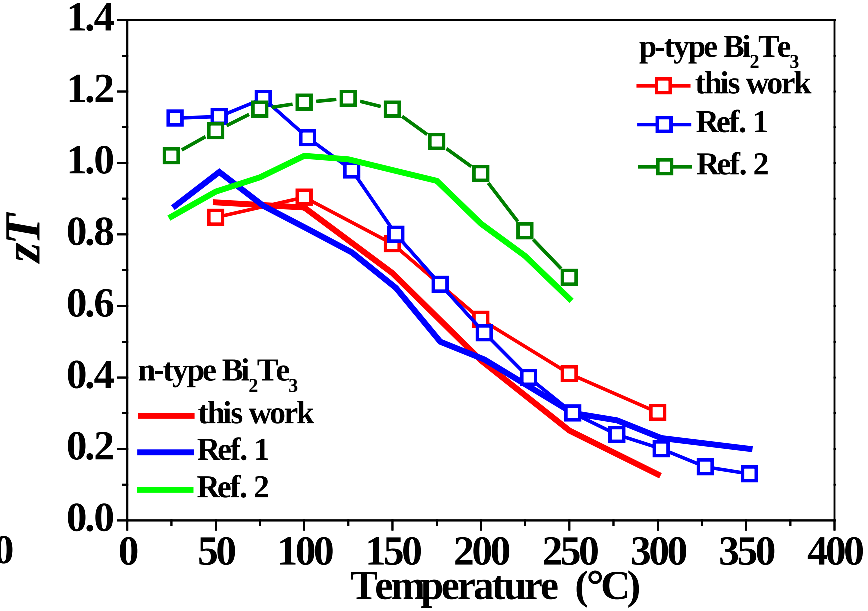


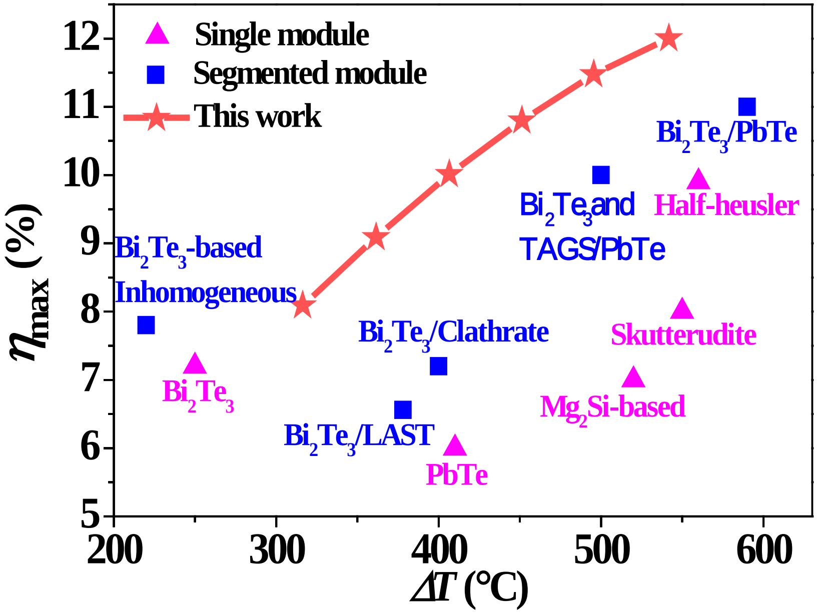
- ΔT=316 ℃, η_{max}= 8%; ΔT=541 ℃, η_{max}= 12%;
- Experimental data agree well with the simulated results;
- η_{max} reaches ~97% of the ideal efficiency based on the TE materials themselves.

Energy Environ. Sci., 2017, 10, 956-963.

Comparison with the literatures



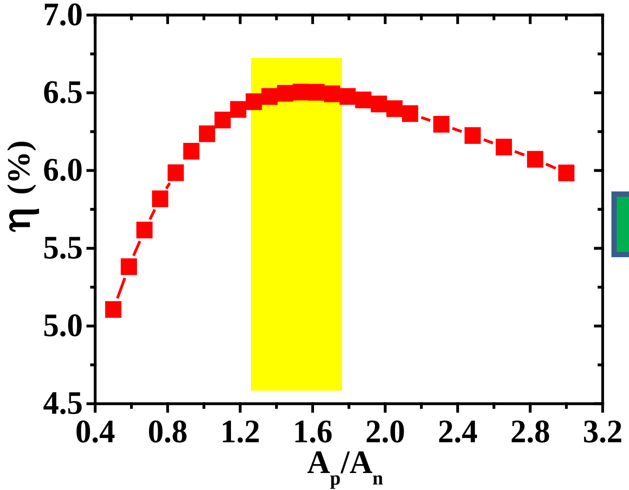


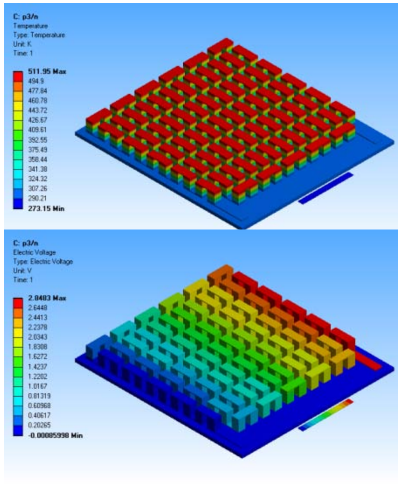


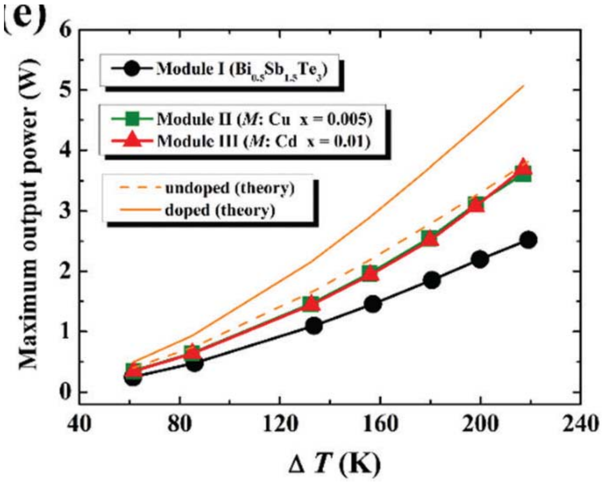
- Using Low zT value TE materials (scaling up fabricated or commercial) can achieve a high η_{max} TE modules, due to the rational structure design and minimum energy losses.

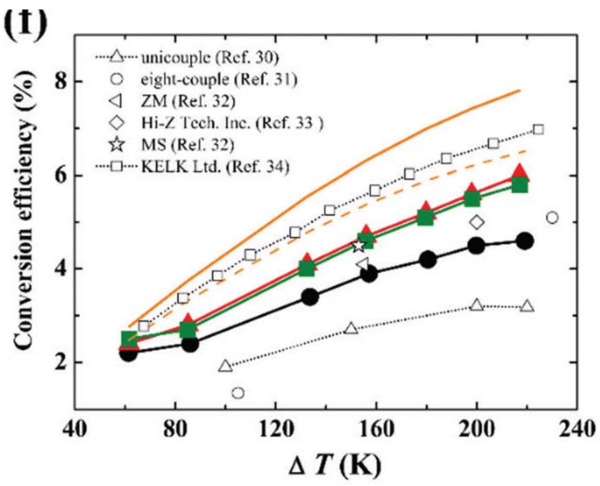
Topologic design application: single-stage BT module

Structure optimize | **Performance simulation** | **Module integration**









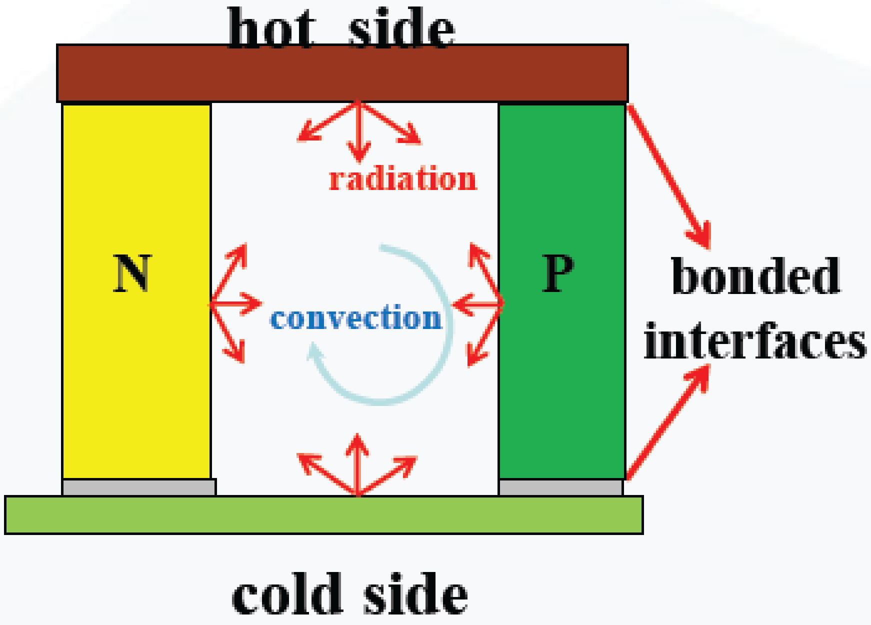
Combining the high ZT p-type Bi_2Te_3-based materials and optimal structure, the module's efficiency reached 6.0% @ ΔT= 217 K, which superior to commercial modules.

