

# Influence of temperature and aging on the thermal contact resistance in thermoelectric devices

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## Abstract

In the first phases of the development of thermoelectric systems, such as the thermoelectric generators, when the thermal design is carried out, the most important parameters affecting the performance are the thermal resistances of the components. This paper focusses on the thermal contact resistance (TCR), analyzing the influence of aging and temperature on different thermal interface materials (TIM), i.e., thermal paste, graphite and indium. In previous papers, TCR has been studied depending on parameters such as surface roughness, bonding pressure, thermal conductivity and surface hardness. However, in thermoelectric applications, a relevant aspect to consider when choosing a TIM is aging due to thermal stress. The exposure of this type of materials to high temperatures for long periods of time leads to deterioration, which causes an increase in the thermal contact resistance which impairs the conduction of the heat flow. Therefore, there is a need to study the behavior of thermal interface materials exposed to temperatures typical in thermoelectric generators, to make a correct selection of the TIM. It has been observed that exposure temperatures of around 180 °C induce a significant increase in the thermal impedance of the three TIM's under study, although this effect is much more relevant for the thermal paste. The contact comprising steel, thermal paste and ceramic presents a 300% increase in the thermal impedance after 70 days of aging, whereas that exceeds 185% for the contact of aluminum, thermal paste and ceramic. In the tests with exposure temperature of 60 °C, there is no observed decrease in the thermal impedance.

**Keywords:** Thermal contact resistance, Thermoelectric devices, Thermal interface material, Thermal aging, Ceramic steel contact, Ceramic aluminum contact.

## Nomenclature

$\Delta T$  Temperature difference between the fluxmeter surfaces in contact with the sample [°C]

A cross-sectional area of fluxmeter [m<sup>2</sup>]

$k_A$  Aluminum thermal conductivity [W/m °C]

$k_C$  Ceramic thermal conductivity [W/m °C]

$k_{\text{fluxmeter}}$  fluxmeter thermal conductivity [W/m °C]

$k_S$  Steel thermal conductivity [W/m °C]

$L_A$  Aluminum thickness [m]

$L_c$  Ceramic thickness [m]

$L_i$  sensors position in the fluxmeter “i= 1 to 4” [m]

$L_S$  Steel thickness [m]

$\dot{Q}$  heat flow through the contact between fluxmeters [W]

$R_c$  Thermal contact resistance, TCR [°Cm<sup>2</sup>/W]

$T_{av}$  Average sample temperature during the tests [°C]

$T_i$  Temperature sensors in the fluxmeter “i = 1 to 6” [°C]

$T_{3-}$  Bottom fluxmeter temperature in contact with the sample [°C]

45	$T_4$	Upper fluxmeter temperature in contact with the sample [°C]
46	$u(Z_g)$	uncertainty of Global thermal impedance [°Cm <sup>2</sup> /W]
47	$u(T_i)$	uncertainty of temperature sensors in the fluxmeter “i= 1 to 6” [°C]
48	$u(L_i)$	uncertainty of sensors position in the fluxmeter “i= 1 to 4” [m]
49	$u(\dot{Q})$	uncertainty of heat flow through the contact between fluxmeters [W]
50	$Z_k$	Thermal impedance due to the conductivity, [°Cm <sup>2</sup> /W]
51	$Z_g$	Global thermal impedance of the sample, [°Cm <sup>2</sup> /W]

52

### 53 INTRODUCTION

54 The development of thermoelectric applications requires adequate thermal designs to guarantee efficient  
55 operation. Specifically, the thermal resistance of all the components must be optimized. In this regard, the  
56 thermal contact resistances (TCR) between all the components must be as low as possible, that being the  
57 reason why thermal interface materials (TIMs) are used. Unfortunately, aging of TIMs is proven to increase  
58 the TCRs, which impairs the heat transfer between the thermoelectric modules and the heat exchangers,  
59 causing a decrease in the performance of the thermoelectric application.

60 The use of TIMs to reduce TCRs has been studied thoroughly in the field of electronics, as can be seen in  
61 the reviews presented in [1], [2], [3]. In addition, reference [4] presents a study on TIMs for high  
62 performance flip-chip-ball-grid arrays (HFCBGAs), whereas [5] conducts so for insulated gate bipolar  
63 transistors (IGBTs). Likewise, reference [6] summarizes the research on TIM reliability, with special  
64 interest in methodologies and results of several testing procedures. Complementary, there are studies of  
65 aging due to radiation on electronics for space applications [7], and studies on the effect of aging of TIMs  
66 in power electronics subjected to either thermal cycling [8-9] or isothermal [10] conditions.

67 Common test benches for TCR measurement applies the steady-state method, similar to the ASTM-D5470  
68 standard method [11]. The literature presents several examples of them, such as that described in [12],  
69 which uses a screw to apply pressure and cotton as insulator; or the one introduced in [13], which is used  
70 to measure the thermal contact conductance (TCC) of structural materials. More interesting is the bench  
71 described in [14], in which the applied pressure is controlled by a computer-based algorithm, and a vacuum  
72 chamber is used to remove convection losses. The TCR is obtained through the calculation of the  
73 temperature-dependent thermal conductivity of the rods used for determination of the heat flow.

74 Furthermore, a technical review of characterization methods of TCRs in TIMs can be found in [15].  
75 Recently, a review of the performance and characterization of TIMs for electronic applications is available  
76 in [16].

77 However, aging of TIMs has been scarcely studied in thermoelectric applications [17], despite its huge  
78 impact on the final efficiency of these systems. This is the main goal of this paper. The knowledge on the  
79 TIM performance (that is, its influence on the TCR) for long periods at high temperatures is essential to  
80 ensure the efficient performance of a thermoelectric system over its working life. The selection of the best  
81 TIM for each application would lead to reductions in both maintenance and economic cost.

82 To fulfill this goal, the test bench presented in this paper has been developed and tested. This bench was  
83 designed specifically for thermoelectric applications. It allows the calculation of TCR of several TIMs  
84 depending on the temperature and pressure, including also the effect to aging. The test bench is presented  
85 in the following section; then, the measurement methodology for TCR characterization is introduced, along  
86 with the aging protocol. After that, the experimental results on the evolution of TCRs are described; and  
87 finally, some conclusions and perspectives are presented.

## 88 **EXPERIMENTAL SET-UP**

89 An original experimental steady-state test bench has been designed and constructed, which can be seen in  
90 Fig.1. It is installed inside an environmental chamber with controlled temperature and humidity (60% in all  
91 the tests), wherein the air can be considered to have zero velocity. The test bench comprises two solid  
92 blocks (called fluxmeters) between which the TIM is placed, a heat source installed at the bottom, and a  
93 forced-convection heat sink at the top. Heat is forced to flow through the fluxmeters and the sample, while  
94 the pressure between them is controlled. The heat source is composed of four electric cartridges inserted in  
95 a solid piece of steel, providing up to 400 W (100 W per cartridge). Finally, the fluxmeters are made of 304  
96 AISI INOX steel. They have presented no significant variation in their thermal conductivity along the tests.

