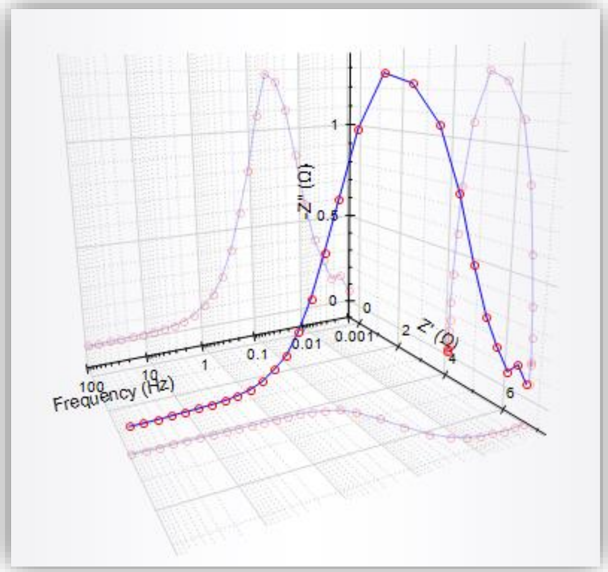
Applications of impedance spectroscopy in thermoelectricity



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Outline

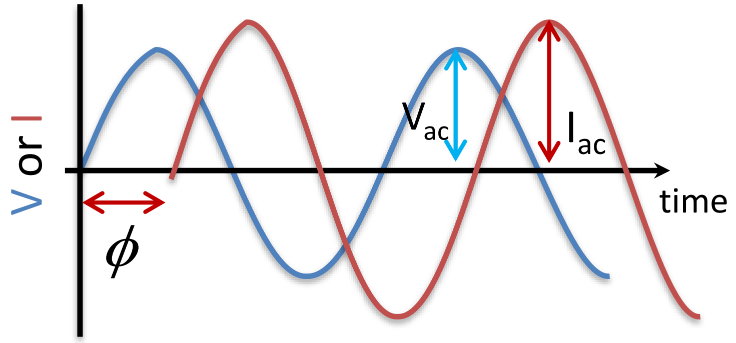
1. Introduction to the impedance method
2. Equivalent circuits for thermoelectricity
3. Module characterisation
4. Convection heat transfer coefficient determination
5. Thermal contact resistance determination
6. Acknowledgements

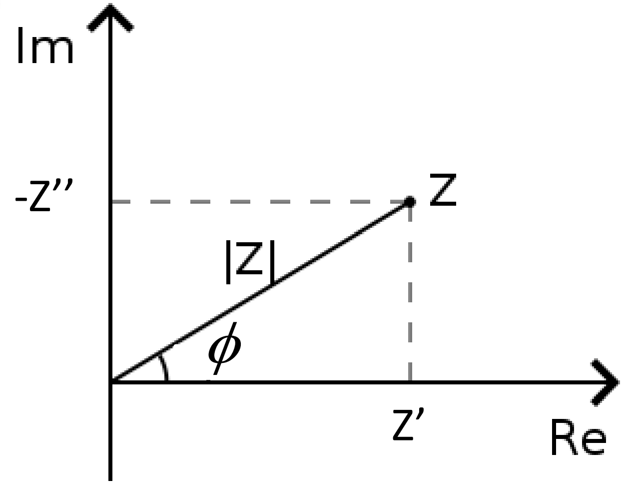
Why exploring impedance in thermoelectrics?

- It is **widely used** in a lot of different fields (solar cells, fuel cells, corrosion, supercapacitors, batteries, etc.).
- Powerful and very **reliable equipment** are **available in the market**.
- It allows the **separation** of the **physical processes** occurring in a device.

The perturbation and the system response

- A **small amplitude** sinusoidal **current wave** of a certain frequency is applied
- The system responds with a **voltage wave proportional** to the current that can be shifted in time (phase)