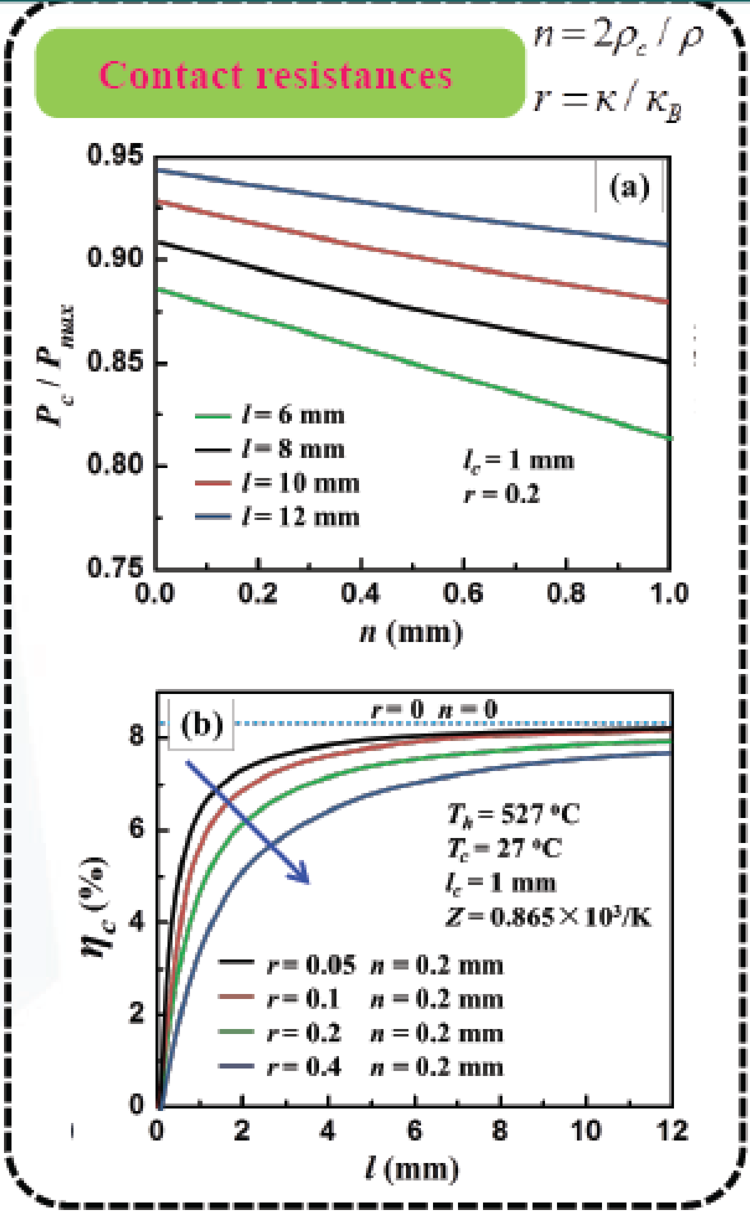
Thanks to *Dr.* Hsu-Shen Chu from ITRI for module integration.

Energy Environ. Sci., 2016, 9, 3120-3127.

Device assembly factor: Energy loss

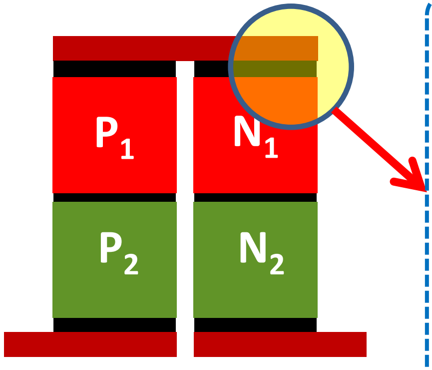
Device assembly loss： electrical & thermal

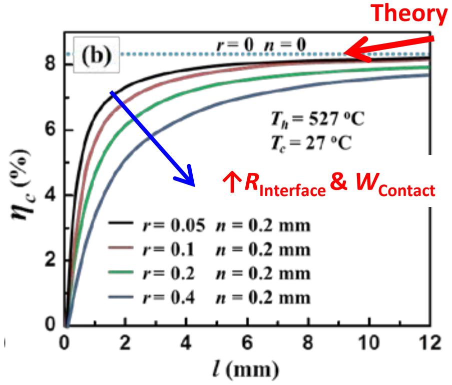




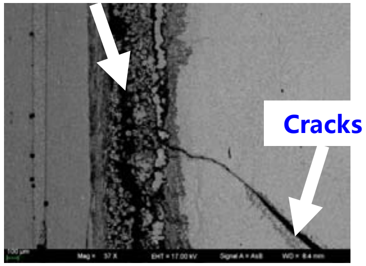
- ◆ Electrical and thermal resistances at various interfaces
- ◆ Thermal radiation and/or convection through the element gaps
- ➢ The actual output performances below the theoretical values

Interface : Key issue of high efficiency & high reliability

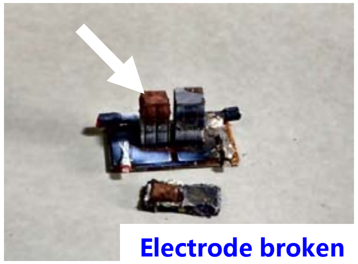




Interfacial diffusion



Oxidation



SKD module：500°C×10 days in air

Working Conditions:

- ➢ High Temp.
- ➢ Large Temp. Diff.
- ➢ Oxide & humidity
- ➢ Corrosively
- ➢ Thermal shock
- ➢ Vibration

Interface

Electrical
- ✓ High electrical conductivity
- ✓ Low interfacial resistivity

Thermal
- ✓ High thermal conductivity
- ✓ Low contact resistivity

Mechanical
- ✓ Good bonding strength
- ✓ Minimize residual stress

Chemical
- ✓ Stable at high temperature
- ✓ Block interfacial diffusion

- ◆ Complex working conditions caused a complex interface behavior.
- ◆ Most of structural and functional failures of TE modules are caused by the evolution of the interfaces.
- ◆ A high reliable interface with minimized energy loss is the key issue to realize a TE module with both high efficiency and high reliability.

Adv. Eng. Mater., 2016,18, 194

Interfacial physical properties: Influent performance

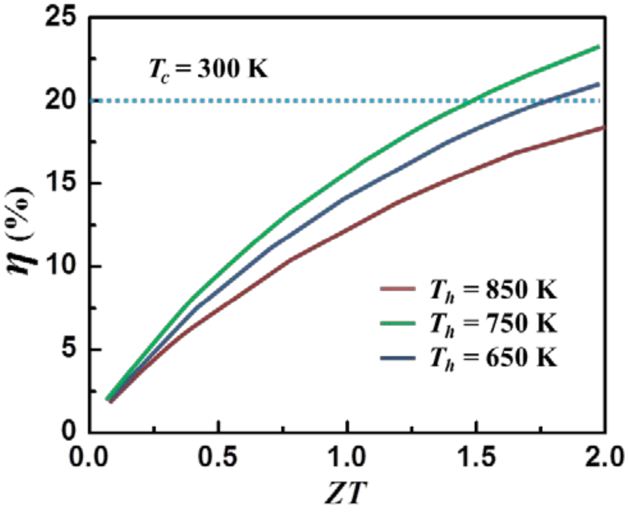
Output power & efficiency:

$$P=\frac{S^2(T_h-T_c)^2}{4R}\cdot\frac{1}{(1+n/l)(1+2rw)^2}$$

$$\eta=\frac{T_h-T_c}{T_h}\left\{(1+2rw)^2\left[2-\frac{1}{2}\left(\frac{T_h-T_c}{T_h}\right)+\frac{4}{ZT_h}\left(\frac{1+n/l}{1+2rw}\right)\right]\right\}^{-1}$$

Efficiency vs. *ZT*

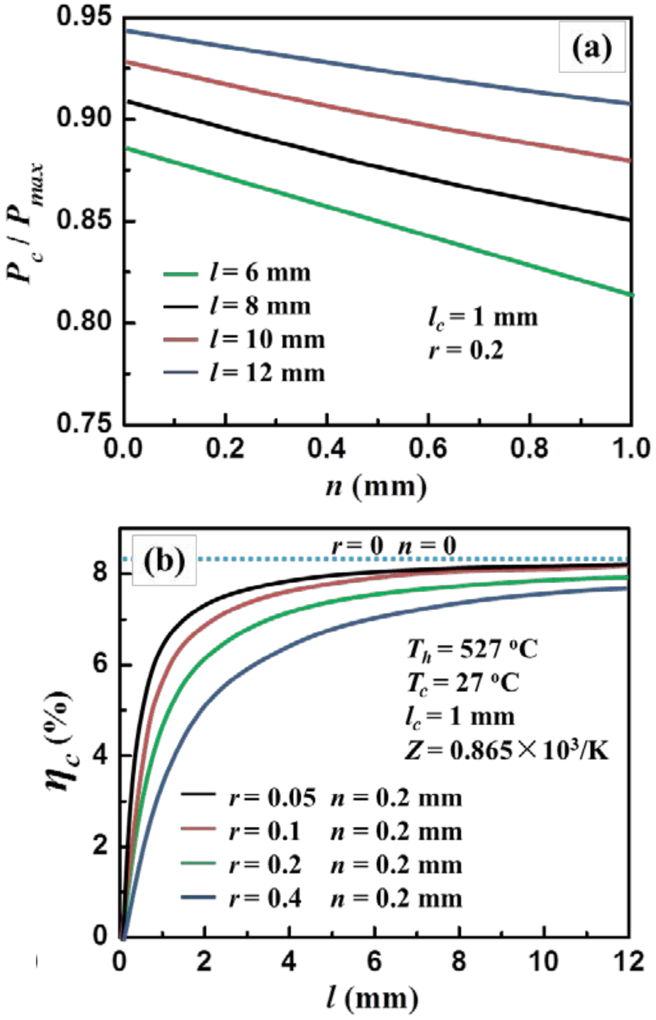
$$ZT=\frac{S^2\sigma}{\kappa}T$$



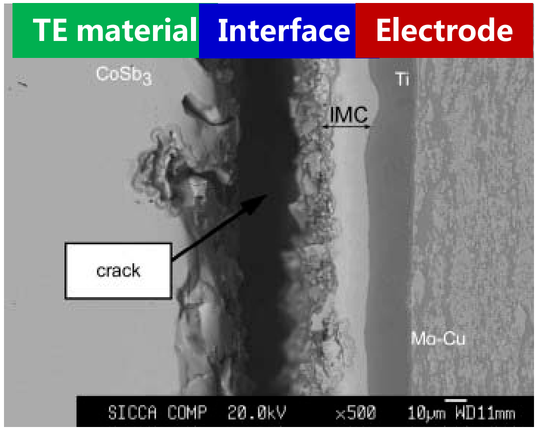
Interface influence

$n=2\rho_c/\rho$

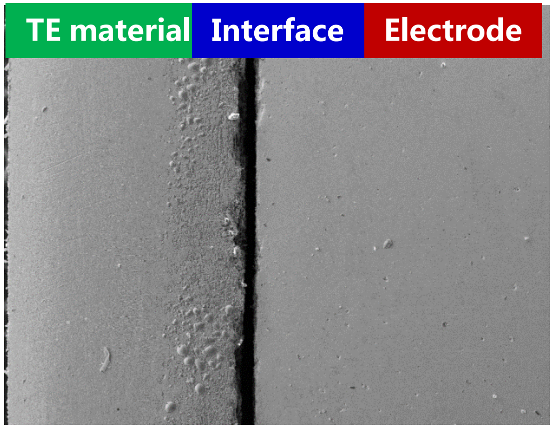
$r=\kappa/\kappa_B$



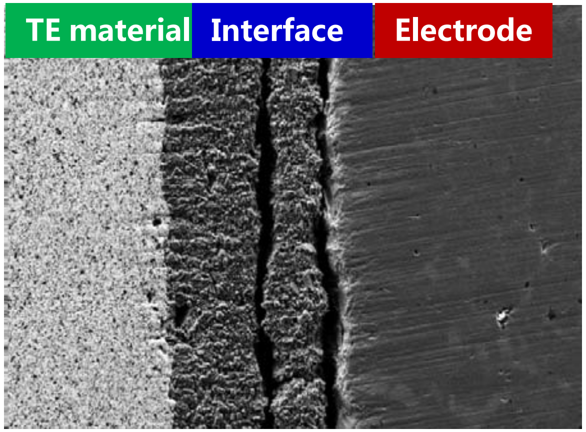
Interfacial chemical properties: Influent reliability