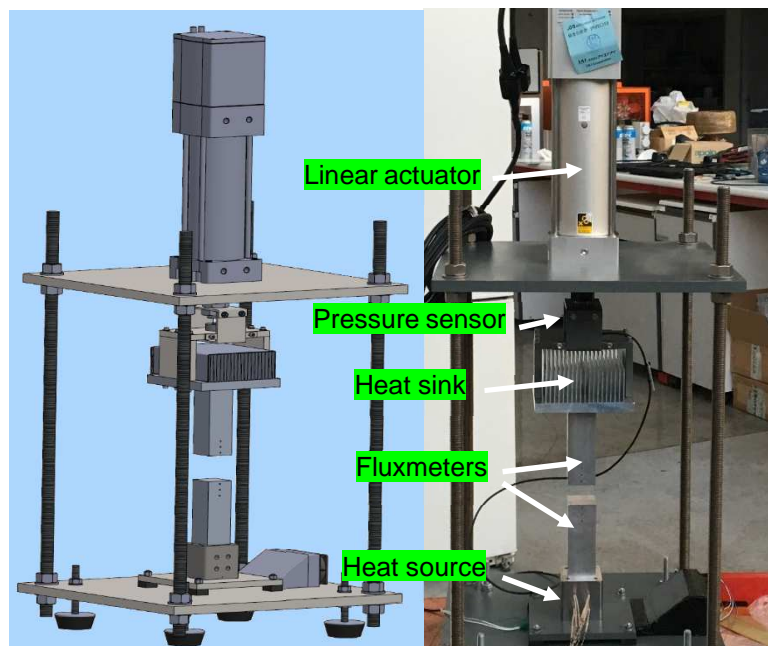
97 The fluxmeter at the bottom connects the heat source and the sample, and has a base area of 40x40 mm<sup>2</sup>,  
98 similar to that of the sample and also to that presented by common thermoelectric modules. The fluxmeter  
99 at the top presents also 40x40 mm<sup>2</sup> of base area and connects the sample and the heat sink. The heat sink is  
100 made of 6063 T5 aluminum, presents base area of 150x150 mm<sup>2</sup> and 10 mm of base height, and includes  
101 fins of 30 mm in height and 1.5 mm in thickness separated 3 mm, and a fan to produce forced convection.  
102 Given that the exposed surfaces of the test bench (fluxmeters and sample) are covered by a thick layer of

103 insulator (see Fig. 2), the heat flow can be considered unidimensional, flowing through the fluxmeter, the  
104 sample and the heatsink.

105 Six Pt-100 temperature probes (model FPA15L0100, with measuring range from  $-50^{\circ}\text{C}$  to  $500^{\circ}\text{C}$ ,  
106 uncertainty of  $0.1^{\circ}\text{C}$ , and diameter of 1.5 mm), are inserted in holes made on the fluxmeters, which present  
107 diameter of 1.75 mm and depth of 20 mm (see Fig. 3). An additional probe measures the ambient  
108 temperature. The uncertainties associated to the temperatures, lengths and diameters were calculated in the  
109 calibration laboratory Applus + AC6, located in Navarre (Spain).

110 The bench includes the linear actuator RCP2-RA10C (built by IAI AMERICA), which presents a capacity  
111 of up to 6 kN. It allows to perform the tests with the assembly pressure recommended by the manufacturers  
112 of thermoelectric modules. The pressure sensor K-1613 (LORENZ MESSTECHNIK), with an upper limit  
113 of 10 kN, allows the measurement of the tension to which the sample is subjected and the control of the  
114 force of the actuator, to ensure that the sample is at the desired pressure. All data is recorded with an  
115 ALMEMO 5690-2M09TG3 connected to a personal computer.

116 The accuracy in the measurement depends on the precise account of the heat flowing through the sample,  
117 so heat losses must be minimized. To do so, the upper and lateral sides of the fluxmeters are insulated with  
118 two layers of insulation material (an inner layer of rock wool and an outer layer of neoprene).



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Fig.1. Test bench

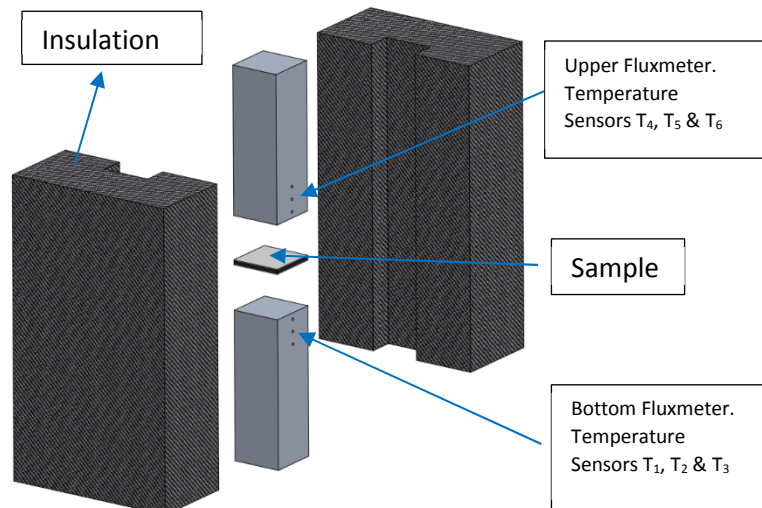


Fig. 2. Position of temperature sensors and fluxmeter insulator

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## 122 THERMAL CHARACTERIZATION METHOD

123 Previous works of our research group, both on thermoelectric refrigeration [18] and thermoelectric power  
 124 generation [19], [20], [21], made evident that a precise characterization of the TIM is required in  
 125 thermoelectric systems. To describe the thermal characterization method and further selection of the TIM,  
 126 the conditions described in [19] have been adopted. That paper presents a real thermoelectric generator that  
 127 harvest heat from the hot gases in a chimney, as can be seen in Fig. 3. Several thermoelectric modules (with  
 128 outer layer of ceramic material) are installed between the hot surface of the chimney (made of steel) and  
 129 the heat sinks (made of aluminum). TIMs are installed to improve the contact between them. Therefore,  
 130 two different testing configurations arise from this application:

- 131 • STEEL-TIM-CERAMIC, (S-TIM-C): The sample is a sandwich composed of a layer of AISI-304  
 132 steel (base area of 40x40 mm<sup>2</sup>, thickness of 4.5 mm,  $k_S$  of 16 W/m°C) and a layer of Al<sub>2</sub>O<sub>3</sub> ceramic,  
 133 Alumina 96% PER MI 866-1005 Marlow Industries, inc. (base area of 40x40 mm<sup>2</sup>, thickness of  
 134 0.75 mm,  $k_C$  of 35 W/m°C) connected by a TIM. This assembly represents the contact between the  
 135 chimney and the hot face of a thermoelectric module.
- 136 • CERAMIC-TIM-ALUMINUM, (C-TIM-A): The sample is a sandwich composed of a layer of  
 137 6063-T5 aluminum (base area of 40x40 mm<sup>2</sup>, thickness of 10 mm,  $k_A$  of 196 W/ °C) and a layer  
 138 of Al<sub>2</sub>O<sub>3</sub>, with a TIM between them. This assembly represents the contact between the cold face  
 139 of a thermoelectric module and the heat sink.