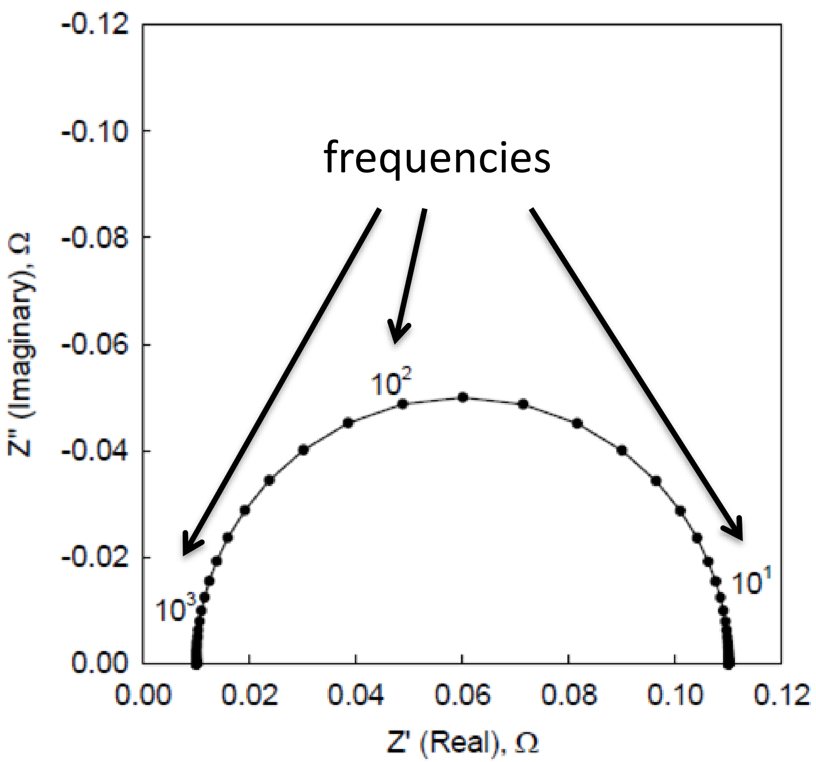
$$\frac{V_{ac}}{I_{ac}} = |Z| = \sqrt{Z'^2 + Z''^2}$$

$$\phi = arc\ \tan\left(\frac{Z''}{Z'}\right)$$

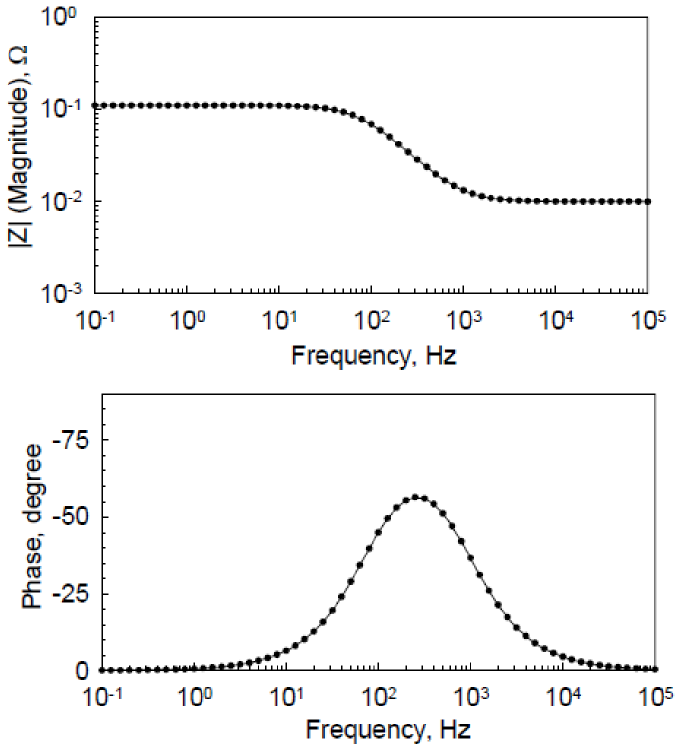
1. Introduction to the impedance method

The impedance spectrum

Z is obtained for a wide **range of frequencies** (e. g. 1 MHz to 10 mHz), obtaining one point in the spectrum per each frequency.



Impedance spectrum (Nyquist plot)



Parameters vs frequency (Bode plots)