Skutterudite module
Failure position：IMC



Half-Huesler module
Failure position：IMC



SiGe module
Failure position：IMC

Structural failure:

- **Position:** Interface of intermetallic compounds.
- **Reasons:** Interfacial diffusion, chemical reaction, oxidation, sublimate, ……

Interface design must combining contact properties (strength, conductivity) & diffusion barrier.

Principle of choosing diffusion barrier(buffer) layer

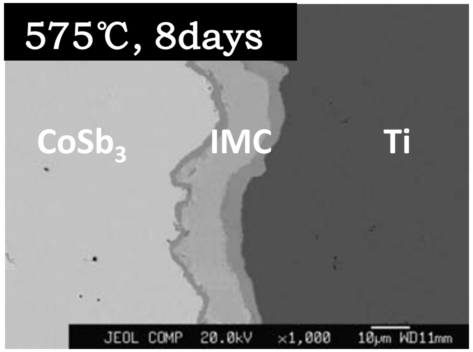
Qualitative principle:

- Chemical : Reaction possible → Improve the bonding strength improvement
  Low diffusion speed → Improve the interface reliability
- Thermal & electrical : 10^1~10^2 higher than TE materials → Reduce energy loss
- CTE: close to TE materials → Reduce the stress
- Others: Melting point >> Working temperature

Candidate		Diffusion properties	MP/°C	σ (10^6 S/m)	κ (W/cm.K)	CTE (ppm)
Matrix	**SKD**	——	873	0.06~0.2	0.02~0.03	9~11
Buffer layer	**Ti**	?	1700	2.4	2.2	8.4
	Zr	?	1852	2.4	2.3	5.8
	Hf	?	2227	2.8	2.3	5.9
	Nb	?	2467	8	5.4	7.1
	Ta	?	3014	8	5.8	6.6
	Cr	?	1857	7.9	9.4	6.2
	Mo	?	2617	19	13.8	5.4

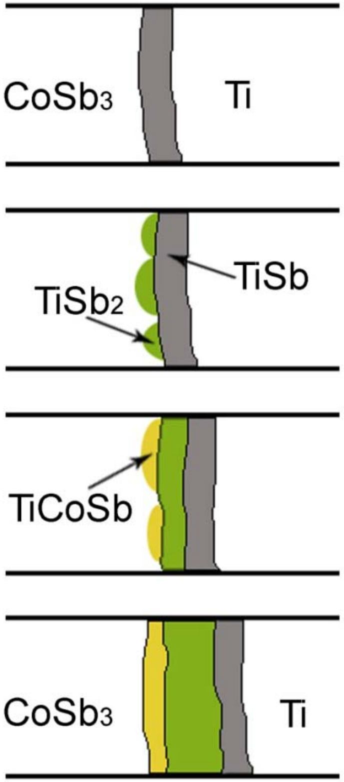
***Interfacial diffusion properties must be obtained by experiment.**

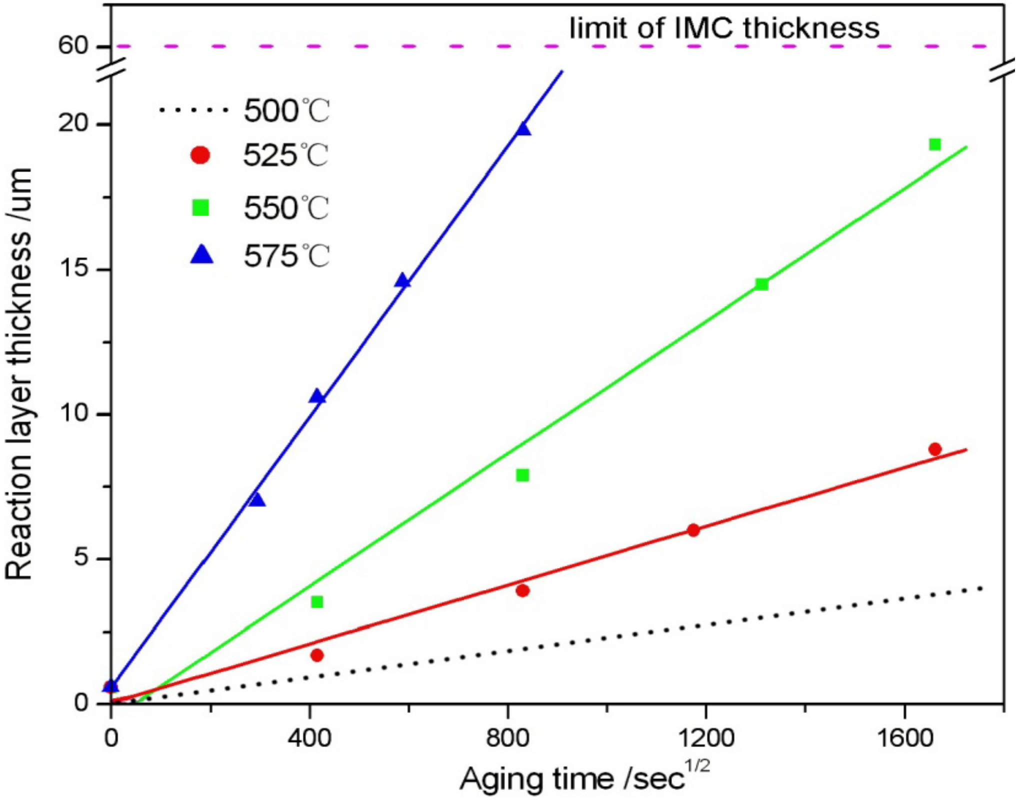
Degradation mechanism: Inter-diffusion at interface



CoSb$_3$/TiSb/Ti → CoSb$_3$/***TiCoSb***/***TiSb$_2$***/TiSb/Ti

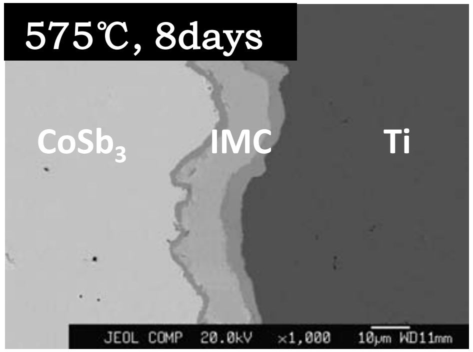
The interfacial microstructure evolution is mainly caused by the Sb diffusion to form intermetallic compound layer and this layer causes the degradation and final destruction.

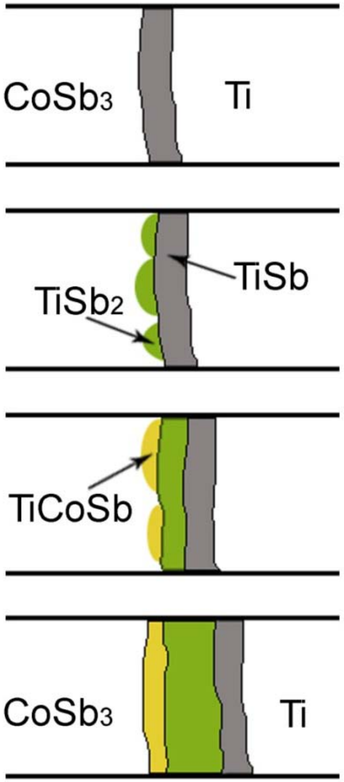




J. Alloys Compd. **2009**, 477, 425

Optimization of interface microstructure





The high activity of Ti interlayer benefits the sintering/bonding between SKD and MoCu electrode, but also causes the degradation due to the inter-diffusion at high temperature.

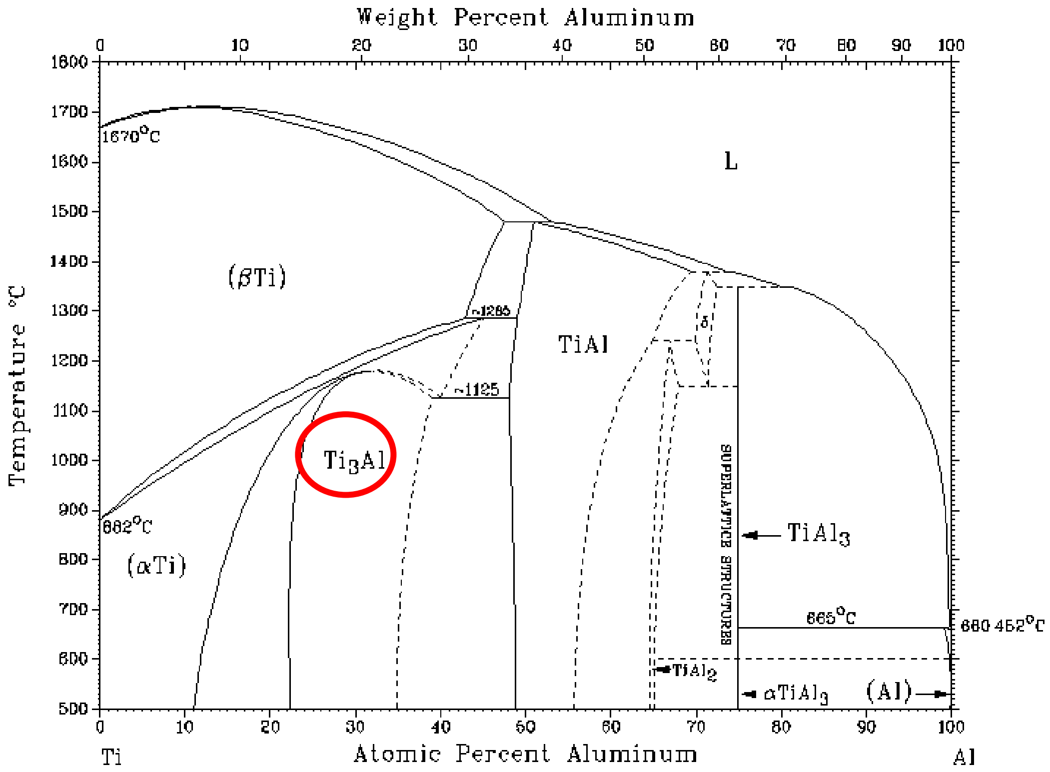
Optimization of interfacial microstructure and bonding technique:

Keep low interfacial R (T&E) → efficiency

Depress high-T inter-diffusion → reliability

Tune high temperature activity (diffusion and reaction) of interlayer: find new Ti-based alloy as interlayer instead of Ti.