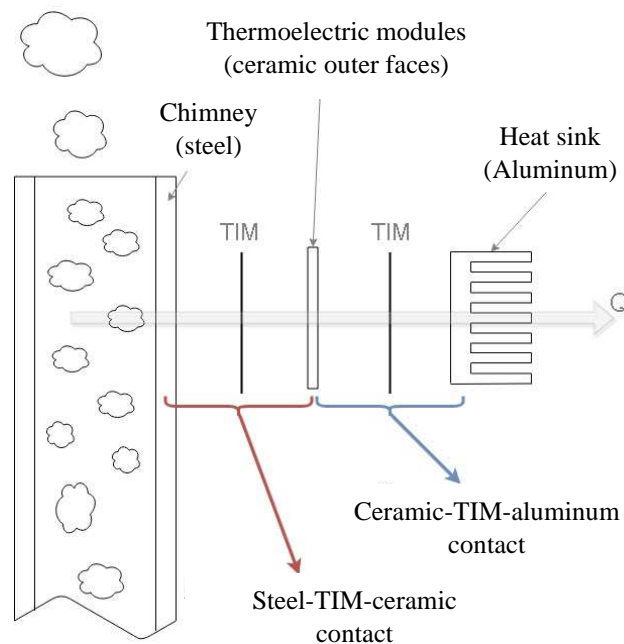
140 Three different TIMs are studied for both configurations:

- 141 • Graphite pad HT2505 (G) (GrafTech International Holdings Inc).
- 142 • Phase Change Material, Indium (I). KITEA-85553 Indium Corporation.
- 143 • Polysynthetic oils thermal grease Artic Silver 5, which contains micronized silver, sub-micron
- 144 zinc oxide, aluminum oxide and boron nitride particles (P).

145 The expected aging due to thermal stress in a TIM is an aspect to consider when choosing the TIM for a  
146 specific thermoelectric application. Aging causes a deterioration of the material, with an increase in its  
147 thermal contact resistance that impairs the heat conduction. Therefore, it is necessary to study the  
148 performance of these materials at high temperatures for long periods of time.

149 Most of TIMs in the literature are used for heat dissipation in electronic devices, whose maximum  
150 temperature hardly exceeds 100°C. However, in thermoelectric applications, especially in electric power  
151 generation, higher temperatures are reached, as occurs in the application presented in this paper, where the  
152 temperature of the outer surface of the chimney lies around 180°C. Consequently, 180°C is selected as aging  
153 temperature for the TIM in contact with the chimney. Similarly, 60°C is selected as aging temperature for  
154 the TIM in contact with the heat sink, as lower temperatures are expected in this component. The aging  
155 process is conducted in two ovens that maintain the temperature constant at 60°C and 180°C respectively.  
156 Therefore, as two aging temperatures have been tested for three different TIMs in two configurations, a  
157 total of 12 studies have been conducted.

158



159

160 Fig. 3. Scheme of the contacts in a thermoelectric generator

161 A specific configuration has been used for each contact, as can be seen in Fig. 4. In steady state, the global  
 162 thermal impedance  $Z_g$  is defined as the temperature gradient between the faces of the fluxmeters in contact  
 163 with the sample, per unit of heat flux passing thorough the interface area, ( $\dot{Q}/A$ ).

$$164 \quad Z_g = \Delta T / (\dot{Q}/A) \quad (1)$$

165 The term  $\Delta T$  ( $= T_{3'} - T_{4'}$ ) is the temperature difference between the surfaces of the fluxmeters. These  
 166 temperatures are obtained by extrapolation from the temperatures measured by the probes located along the  
 167 fluxmeters when the thermal equilibrium is reached, according to Eqs. 2 and 3.

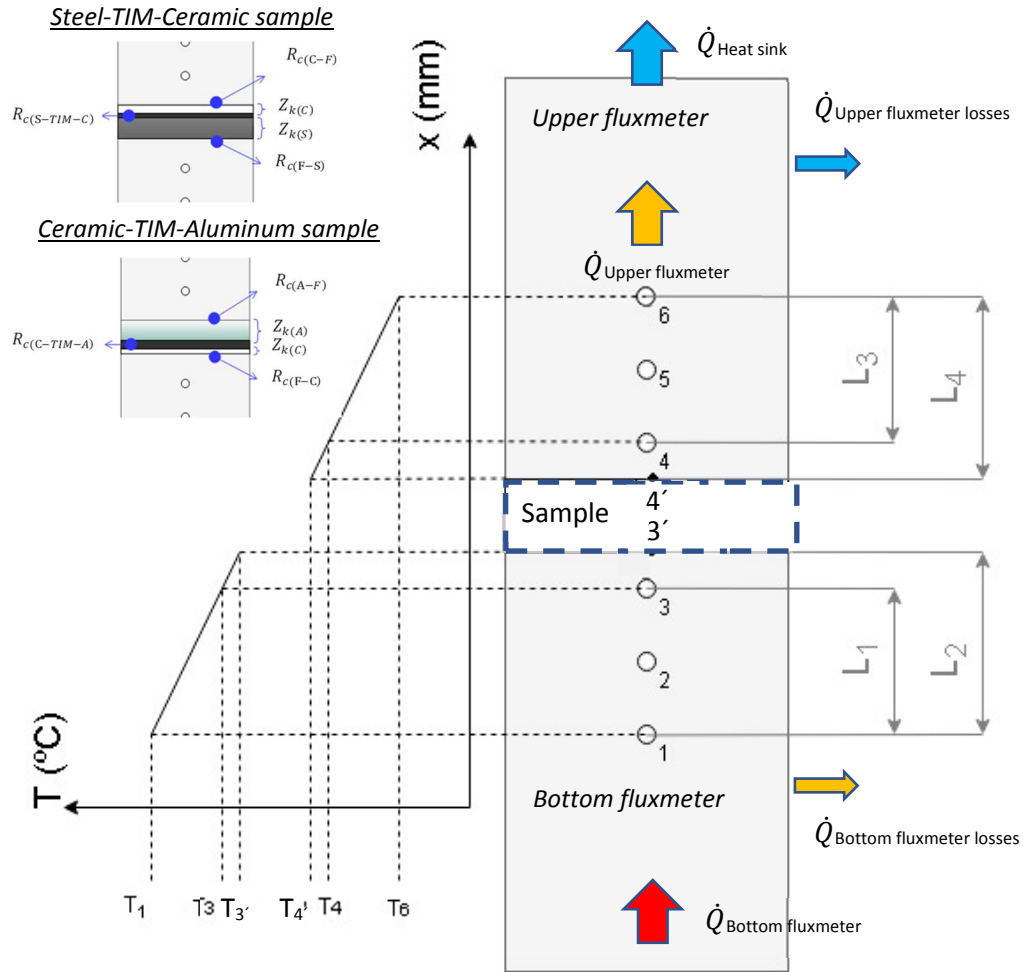
$$168 \quad T_{3'} = \frac{L_2}{L_1} T_3 + \left(1 - \frac{L_2}{L_1}\right) T_1 \quad (2)$$