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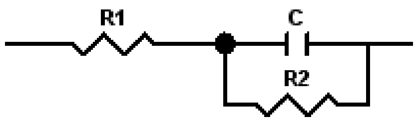
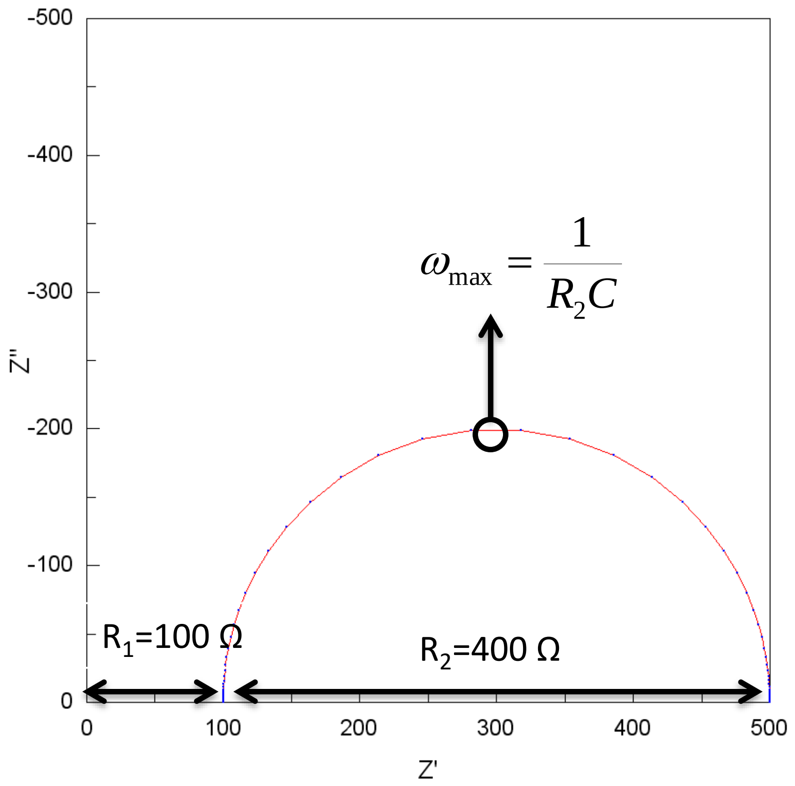
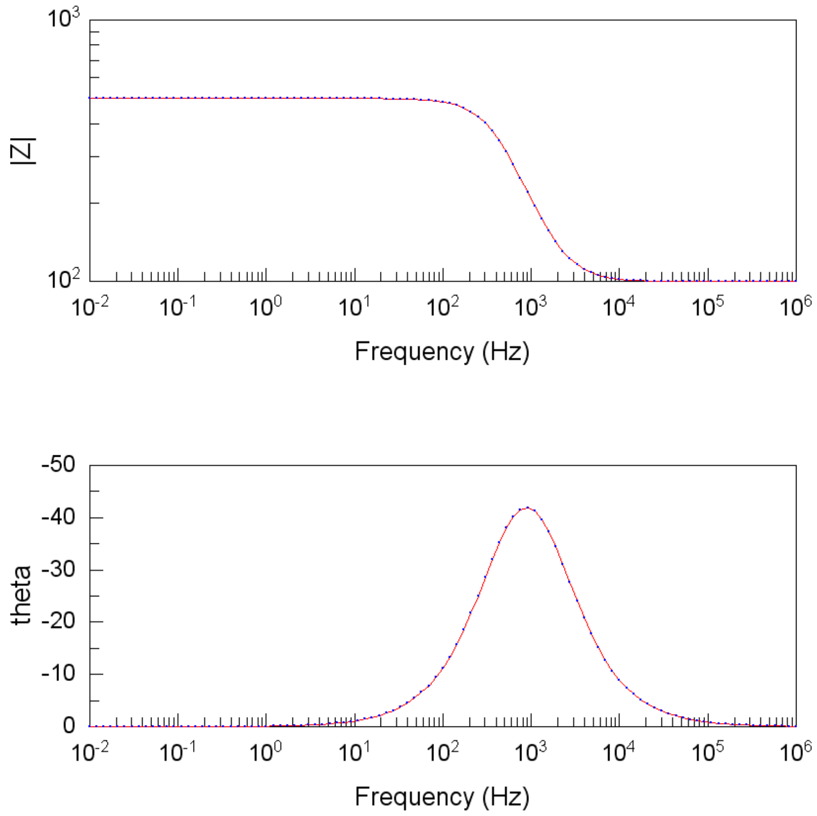
Equivalent circuits



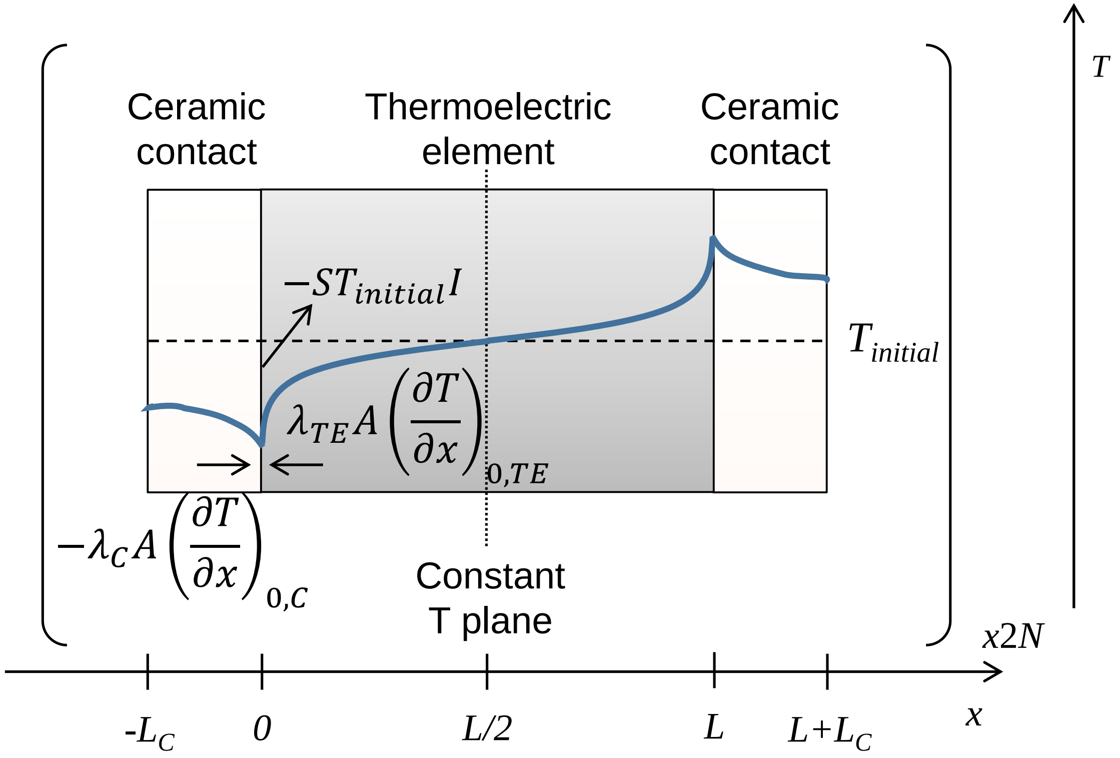
$$Z = R_1 + \frac{R_2}{1 + j\omega C R_2}$$