Ti-Al alloy: Balance high-T stability & bonding activity

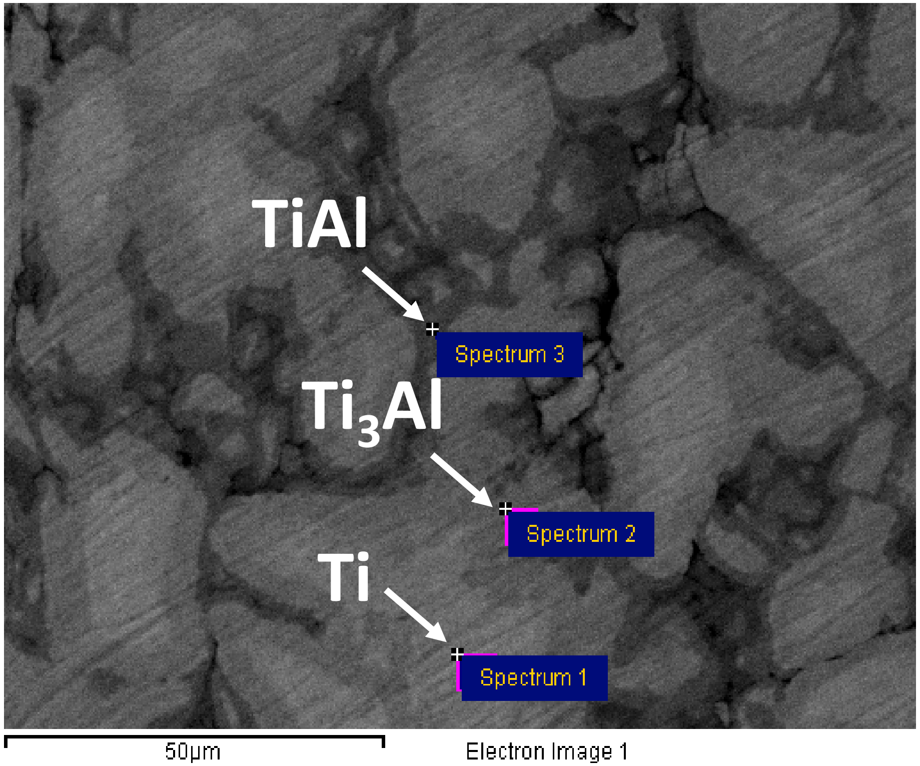


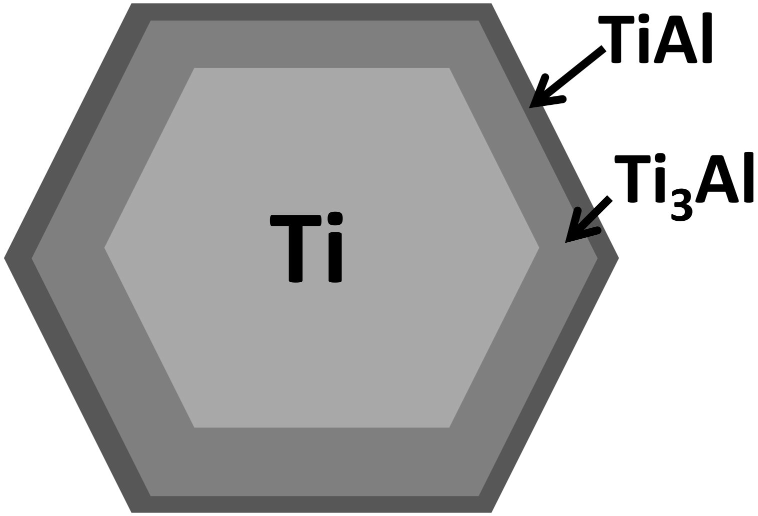
In Ti-Al binary system, there exist several electrically conductive compounds with high-T stability, which are known easily form by solid state reaction.

- Maintain high reaction activity during sintering.
- Suppress activity after sintering or during high-T service due to the formation of stable Ti_xAl.
- The Ti_xAl layer should not raise interfacial thermal/electrical resistance, while maintaining good bonding strength.

Microstructure of Ti-Al interlayer

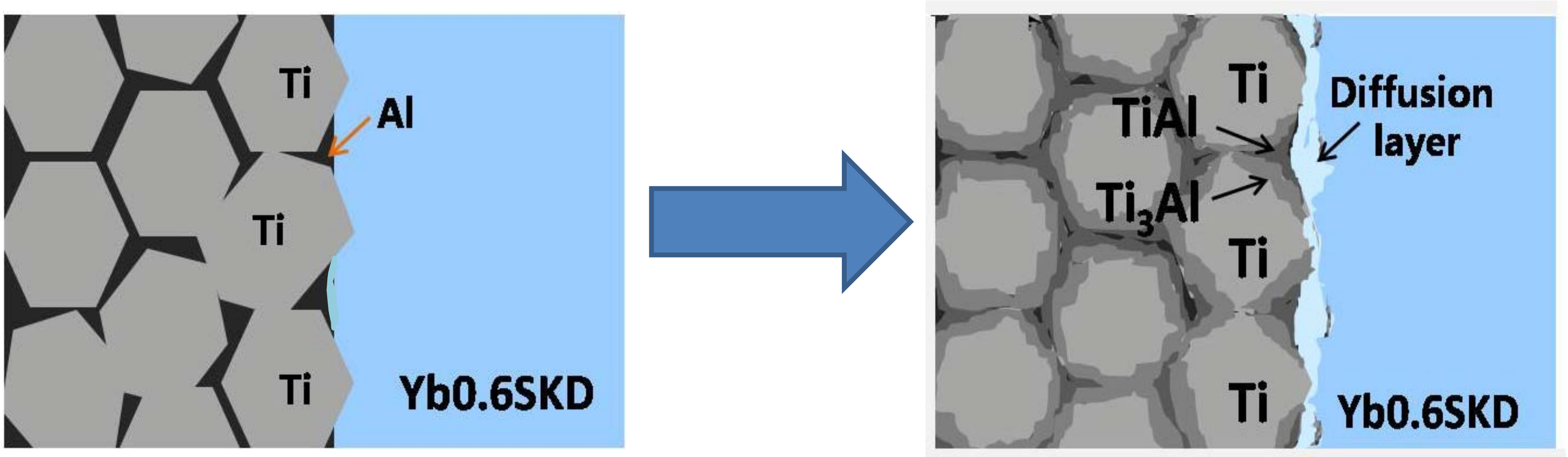
after annealing at 550°C





SEM and EPMA indicate the formation of Ti_3Al and TiAl alloys on the surface of Ti particles during high temperature annealing.

Sketch: Formation of Ti/Ti_3Al “core-shell” structure



Before sintering:

MoCu / (Ti+Al) / SKD

→ Ti & Al metals possess high activity and enable reaction and bonding during sintering.

After sintering or during high temperature operation:

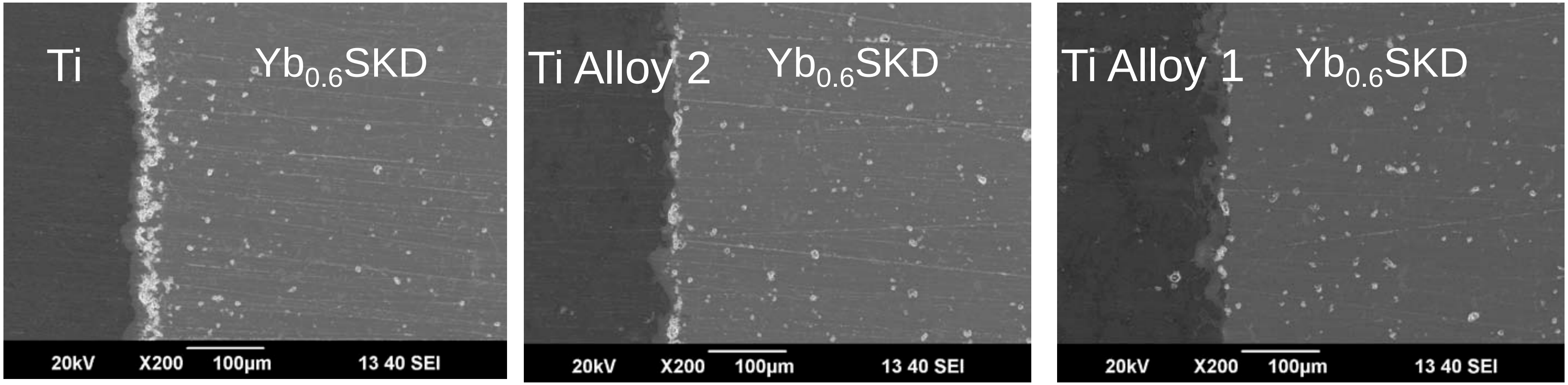
Ti+Al → Ti+Ti_3Al (or TiAl)

→ Ti/Ti_3Al core/shell structure prevents further Ti-diffusion and realize high stability.

J. Alloys Compd. **2014**

Stability of Ti-Al interlayer: Microstructure

High temperature stability for different Ti-based alloy interlayers.