$$169 \quad T_{4'} = \frac{L_4}{L_3} T_4 + \left(1 - \frac{L_4}{L_3}\right) T_6 \quad (3)$$

170 Heat is supplied at one end (generator, heat source) and dissipated at the other (heat sink), as Fig. 1 displays.  
 171 With the adequate insulation, the heat flow can be quantified as the thermal energy produced in the  
 172 generator minus the thermal energy that leaks through the insulator (see Fig. 2). The heat flow through  
 173 fluxmeter at the bottom has two terms: the leaks plus the heat flowing through the sample. Considering that  
 174 the sample is thin, and the sides are well insulated, the heat flowing through the sample is equal to that  
 175 flowing through the fluxmeter at the top. Therefore, by knowing the thermal conductivity of the fluxmeter  
 176 and the temperatures measured by  $T_4$  and  $T_6$ , (whose separation is precisely known), we can determine the  
 177 heat flux passing thorough the interface area ( $\dot{Q}$ ) with Eq. (4).

$$178 \quad \dot{Q} = k_{fluxmeter} \frac{T_4 - T_6}{A * L_3} \quad (4)$$

179 The thermal conductivity of the fluxmeter (AISI-304 stainless steel) used to calculate the heat flux was  
 180 taken at the average temperature between the sensors  $T_4$  and  $T_6$ . The temperature-dependent thermal  
 181 conductivity is obtained from [22].



182

183 Fig. 4. Configurations for the contacts and temperature sensors. Thermal and heat flux distribution.

184 Figure 4 shows the configuration of the two contacts and the thermal impedances that come from them.

185 The global thermal impedance in the case of the steel-ceramic contact  $Z_{g(S-TIM-C)}$  is composed of the

186 conductive thermal impedances of the ceramic material ( $Z_{k(C)}$ ) and steel ( $Z_{k(S)}$ ), along with three thermal

187 contact resistances: two due to the thermal contact resistances between the fluxmeters and the sample,

188 (ceramic and steel  $R_{c(F-S)}$  and  $R_{c(C-F)}$ ), and the thermal contact resistance inherent to the TIM ( $R_{c(S-TIM-C)}$ ).

189 The calculation of the global thermal impedance in the case of aluminum-contact,  $Z_{g(S-TIM-C)}$ , is similar but

190 substituting steel for aluminum.

191 The global thermal impedance of the Steel-TIM-Ceramic sample presents the following expression:

192 
$$Z_{g(S-TIM-C)} = R_{c(F-S)} + Z_{k(S)} + R_{c(S-TIM-C)} + Z_{k(C)} + R_{c(C-F)} \quad (5)$$

193 And the global thermal impedance of the Ceramic-TIM-Aluminum sample is:



194  $Z_{g(C-TIM-A)} = R_{c(F-C)} + Z_{k(C)} + R_{c(C-TIM-A)} + Z_{k(A)} + R_{c(A-F)}$  (6)

195 The thermal impedance for the layers of aluminum, steel and ceramic are respectively:

196  $Z_{k(A)} = \frac{L_A}{k_A}$  (7)

197  $Z_{k(S)} = \frac{L_S}{k_S}$  (8)

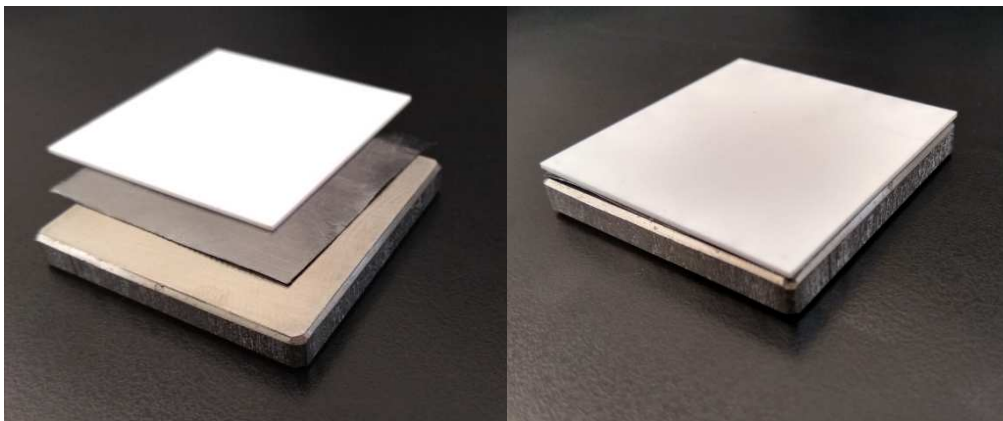
198  $Z_{k(C)} = \frac{L_C}{k_C}$  (9)

199 In our research, these impedances result to be  $Z_{k(A)} = 0.509 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ ;  $Z_{k(S)} = 2.81 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ ;  $Z_{k(C)}$   
 200  $= 0.214 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ ;

201 The thermal contact resistances between the fluxmeters and the ceramic-TIM-steel sample ( $R_{c(F-S)}$  and  $R_{c(C-}$   
 202  $F)$ ), as well as those between the fluxmeters and the ceramic-TIM-aluminum sample ( $R_{c(A-F)}$  and  $R_{c(F-C)}$ ),  
 203 are calculated prior to performing the tests. In all the cases, we have assumed that these values do not  
 204 change due to the use of fresh graphite pads, and therefore these contacts do not suffer aging. The thermal  
 205 contact resistances between the fluxmeters and the samples turn out to be  $R_{c(F-S)} = 0.535 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ ;  $R_{c(C-}$   
 206  $F) = R_{c(F-C)} = 0.576 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ ;  $R_{c(A-F)} = 0.438 \times 10^{-4} \text{ m}^2 \text{ }^\circ\text{C/W}$ .