Model considerations

- **Module of 2*N* thermoelements with the typical architecture**.
- **Adiabatic conditions** (no heat exchanged with surroundings), i.e. suspended module in vacuum.
- All thermal and TE **parameters independent on temperature**.
- System is **initially at thermal equilibrium** at temperature $T_{initial}$.
- **Joule effect** is **neglected** both in the bulk and at the junctions.
- **Spreading-constriction** effects due to differences in areas between legs and ceramics is discarded.
- Thermal influence of the **Cu contacts** is **neglected**.



2. Equivalent circuits in thermoelectricity

Impedance function

$$V = IR_\Omega + S\left[T(L) - T(0)\right]$$

$$Z(t) = \frac{V}{I} = R_\Omega + \frac{S\left[T(L) - T(0)\right]}{I}$$

time domain (t)

$\mathcal{L}\{\Delta T\}=\theta \quad \mathcal{L}\{I\}=i_0$

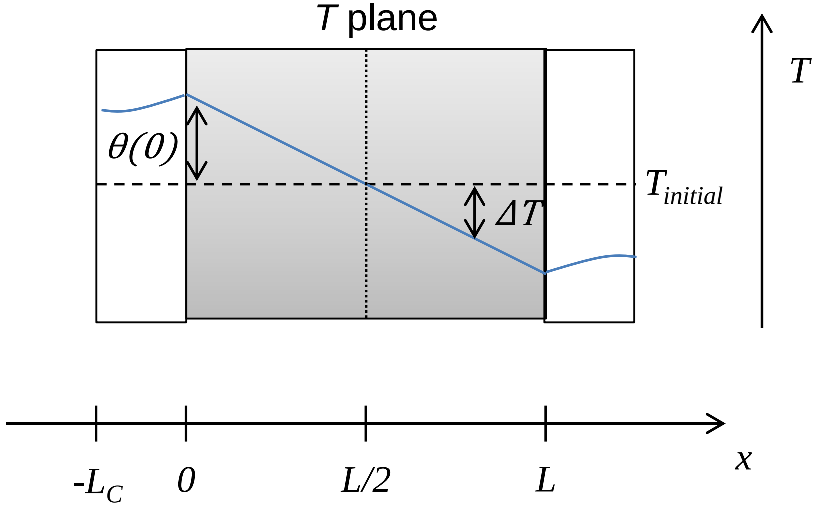
$\left[T(L) - T(0)\right] \rightarrow -2\theta(0)$

$$Z(j\omega) = R_\Omega - \frac{S2\theta(0)}{i_0}$$

frequency domain (*jω*)

To know the impedance function we **need to know the T difference** at *x*=0 as a function of frequency

Solve heat equation

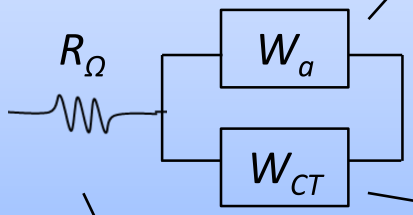


$V=V(0)-V(L)$, R_Ω=total ohmic resistance, $\omega=2\pi f$, f is the frequency, $j=\sqrt{-1}$