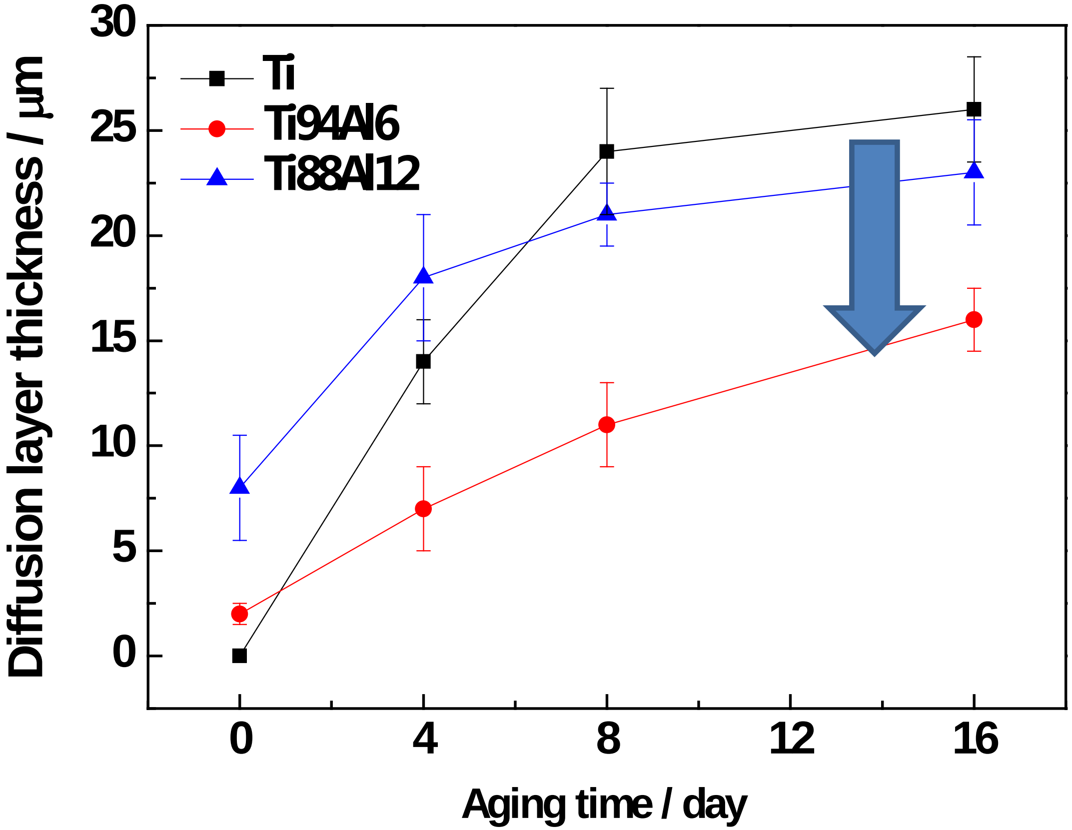


SEM images of cross section of elements after aging at 600°C for 16 days in vacuum.

- **No (or less) formation of TiCoSb brittle phase on the interface.**
- **The formation of TiCoSb intermetallic layer is effectively prevented by Ti_xAl alloying.**

J. Alloys Compd. **2014**

Stability of Ti-Al interlayer: Diffusion layer

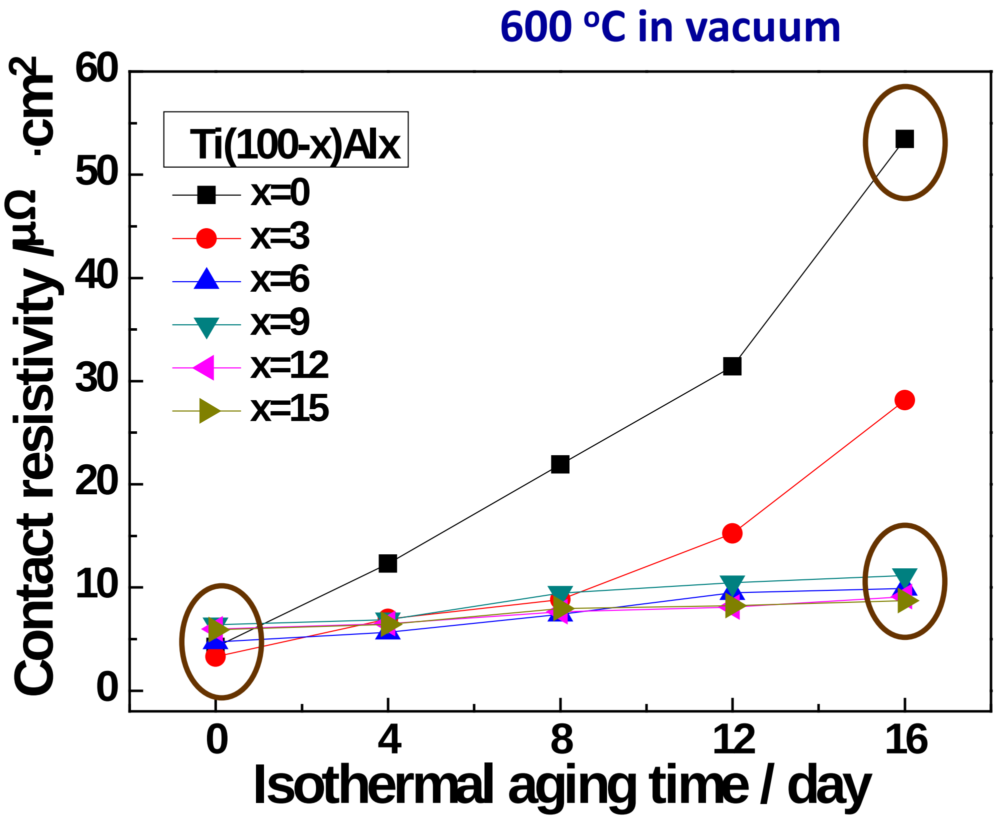


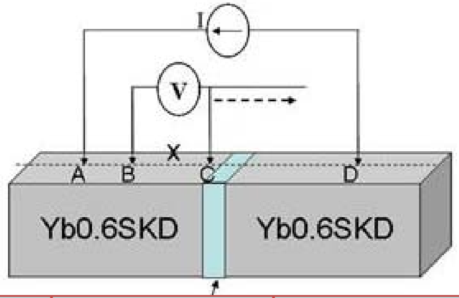
At 600°C in vacuum, 16 days

The growing speed of diffusion layer was greatly decreased after alloying Ti and Al.

J. Alloys Compd. **2014**

Interfacial resistance of $Ti_{100-x}Al_x$ / $Yb_{0.6}Co_4Sb_{12}$ joints





X	ρ_{c0} (μΩ•cm²)	ρ_{c16} (μΩ•cm²)
0	4.2	53
3	3.3	28
6	4.7	10
9	6.4	11
12	6	9
15	5.9	9

☐ *Ti interlayer:* *ρ_c increased by 12 times of its initial value after aging.*

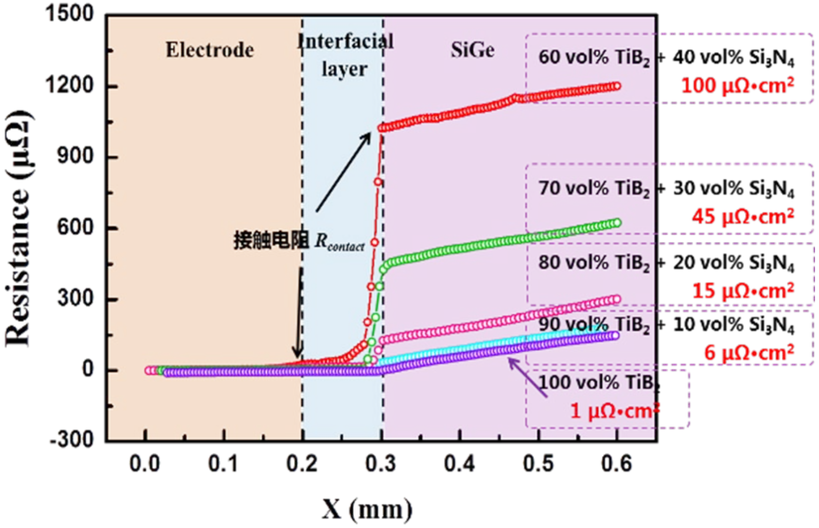
☐ *Ti-Al alloying interlayer:* *ρ_c changed less than twice after aging.*

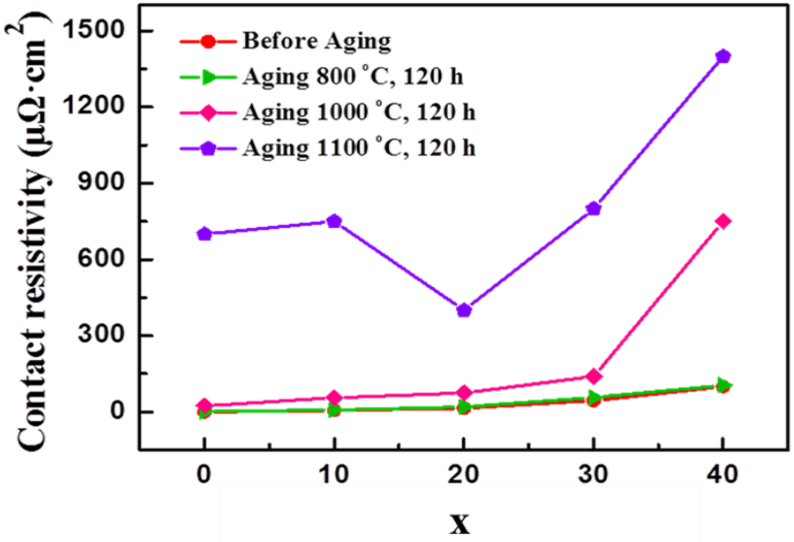
J. Alloys Compd. **2014**

Interface design target: Low energy loss & high reliability

◆ **SICCAS established interface solutions for several TE materials :**

TE materials	Electrode					Barrier layer	ρ_{inter} ($\mu\Omega cm^{-2}$)	$\Delta\ \eta$
	Content	σ (S/m)	κ (W/m•K)	CTE ($10^{-6}K^{-1}$)				
				Electrode	TE			
Bi_2Te_3	AlCu	3×10^7	300	20	15~22	Ni-Mo	< 10	< 3%
$CoSb_3$	MoCu	2.6×10^7	98	10.3	9.7~12.8	Ti-Al	< 10	< 3%
Half-heusler	MoCu	2.6×10^7	98	10.3	9.8~11.5	Zr-Al	< 5	< 2%
SiGe	W-Si_3N_4	4.4×10^6	49	4	4	TiB_2- Si_3N_4	< 20	< 5%