207 The thermal contact resistance of the TIM ( $R_{c(TIM-)}$ ) is the only term of the global thermal impedance  $Z_g$   
 208 that is affected by the aging process. Therefore, the variation in the global thermal impedance due to aging  
 209 shows the trend in the thermal contact resistance of the TIM.

210 The testing protocol includes, in the first place, the assembly of the sample (TIM, ceramic plate and steel  
 211 or aluminum block), as can be seen in Fig. 5, which is then installed between the fluxmeters.



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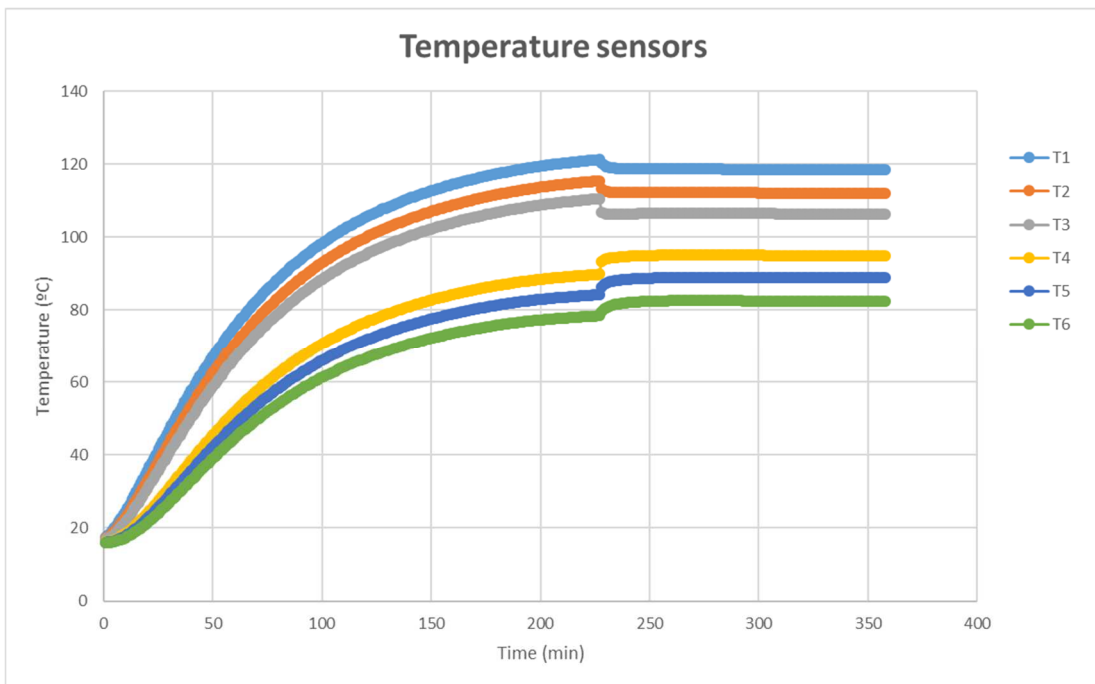
Fig. 5. Assembly of Steel-graphite-ceramic sample

214 Subsequently, the electrical cartridges are connected to a DC power supply to generate and control the heat  
 215 flow. The data provided by the temperature and pressure sensors is monitored in real time, as can be seen  
 216 in Fig. 6 and Fig. 7. Therefore, when temperatures reach the steady state, the actuator is activated to apply  
 217 the required pressure. It is important to do this step after the complete stabilization of the temperatures, to  
 218 ensure the complete dilatation of all the components. Finally, once both the pressure and the temperatures  
 219 are stable again, the data is recorded.

220 After the first test, the samples are introduced in the corresponding oven to start the aging process. The  
 221 samples with the TIM remain in the oven for 70 days (1680 hours). Within that period, the tests are  
 222 conducted every 20 days. Every sample is taken from the oven and installed in the test bench. Once it  
 223 reaches room temperature, heat flow is generated until the average temperature  $T_{av}$  between  $T_3$  and  $T_4$  lies  
 224 between 97 °C and 103 °C. The temperature evolution during this process is shown in Fig. 6. This average  
 225 temperature is equal in all the tests, in order to obtain comparable results.

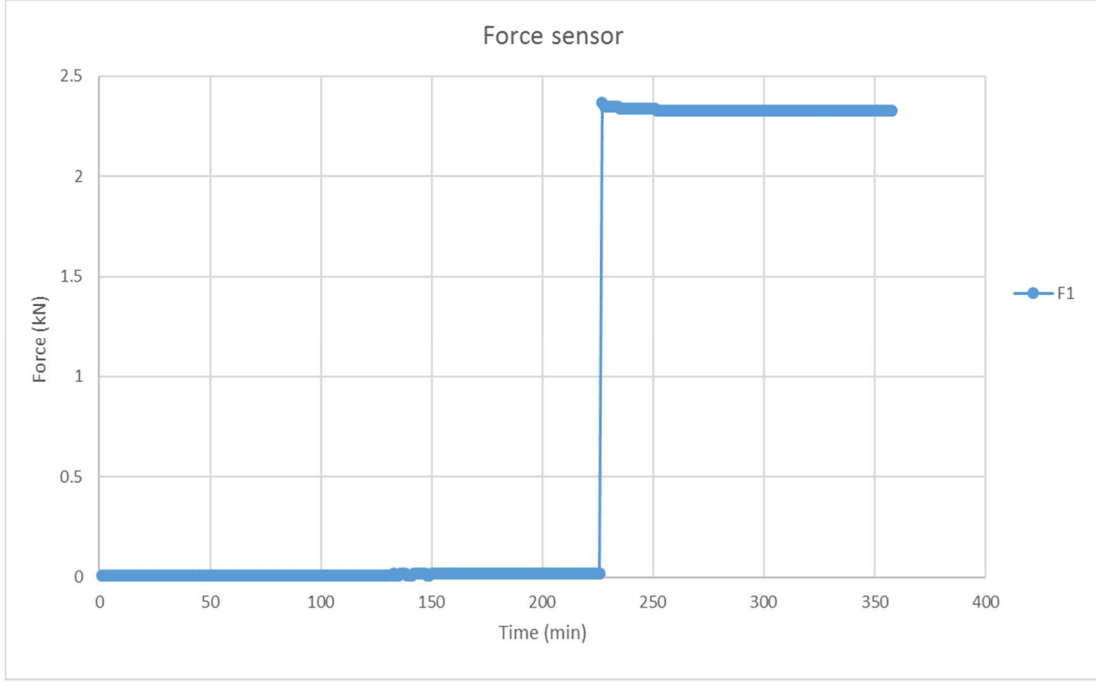
226 
$$T_{av} = \frac{T_3 + T_4}{2} \tag{10}$$

227 The thermoelectric modules used in our previous works are Marlow TG12-6, which are recommended to  
 228 operate under a pressure of 1.4MPa. This is the pressure applied in all the tests. In the oven, no pressure  
 229 was applied to the samples.