2. Equivalent circuits in thermoelectricity

Equivalent circuit elements

$$Z = R_\Omega + \left(\frac{1}{Z_{W_{CT}}^{-1} + Z_{Wa}^{-1}} \right)$$



R_Ω is the total ohmic resistance

$$R_\Omega = 2N\rho\frac{L}{A} + R_{parasitic}$$

W_{CT} is a Constant *T* Warburg

$$Z_{W_{CT}} = R_{TE}\left(\frac{j\omega}{\omega_{TE}}\right)^{-0.5} \tanh\left\{\left(\frac{j\omega}{\omega_{TE}}\right)^{0.5}\right\}$$

$$R_{TE} = 2N\frac{S^2 T_{initial} L}{\lambda_{TE} A}$$

Thermoelectric resistance

$$\omega_{TE} = \frac{\alpha_{TE}}{(L/2)^2}$$

Characteristic frequency

W_a is an adiabatic Warburg

$$Z_{Wa} = R_C\left(\frac{j\omega}{\omega_C}\right)^{-0.5} \coth\left\{\left(\frac{j\omega}{\omega_C}\right)^{0.5}\right\}$$

$$R_C = 4N\frac{S^2 T_{initial} L_C}{\lambda_C A}$$

Thermoelectric resistance from ceramic

$$\omega_C = \frac{\alpha_C}{(L_C)^2}$$

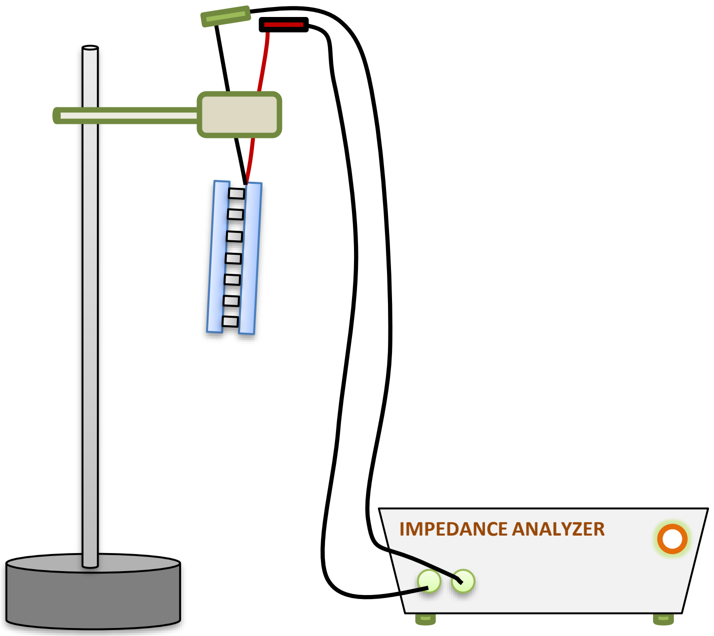
Characteristic frequency from ceramic

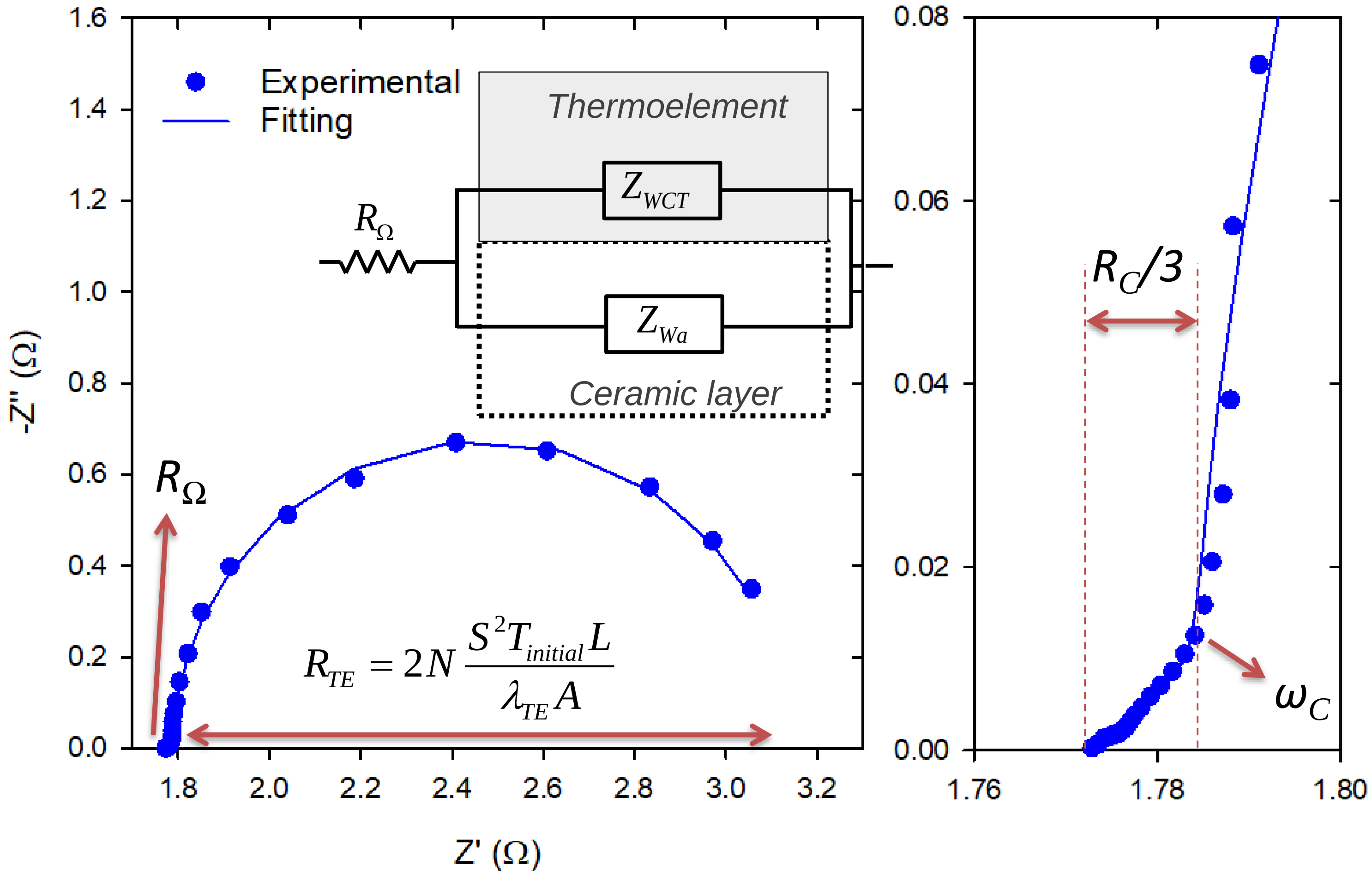
The **fitting** of experimental measurements to the equivalent circuit **provides**: R_Ω, R_{TE}, ω_{TE}, R_C and ω_C.
The resistances contain all the **interesting TE properties** (*S* and *λ* of the legs and the R_Ω of the module).