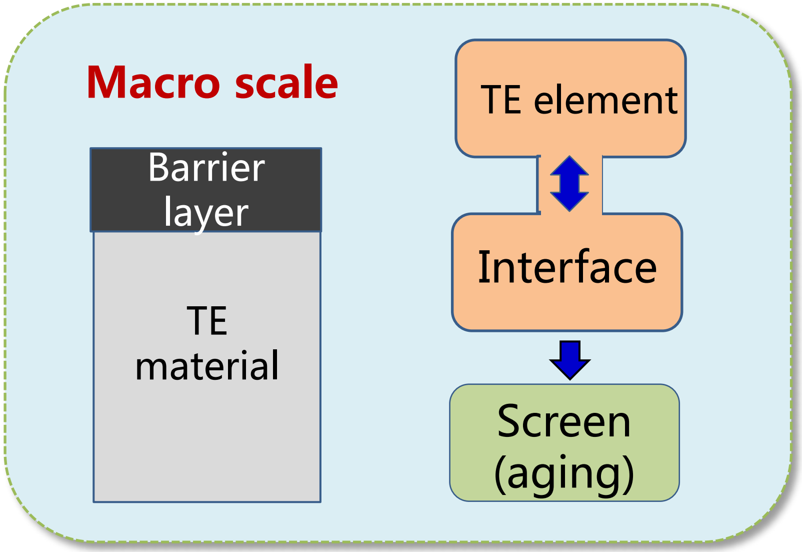




Ceram. Int. **2016**, 42, 8044-8050

high throughput strategy: Screen interfacial diffusion barrier

Traditional method



Macro scale

Barrier layer

TE material

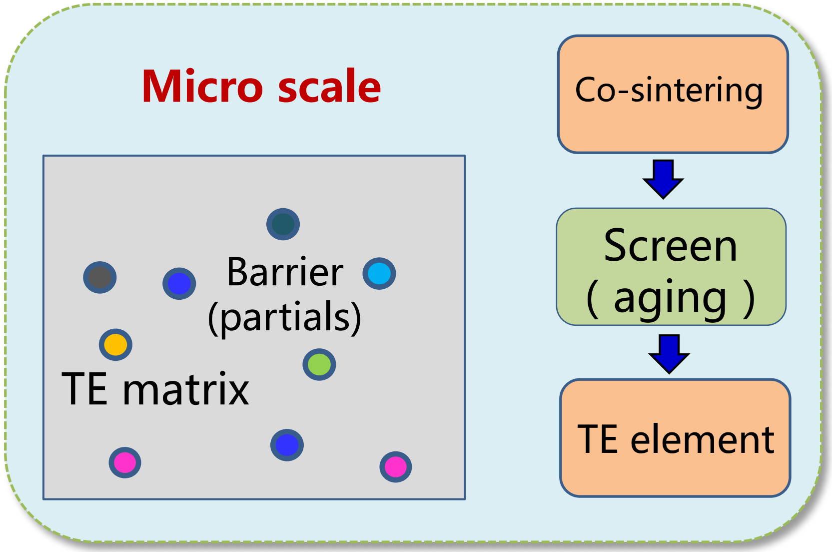
TE element

Interface

Screen (aging)

- Interface got based on the TE elements
- High cost in try error experiments of TE elements fabrication
- Long time of screening for one by one selection: months ~ years

High throughput strategy



Micro scale

Barrier (partials)

TE matrix

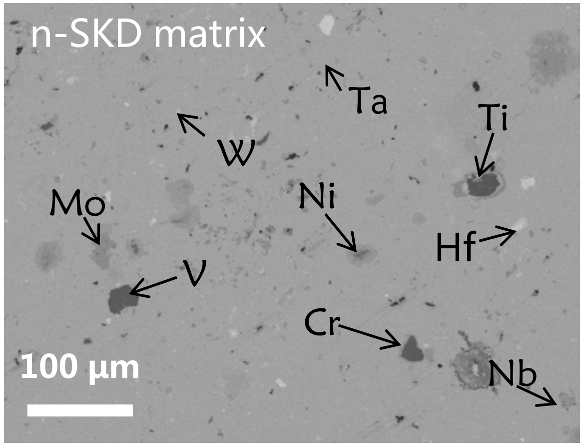
Co-sintering

Screen (aging)

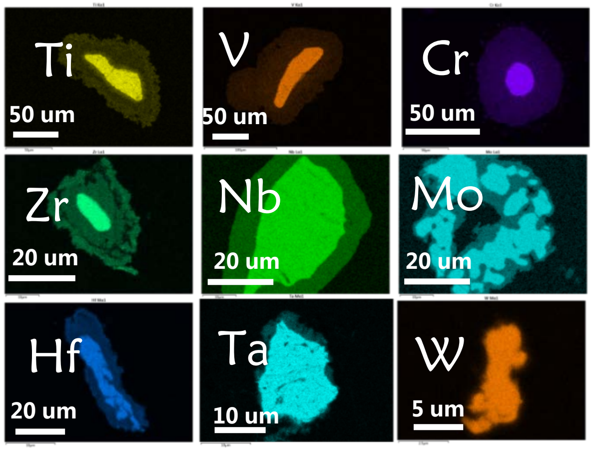
TE element

- Interface got NOT based on the TE elements
- Low cost in try error experiments of TE elements fabrication
- Short time of screening for one-time selection: weeks

high throughput strategy: Screen interfacial diffusion barrier



650°C×2 days



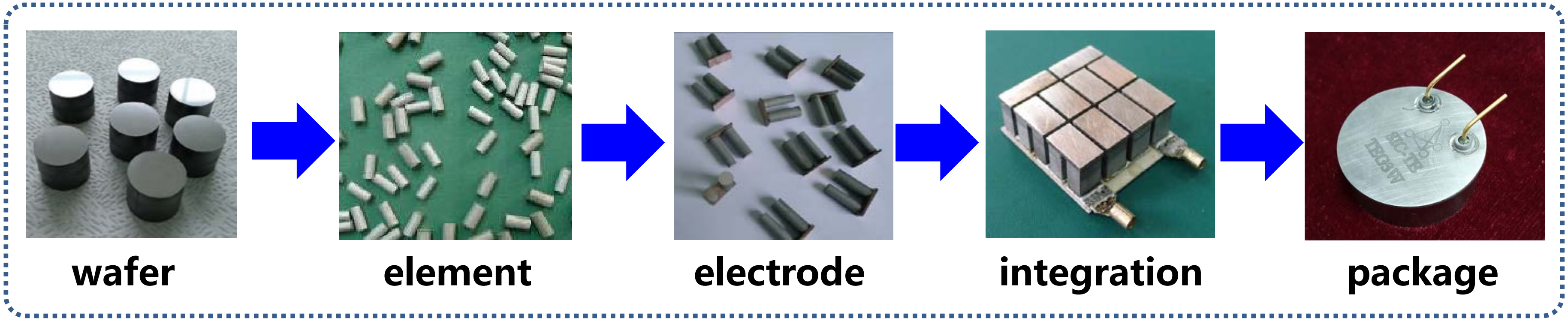
Barrier layer		**Diffusion layer** (650°C, 2D)	
Element	CTE/ppm	Content	Thickness/μm
V	8.3	VSb(?)	~50
Cr	6.2	CrSb ?	~40
Ti	8.4	TiSb, $TiSb_2$, TiCoSb	~30
Zr	5.8	$ZrSb_2$	~20
Hf	5.9	$HfSb_2$ HfCoSb	~15
Mo	5.4	Mo_3Sb_7	~10
Nb	7.0	$NbSb_2$	< 10
Ta	6.6	$TaSb_2$ TiCoSb	~5
W	4.6	N/A	N/A

*** Cost and fabrication technique should be considered in selection.**

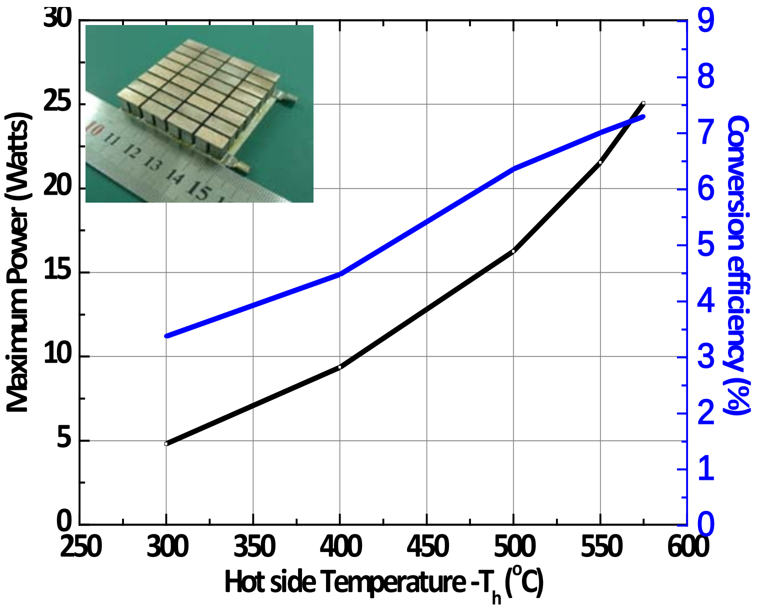
J. Mater. Res. **2018**, (under review)