230

231 Fig. 6. Temperature sensors monitored in real time, testS-G-C\_60 (aging of 20 days)



232

233

Fig. 7. Force sensor monitored in real time, testS-G-C\_60 (aging of 20 days)

234

The uncertainty in the measurement of the global thermal impedance  $Z_g$  is calculated by the uncertainty

235

propagation method. We assume that the uncertainty of the longitudinal heat flux  $u(\dot{Q})$  is 5% and is

236

independent of the rest of the variables. Equation 11 relates  $Z_g$  to the temperatures and positions of the

237

probes. This equation is obtained by the combination of eqs. 1, 2 and 3.

238

$$Z_g = A \left( \frac{T_1}{\dot{Q}} - \frac{L_2 T_1}{L_1 \dot{Q}} + \frac{L_2 T_3}{L_1 \dot{Q}} - \frac{T_6}{\dot{Q}} + \frac{L_4 T_6}{L_3 \dot{Q}} - \frac{L_4 T_4}{L_3 \dot{Q}} \right) \quad (11)$$

239

Then, eq. 12 provides the uncertainty in the measurement of the global thermal impedance, wherein the

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uncertainties associated to temperatures and lengths were calculated in a calibration laboratory (Applus +

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AC6, Navarre, Spain).

242

$$u(Z_g) = A \sqrt{\left[ \frac{L_2(T_1 - T_3)}{L_1^2 \cdot \dot{Q}} \right]^2 u^2(L_1) + \left[ \frac{T_3 - T_1}{L_1 \cdot \dot{Q}} \right]^2 u^2(L_2) + \left[ \frac{1}{\dot{Q}} - \frac{L_2}{L_1 \cdot \dot{Q}} \right]^2 u^2(T_1) + \left[ \frac{L_2}{L_1 \cdot \dot{Q}} \right]^2 u^2(T_3) + \left[ \frac{L_4(T_6 - T_4)}{L_3^2 \cdot \dot{Q}} \right]^2 u^2(L_3) + \left[ \frac{T_6 - T_4}{L_3 \cdot \dot{Q}} \right]^2 u^2(L_4) + \left[ -\frac{L_4}{L_3 \cdot \dot{Q}} \right]^2 u^2(T_4) + \left[ -\frac{1}{\dot{Q}} + \frac{L_4}{L_3 \cdot \dot{Q}} \right]^2 u^2(T_6) + \left[ -\frac{T_1}{\dot{Q}^2} + \frac{L_2 T_1}{L_1 \dot{Q}^2} - \frac{L_2 T_3}{L_1 \dot{Q}^2} + \frac{T_6}{\dot{Q}^2} - \frac{L_4 T_6}{L_3 \dot{Q}^2} + \frac{L_4 T_4}{L_3 \dot{Q}^2} \right]^2 u^2(\dot{Q})}$$

243

(12)

244

245 **EXPERIMENTAL RESULTS**

246 In all the tests, we have calculated the evolution over time of the global thermal impedance of the sample.

247 Subsequently, since it is the only term affected by aging, we can deduce the evolution over time of the TCR  
248 in the TIM.

249 Since the main objective is to evaluate the influence of aging on the TCR of the TIM, equal measurement  
250 conditions are used both in temperature and pressure for all samples. The boundary conditions are the aging  
251 temperature, the aging time, the sample temperature during the test ( $T_{av}$ ) and the pressure applied to the  
252 sample during the test.

253 The initial values of the global thermal impedance of the 12 samples can be seen in Table I, whereas those  
254 obtained after the aging process are presented below.

	$Z_{g(S-P-C)}$ [ $10^{-4}m^2C/W$ ]	$Z_{g(S-I-C)}$ [ $10^{-4}m^2C/W$ ]	$Z_{g(S-G-C)}$ [ $10^{-4}m^2C/W$ ]	$Z_{g(C-P-A)}$ [ $10^{-4}m^2C/W$ ]	$Z_{g(C-I-A)}$ [ $10^{-4}m^2C/W$ ]	$Z_{g(C-G-A)}$ [ $10^{-4}m^2C/W$ ]
60 °C	4.35	4.79	4.62	1.96	2.26	2.32
180 °C	4.29	4.74	4.61	1.85	2.38	2.24

255 Table I: Initial values of the global thermal impedance

256

257 **Aging with temperature of 180°C**

258 This section shows the evolution of the global thermal impedance as a function of time in the oven at 180°C,  
259 as well as the uncertainty associated with the value obtained, as can be seen in Figs. 8 and 9.

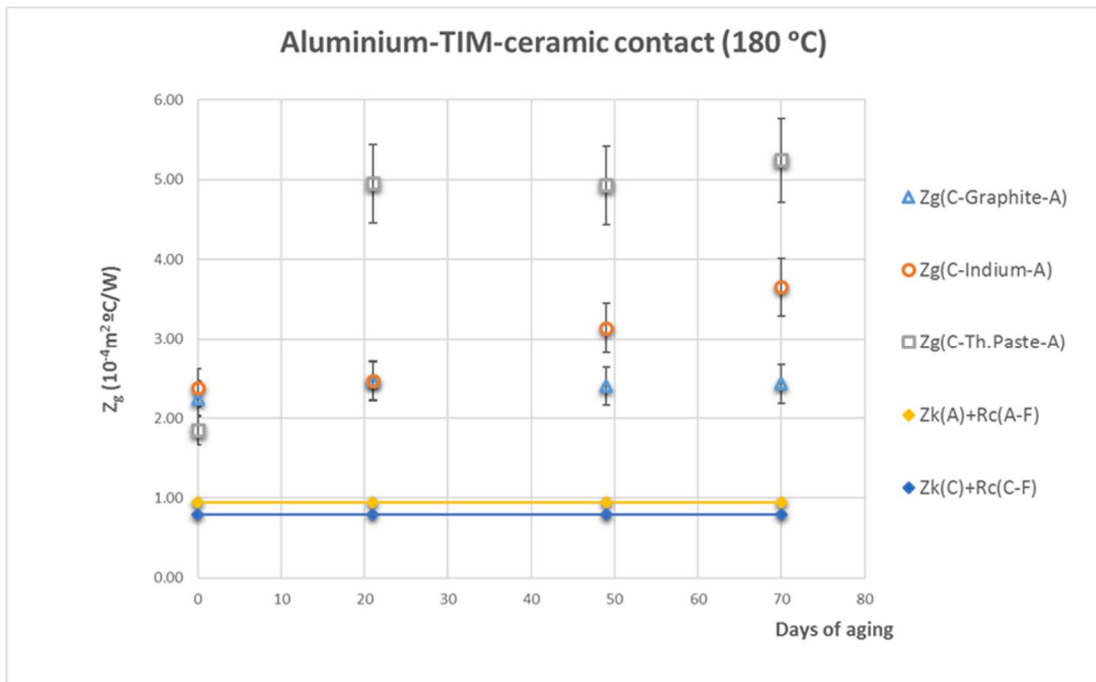


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Fig. 8. Steel-TIM-ceramic contact (180 °C)