From the thermoelement: *S*=Seebeck coefficient, *ρ*=electrical resistivity, α_{TE}=thermal diffusivity, λ_{TE}=thermal conductivity
From the contact: α_C=thermal diffusivity, λ_C=thermal conductivity

3. Module characterisation

Bi-Te commercial module suspended in vacuum (<10^{-4} mbar) at room T





3. Module characterisation

Extracted parameters

The **Seebeck coefficient and thermal conductivity** can be **extracted if the thermal conductivity of the ceramic is known**. The module **internal resistance** and the **ZT** can be **directly** extracted.
All properties show **good agreement** with values of the manufacturer.

$$R_\Omega = 2N\rho\frac{L}{A} + R_{parasitic}$$

$$R_C = 4N\frac{S^2 T_{initial} L_C}{\lambda_C A} = 0.036\ \Omega$$

λ_C=37 W/Km

$$R_{TE} = 2N\frac{S^2 T_{initial} L}{\lambda_{TE} A} = 1.33\ \Omega$$

$$(ZT)_{module} = \frac{R_{TE}}{R} = \frac{2NS^2 T_i L}{\left(R_p + \frac{2NL}{\sigma A}\right)\lambda_{TE} A}$$

R_Ω=1.75 Ω

S=175 μV/K

λ_{TE}=1.44 W/Km

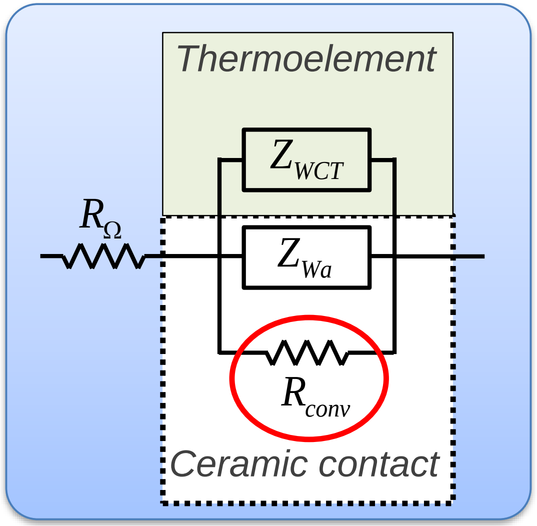
$(ZT)_{module}$=0.76

Suitable method for module properties characterisation

J. García-Cañadas, G. Min, Impedance spectroscopy models for the complete characterization of thermoelectric materials, ***J. Appl. Phys.*** *116* (2014) 174510