Performance of SKD–based TE devices

Integration of SKD module



TYPICAL CURVES



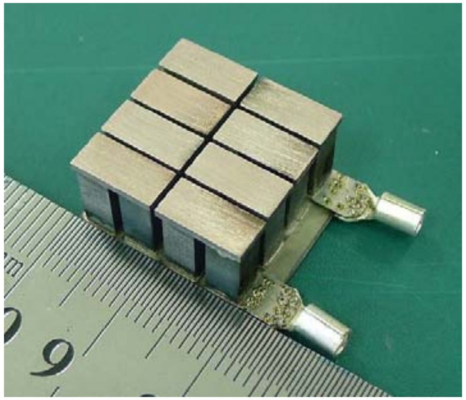
SICCAS TE group website

Data sheet of SKD TE generation modules

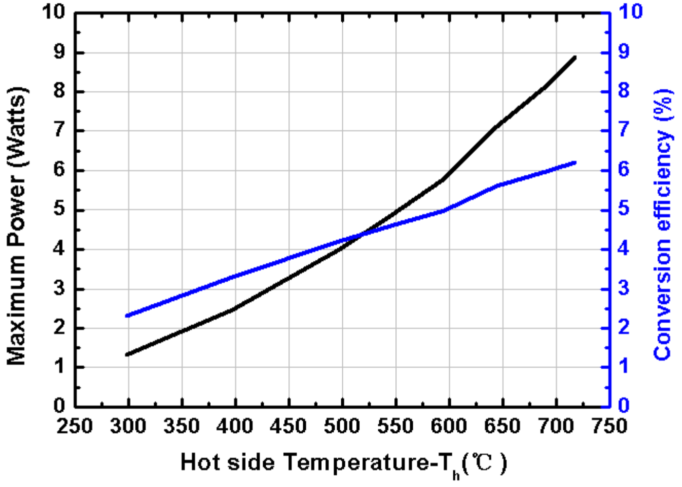
	Dimensions	Couples	Hot-side Temp. (°C)	Cold-side Temp. (°C)	V_{oc} (V)	P_{max} (Watt)	$R_{Inter.}$ (mΩ)
TEG5	Module Size: 20×20×10mm³	8	400	25	0.80	1.97	82
			500	30	1.06	3.30	85
	Dice Size: 4×4×8 mm³		550	30	1.19	4.09	87
			600	35	1.31	4.93	88
TEG10 (H-V)	Module Size: 30×30×10 mm³	18	400	35	1.50	3.85	139
			500	40	1.77	5.72	142
	Dice Size: 4×4×8 mm³		550	45	2.17	7.53	157
			600	45	2.53	9.54	173
TEG10 (L-V)	Module Size: 30×30×10 mm³	10	400	30	0.83	3.74	47
			500	35	1.11	6.33	48
	Dice size: 6×4.5×8 mm³		550	40	1.24	7.86	49
			600	45	1.38	9.51	50
TEG25	Module Size: 50×50×10 mm³	32	400	40	2.62	9.37	180
			500	50	3.52	16.25	188
	Dice Size: 5×5×8 mm³		550	55	4.08	21.55	191
			575	65	4.43	25.08	193

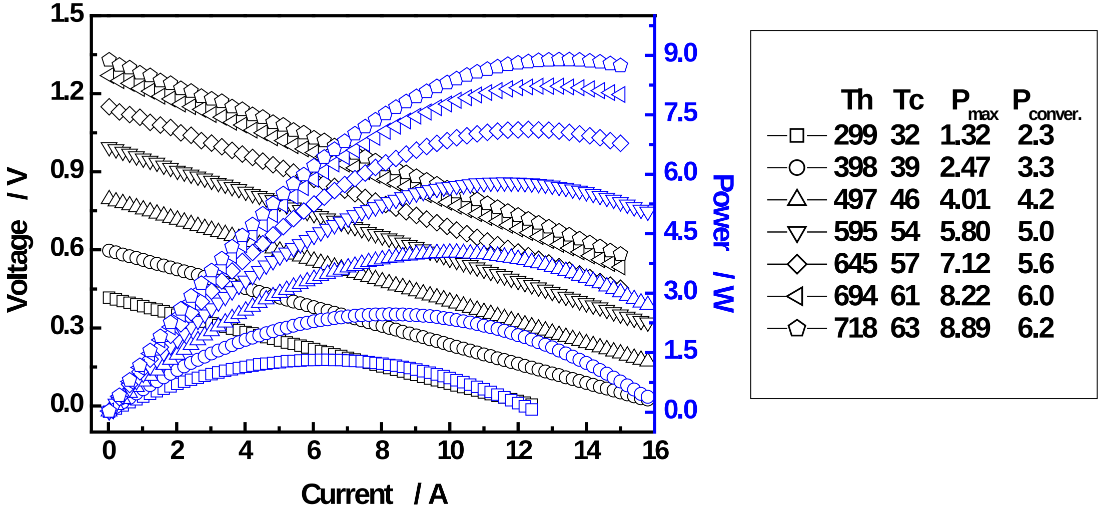
Performance of HH–based TE devices

- ☐ **Pb-free TE modules using HH alloys**
- ☐ **Capability of working at 700 °C (hot side)**
- ☐ **Output power up to 9 Watts (ΔT= 650°C)**
- ☐ **Efficiency up to 6.2% @ ΔT= 650°C**
- ☐ **Dimensions: 20mm×20mm×10mm(H)**
- ☐ **Output power density up to 2.2 W/cm²**



**** HH materials provided by Prof. Tiejun Zhu (Zhejiang University)***



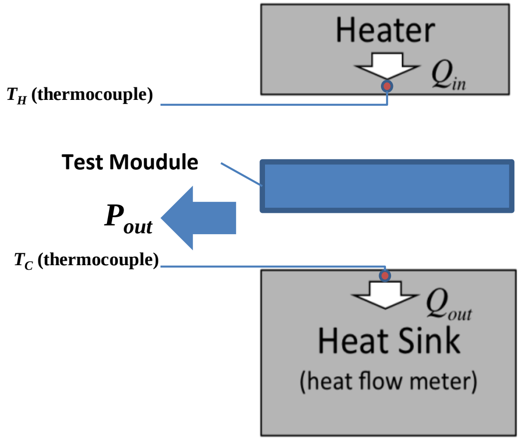


C. Fu, S. Bai, L. Chen: Nature communications (2015)

TEG module testing

1. **Agreement found in Electrical measurement of TE devices.**
2. **Significant measurement issues observed in heat flow measurement.**
3. **Measurement standard not established.**

Including: **Th & Tc, gas pressure, uniaxial pressure;**
Vaccum pump on/off during testing;
Scanning mode (scanning speed, temperature stage, ……)



Definition of efficiency:

$$\eta = \frac{P_{out}}{Q_{in}} \times 100\% = \frac{P_{out}}{Q_{out} + P_{out}} \times 100\%$$

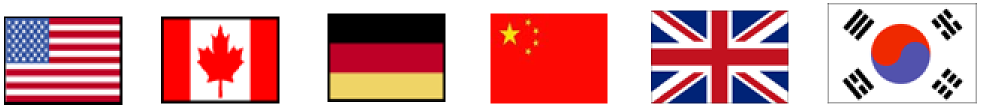
ULVAC-RIKO:PEM-2

Module testing system in AIST

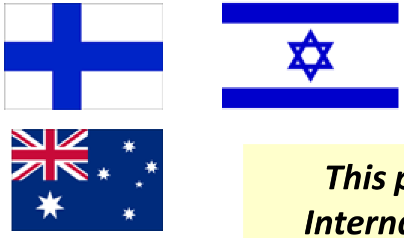
Testing standard of TEG materials and modules

Annex VIII Participants

- IEA-AMT Thermoelectric Annex
  - Annex lead: Oak Ridge National Laboratory (H. Wang)
  - USA: GMZ (G. Joshi); Marlow (J. Sharp); GM R&D (J. Salvador); Army Research Laboratory (P. Taylor), NIST (J. Martin)
  - China: SICCAS (S.Q. Bai, L. Chen)
  - Canada: CANMET(Y.C. Tseng); University of Waterloo (H. Kleinke);
  - Germany: Fraunhofer IPM (J. König)
  - United Kingdom: NPL (A. Cuenat)
  - Korea: Korea Electrotechnology Research Institute (H.W. Lee)



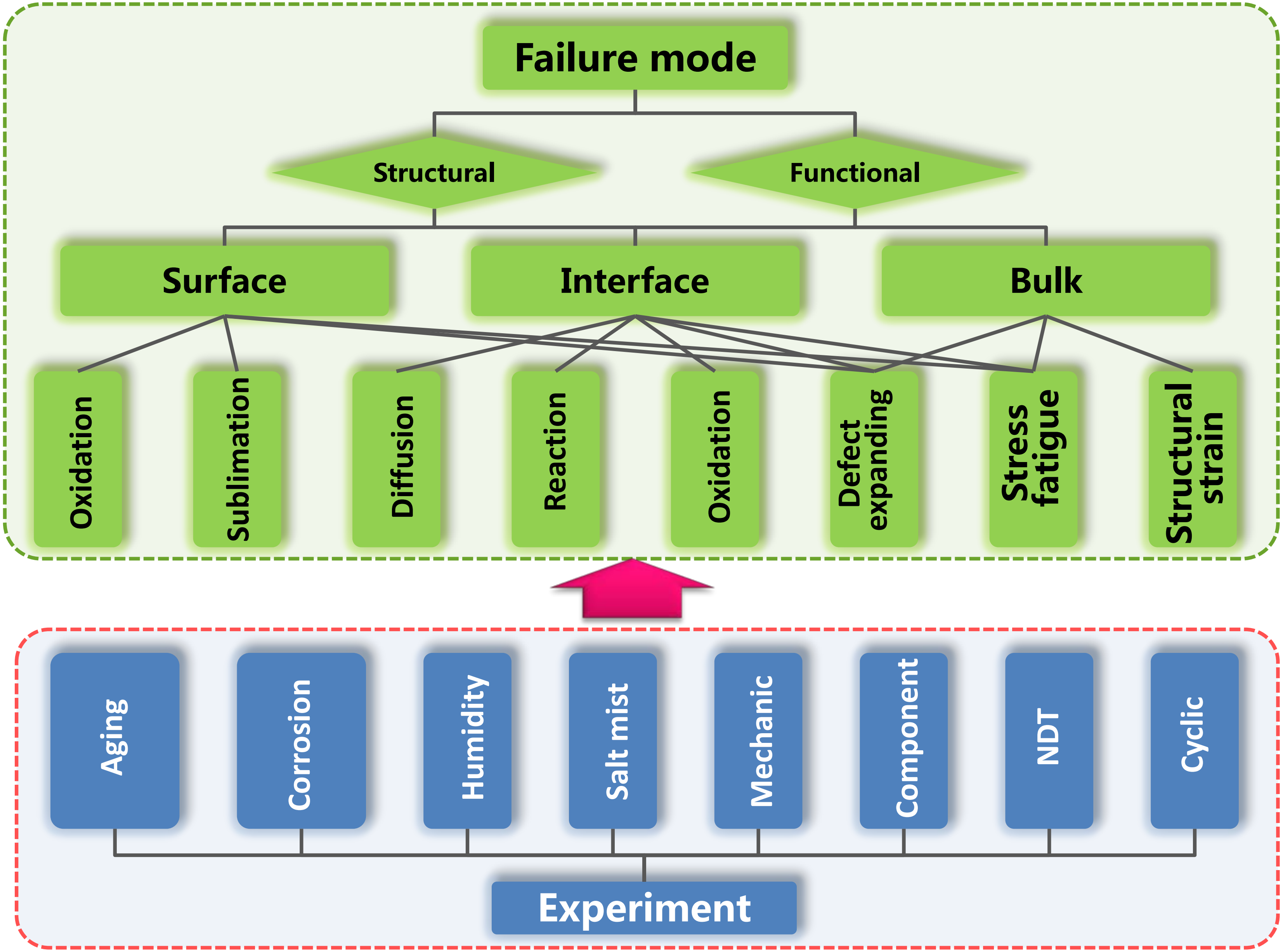
- IEA-AMT members countries:
  - Finland: VTT
  - Israel:
  - Australia:



This project is supported by the International Energy Agency (IEA) under the Implementing Agreement on Advanced Materials for Transportation (AMT)

From Dr. Hsin Wang (ORNL)

TEG module failure mode & service behavior


Failure mode

Structural

Functional

Surface

Interface

Bulk

Oxidation

Sublimation

Diffusion

Reaction

Oxidation

Defect expanding

Stress fatigue

Structural strain

Aging

Corrosion

Humidity

Salt mist

Mechanic

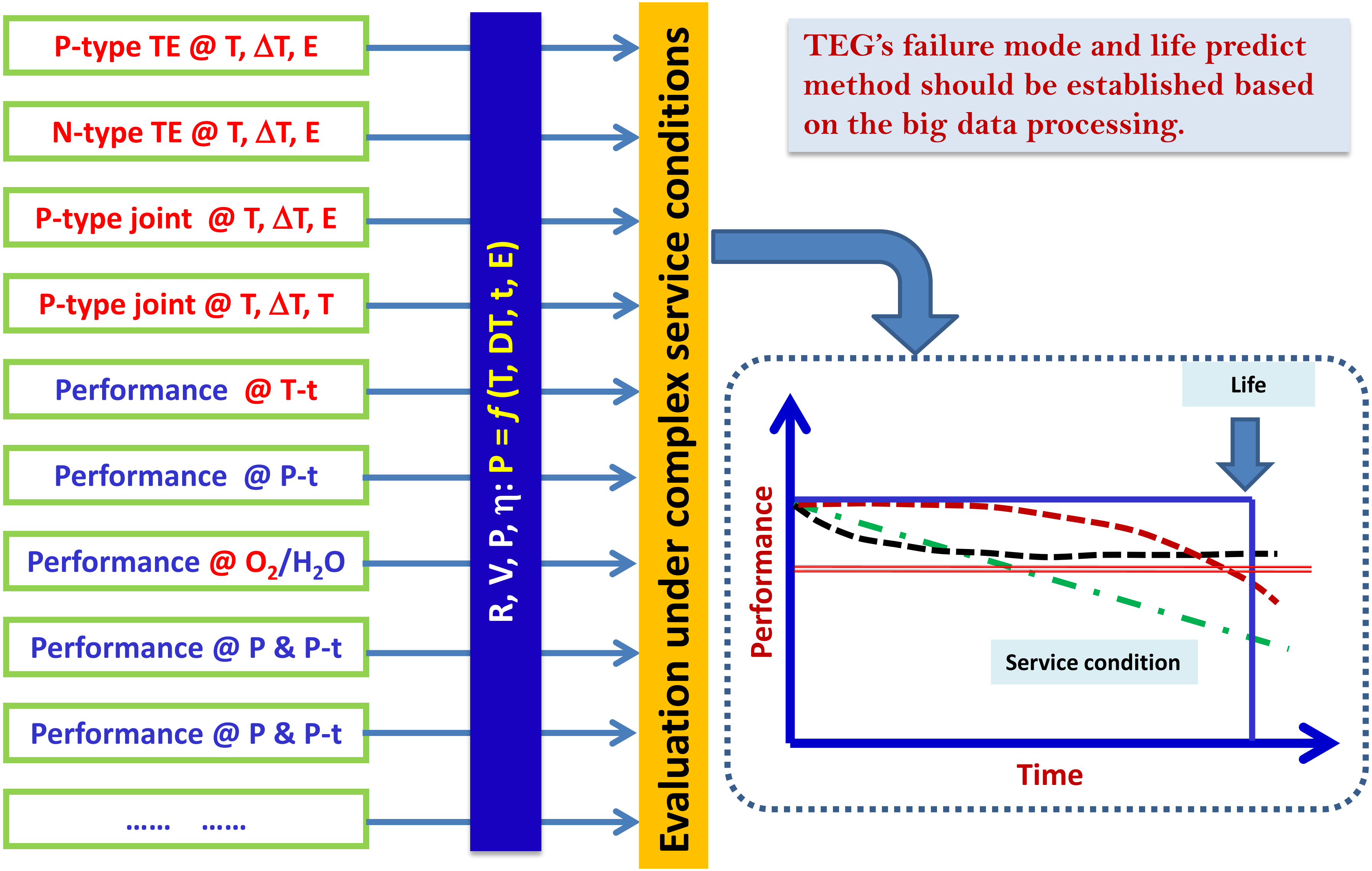
Component

NDT

Cyclic

Experiment

TEG module failure mode & service behavior



P-type TE @ T, ΔT, E

N-type TE @ T, ΔT, E

P-type joint @ T, ΔT, E

P-type joint @ T, ΔT, T

Performance @ T-t

Performance @ P-t

Performance @ O_2/H_2O

Performance @ P & P-t

Performance @ P & P-t

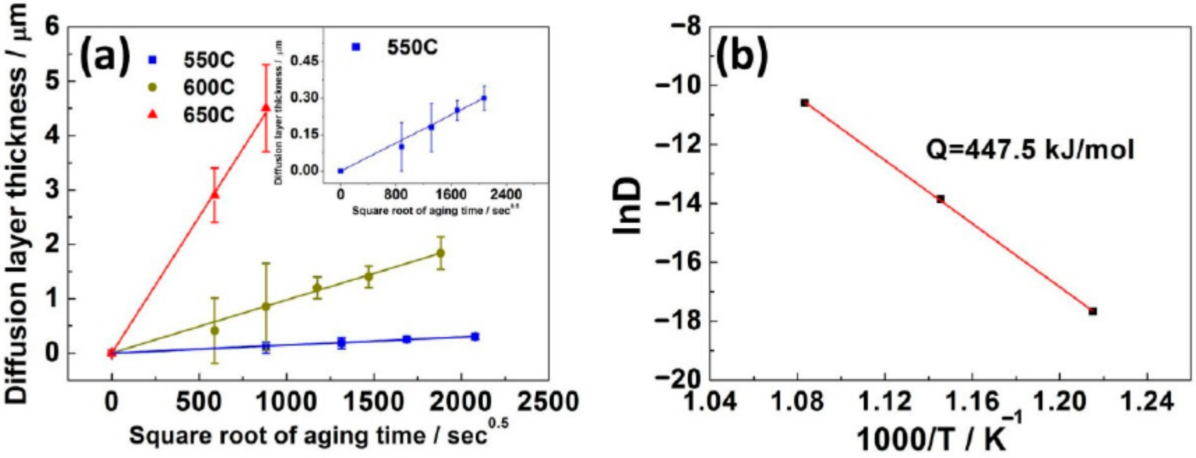
…… ……

R, V, P, η: P = ƒ(T, DT, t, E)

Evaluation under complex service conditions

TEG's failure mode and life predict method should be established based on the big data processing.

Life predict based on thermal dynamics

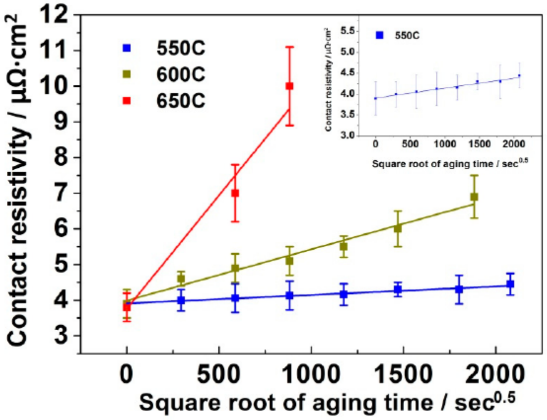


Temperature/°C	$D/10^{-19}$ m^2/s	Y/μm
550	0.21	$1.45 \times 10^{-4} \cdot t^{0.5}$
600	9.53	$9.76 \times 10^{-4} \cdot t^{0.5}$
650	251	$50.1 \times 10^{-4} \cdot t^{0.5}$

Arrhenius equation:

$$Y - Y_0 = (Dt)^{0.5}$$

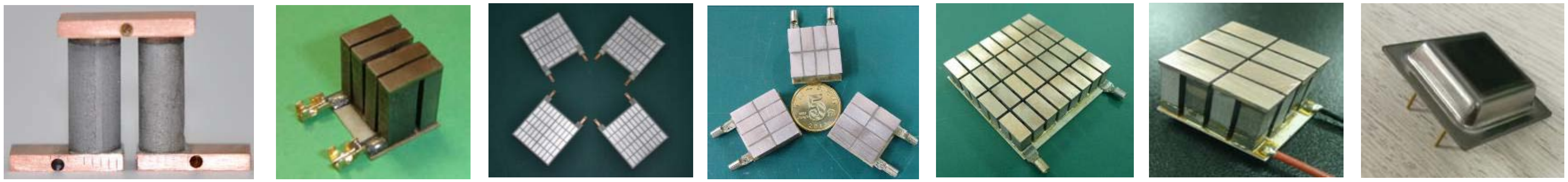
$$D = D_0 \exp\left(-\frac{Q}{RT}\right)$$



Life predict based on the interfacial resistivity

Temperature/°C	Contact Resistivity/μΩ·cm^2	Predicted Service Life (a)/day	Predicted Service Life (b)/day
550	$2.37 \times 10^{-4} \cdot t^{0.5} + 3.91$	7642	53,346
600	$14.1 \times 10^{-4} \cdot t^{0.5} + 3.99$	201	1431
650	$63.5 \times 10^{-4} \cdot t^{0.5} + 3.76$	11	75

Appl. Sci. ***2017, 7, 952***



Thank you for your attention !