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Fig. 9. Aluminium-TIM-ceramic contact (180 °C)

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In all the test, the global thermal impedance shows an increasing trend. In the case of the ceramic-paste-

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steel sample (S-P-C-180), just after 20 days of exposure to 180°C, the thermal impedance increases

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approximately by 200%. In the following periods, this trend continues but at lower rate. The total increase

269 in the 70 days of exposure is 300%. A slightly different pattern is obtained when the ceramic-paste-  
270 aluminum sample (C-P-A-180) is tested, which presents an increase of 167% in the thermal impedance  
271 after 20 days of exposure. Thereafter, this value remains virtually constant, with a total increase of 183%  
272 at the end.

273 The global thermal impedance in the combination of thermal paste and steel increases more significantly  
274 than in the combination with aluminum. This may be due to the fact that aluminum is softer than steel, and  
275 therefore may present a larger effective contact area with the ceramic. Consequently, the area occupied by  
276 the holes and covered by the thermal paste is smaller in the case of aluminum, and the aging of the thermal  
277 paste might be less influential.

278 In the case of the ceramic-indium-steel sample (S-I-C-180), the observed increase is much lower than in  
279 the previous cases. The thermal impedance presents a total increase of 25.5% after 70 days of exposure. In  
280 the tests with ceramic-indium-aluminum sample (C-I-A-180), the increase is 50%. Therefore, the  
281 performance of the indium is significantly better than that of the thermal paste.

282 The ceramic-graphite-steel (S-G-C-180) and ceramic-graphite-aluminum (C-G-A-180) samples have the  
283 best behavior at 180 °C, with a total increase in the thermal impedance of 8.48% and 8.50% respectively  
284 after 70 days of exposure, which are even lower than the measurement uncertainty of the test bench (10%).  
285 Therefore, it could be said that for long periods of more than 70 days of exposure to 180° C, graphite is the  
286 material with greater performance.

### 287 **Aging with temperature of 60°C**

288 The results of the thermal impedance after the tests with exposure temperature of 60°C are shown in Figs.  
289 10 and 11. These figures report minute variations in the thermal impedance of the samples with steel (S-P-  
290 C-60, S-I-C-60 and S-G-C-60), and also of the samples with aluminum (C-P-A-60, C-I-A-60 and C-G-A-  
291 60). In fact, these variations are even lower than the mentioned measurement uncertainty. Therefore, it can  
292 be concluded that there is not significant change in the thermal impedance of the samples when they are  
293 exposed to a temperature of 60 °C for 70 days.

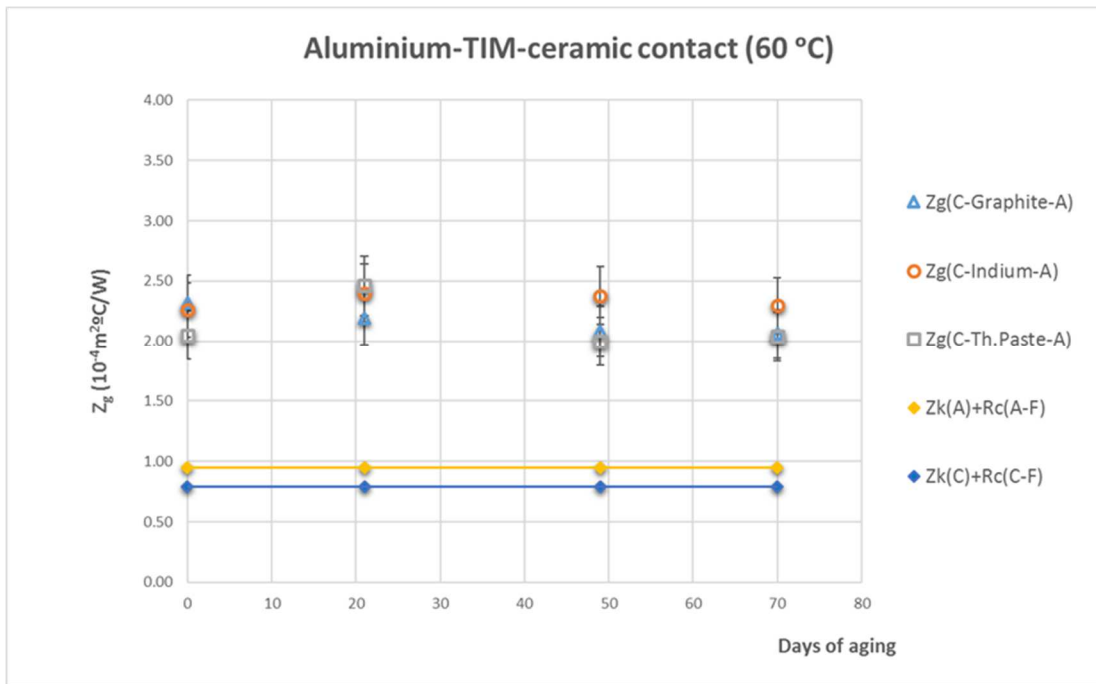


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Fig. 10. Steel-TIM-ceramic contact ( $60^\circ\text{C}$ )



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300 **CONCLUSIONS**

301 An increase in the thermal contact resistance due to TIM degradation entails a decrease in the performance

302 of a thermoelectric application. The test bench presented in this article is intended to evaluate the influence

303 of this thermal contact resistance and serve as guide for the selection of the TIM best suited for each  
304 thermoelectric application. The tests show that the degradation of TIMs makes it necessary to measure the  
305 evolution of the thermal contact resistance over time for different temperature conditions.

306 This paper presents a methodology for evaluation of the influence of TIM aging on the thermal contact  
307 resistance. The testing conditions mimic those of a thermoelectric generator that absorbs heat from the hot  
308 surface of a steel chimney, and releases heat to the ambient through an aluminum heat sink. Two contacts  
309 arise from this configuration: steel-ceramic contact between the chimney and the hot face of the modules;  
310 and ceramic-aluminum contact between the heat sink and the cold face of the modules. For both, three  
311 TIMs have been studied (graphite, indium, and polysynthetic thermal paste) under aging temperatures of  
312 180°C and 60°C, so a total of 12 tests have been conducted.