4. Convection heat transfer coefficient determination

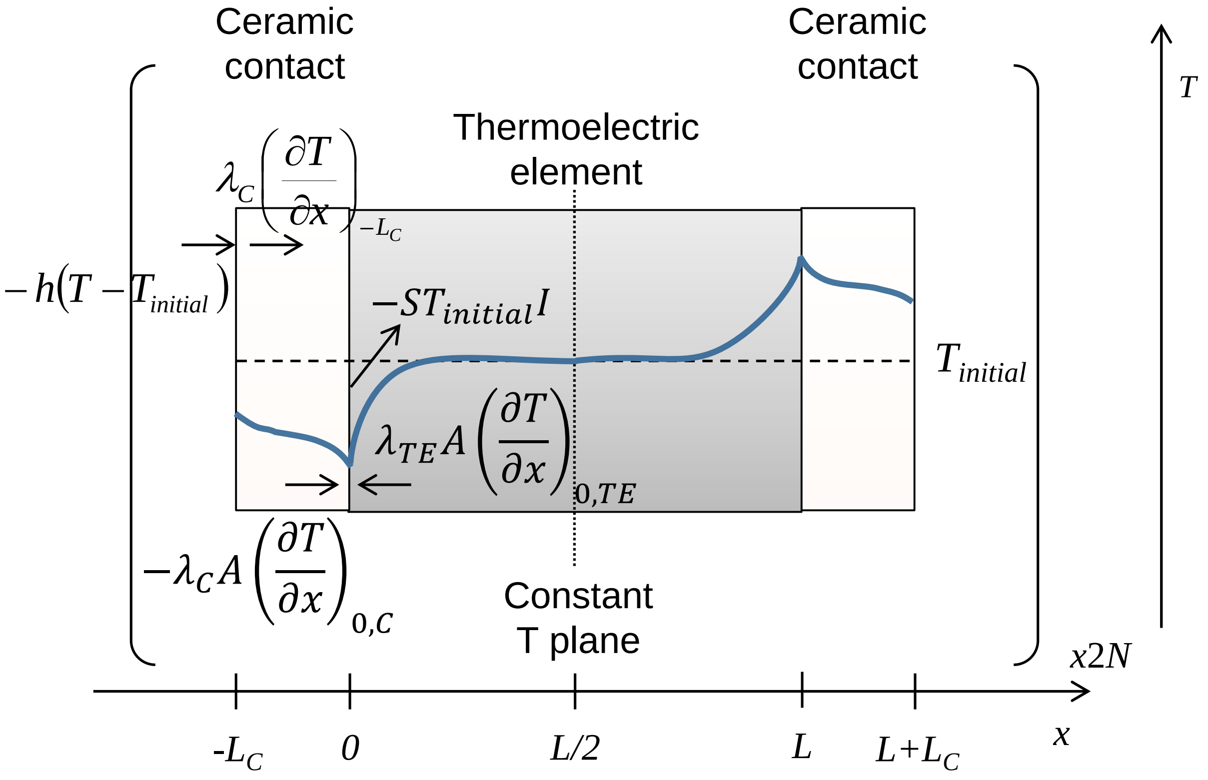
Model considerations



$$R_{conv} = 4N \frac{S^2 T_{initial} \eta}{hA}$$

***h*=convection heat transfer coefficient**

η=Total area of TE legs/ceramic plate area



4. Convection heat transfer coefficient determination

Extracted parameters

The **convection resistance** can be **obtained** from a fitting to the suspended module in vacuum and other fitting in air (or the condition to be evaluated). From R_{conv}, ***h* can be obtained** if S is known.