313 The thermal impedance  $Z_g$  is calculated experimentally, assuming that the only component of  $Z_g$  affected  
314 by aging is the thermal contact resistance inherent to the TIM.

315 The results with aging temperature of 180°C report a significant increase in the thermal impedance due to  
316 aging for the three TIMs, although this increase is much more significant when thermal paste is used.  
317 Therefore, the use of thermal paste as TIM for the hot side of thermoelectric modules should be considered  
318 with caution, due to the experimentally-proven aging of this material at temperatures around 180°C. As for  
319 the other two materials, graphite performs better against aging but indium offers better initial values of  
320 thermal contact impedance. In consequence, it is necessary to study whether the lower cost of graphite  
321 coming from its lower maintenance requirements (as indium must be replaced more frequently than  
322 graphite) counterbalances its higher initial thermal contact resistance compared to indium.

323 The tests at 180 °C carried out with steel indicate a greater increase in the thermal impedance compared to  
324 aluminum. This is explained by the larger hardness of steel, which entails a smaller effective contact surface  
325 and therefore a greater influence of the TIM. Thus, when the TIM degrades, it influences more significantly  
326 those materials with higher hardness, as is the case of steel compared to aluminum.

327 In the tests with 60°C of aging temperature, no variation in the thermal impedance has been found.  
328 Therefore, the use of thermal paste as TIM could be a good option at this temperature, as this material  
329 presents the lowest initial thermal contact resistance.



330 Future research is planned to complete this study. In particular, new experimental analyses are expected for  
331 thermal characterization of TIMs after cycles at different temperatures and corrosive atmospheres.

332

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### 337 **REFERENCES**

- 338 1. P. Jaiswal and C.K. Dwivedi, International Journal of Innovative Technology and Creative  
339 Engineering IJITCE, (2011). [http://ia800305.us.archive.org/34/items/IJITCE/IJITCE\\_May3.pdf](http://ia800305.us.archive.org/34/items/IJITCE/IJITCE_May3.pdf)  
340 Accessed 4 May 2011.
- 341 2. S. Narumanchi, M. Mihalic, K. Kelly and Gary Eesley, in IITHERM Conference Proceedings,  
342 (2008), doi:10.1109/IITHERM.2008.4544297
- 343 3. R. Prasher and CP. Chiu, in Materials for Advanced Packaging. ed. By D. Lu and C. Wong  
344 (Springer, 2017), p. 511.
- 345 4. I. Hu, M. Shih, G. Kao, in IMPACT'15 Conference Proceedings, (2015),  
346 doi:10.1109/IMPACT.2015.7365218
- 347 5. GK. Morris, MP. Polakowski, L. Wei, MD. Ball, MG. Phillips, C. Mosey and R.A. Lukaszewski,  
348 in IWIPD Conference Proceedings, (2015), doi: 10.1109/IWIPP.2015.7295991
- 349 6. J. Due and A. J. Robinson, Appl. Therm. Eng., 50, 455 (2013)
- 350 7. R. A. Sayer, T. P. Koehler, S. M. Dalton, T. W. Grasser, and R. L. Akau, in ASME 2013 Summer  
351 Heat Transfer Conference Proceedings, (2013), doi:10.1115/HT2013-17408
- 352 8. J.-P. Ousten and Z. Khatir, in EPE Conference proceedings, (2011), [https://hal.archives-](https://hal.archives-ouvertes.fr/hal-00628876)  
353 [ouvertes.fr/hal-00628876](https://hal.archives-ouvertes.fr/hal-00628876), Accessed 4 October 2011.
- 354 9. V. Khuu, M. Osterman, A. Bar-Cohen and M. Pecht, IEEE Transactions on Device and Materials  
355 Reliability, Vol. 9, N° 3, 379, (2009)
- 356 10. R. Skuriat, J.F. Li, P.A. Agyakwa, N. Matthey, P. Evans, C.M. Johnson, Microelectron. Reliab. 53,  
357 1933, (2013).
- 358 11. American Society for Testing and Materials, ASTM Standard D5470-06, (2006).

- 359 12. J. Liu, H. Feng, X. Luo, R. Hu, and S. Liu, in *Int. Conf. Electron. Packag. Technol. High Density*  
360 *Packag. ICEPT-HDP Conference Proceedings*, (2010), pp. 116–120.
- 361 13. W. Zongren, Z. Weifang, Y. Mingyuan, *Adv. Mater. Res.*, 337, 774 (2011)
- 362 14. R. A. Sayer, *Heat Transfer Eng.* (2015), doi:10.1080/01457632.2014.932553.
- 363 15. N. Goel, A. Bhattacharya, J.A. Cervantes, R.K. Mongia, S.V. Machiroutu, H.L. Lin, Y.C. Huang,  
364 K.H. Fan, B.L. Denq, C.H. Liu, C.H. Lin, C.W. Tien, J.H. Pan, in *Electron. Packag. Technol.*  
365 *Conf. Proceedings Conference*, (2008), doi:10.1109/EPTC.2008.4763637
- 366 16. M.C. Kumar Swamy and Satyanarayan, *J. Electron. Mater.* (2019), doi:10.1007/s11664-019-  
367 07623-7
- 368 17. T. Sakamoto, T. Iida, T. Sekiguchi, Y. Taguchi, N. Hirayama, K. Nishio and Y. Takanashi, *J.*  
369 *Electron. Mater.* (2014), doi:10.1007/s11664-014-3165-7.
- 370 18. A. Rodríguez, J.G. Vián and D. Astrain, *Appl. Therm. Eng.* (2009),  
371 doi:10.1016/j.applthermaleng.2009.03.005
- 372 19. A. Rodríguez, D. Astrain, A. Martínez, E. Gubía and F.J. Sorbet, *J. Electron. Mater.* (2013),  
373 doi:10.1007/s11664-013-2504-4
- 374 20. A. Rodríguez, D. Astrain, A. Martínez and P. Aranguren, *J. Electron. Mater.* (2014),  
375 doi:10.1007/s11664-014-3097-2
- 376 21. P. Aranguren, D. Astrain, A. Rodríguez and A. Martínez, *Appl. Energy* (2015),  
377 doi:10.1016/j.apenergy.2015.04.077
- 378 22. D. P. Bentz and K. R. Prasad. Rep. No. Building and Fire Research Laboratory (BFRL)-NIST  
379 7401, NIST, Gaithersburg, MD. Publisher: U.S. Department of Commerce (2007).