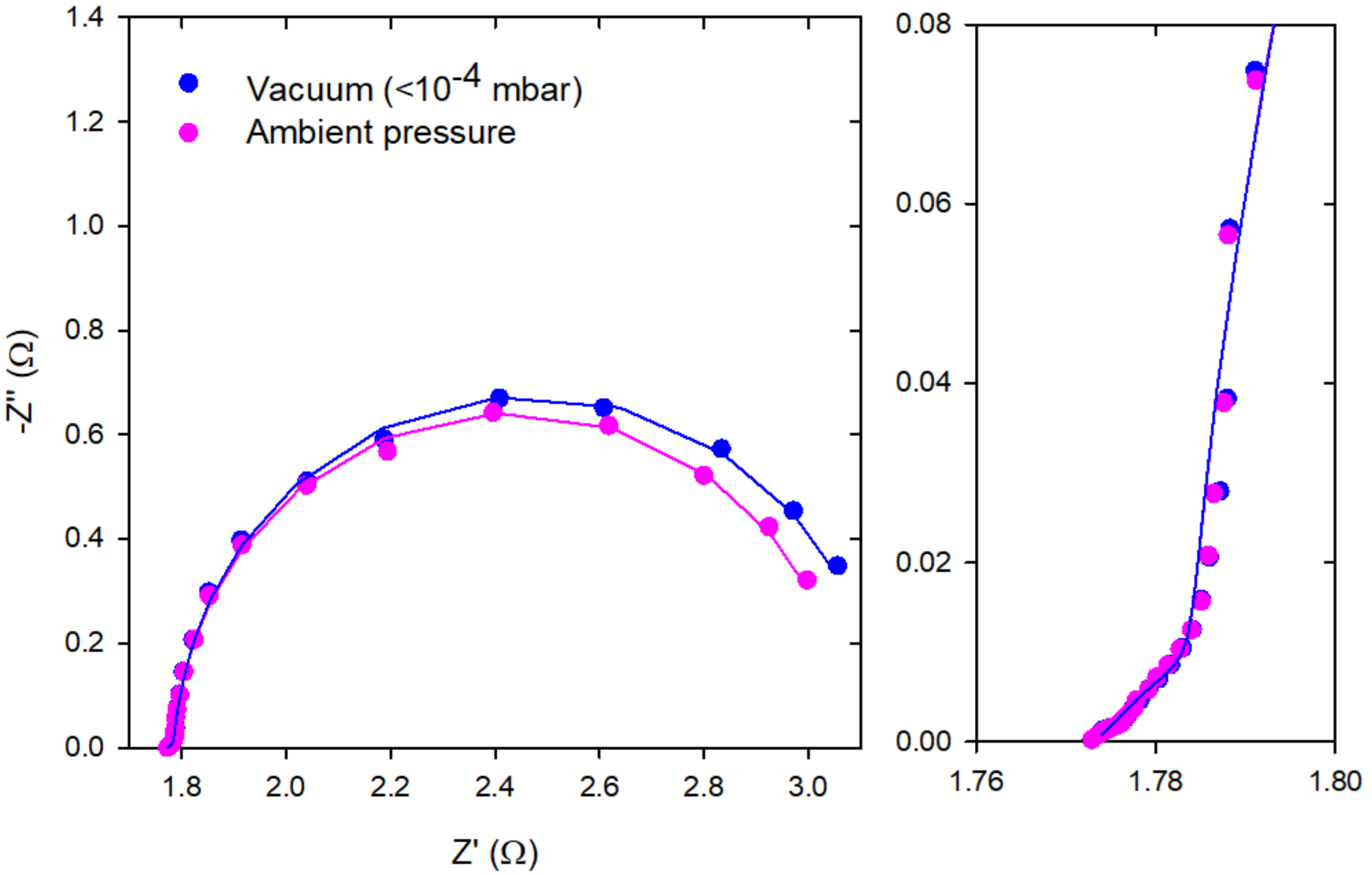
S=192 μV/K

$$R_{conv} = 4N\frac{S^2 T_{initial}\eta}{hA} = 21.75\ \Omega$$

***h*=40.12 W/(m²K)**

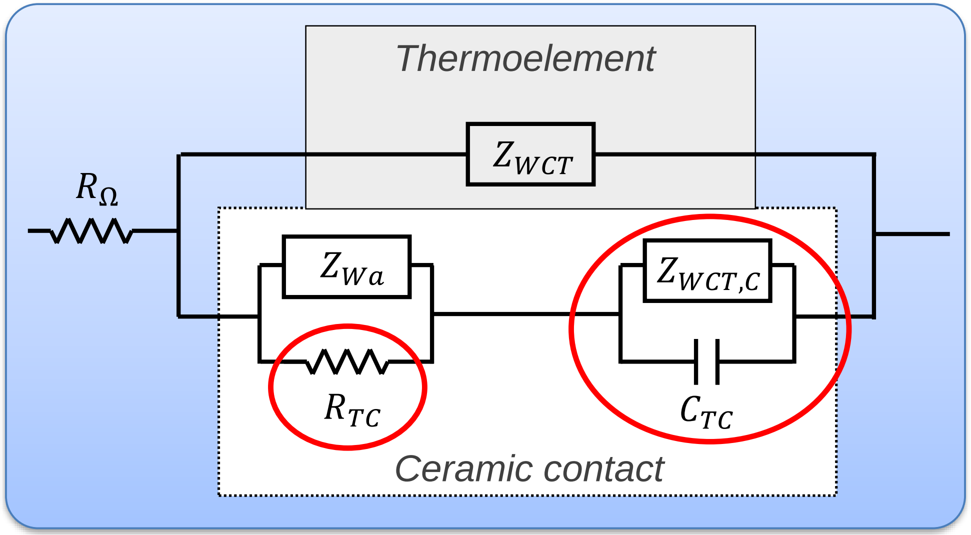
Similar order of magnitude as natural convection 5-25 W/(m²K). Potential ***h* sensor application.**

Higher accuracy [21.6 W/(m²K)] was reached considering spreading-constriction effects [see Mesalam et al. Applied Energy 226 (2018) 1208]

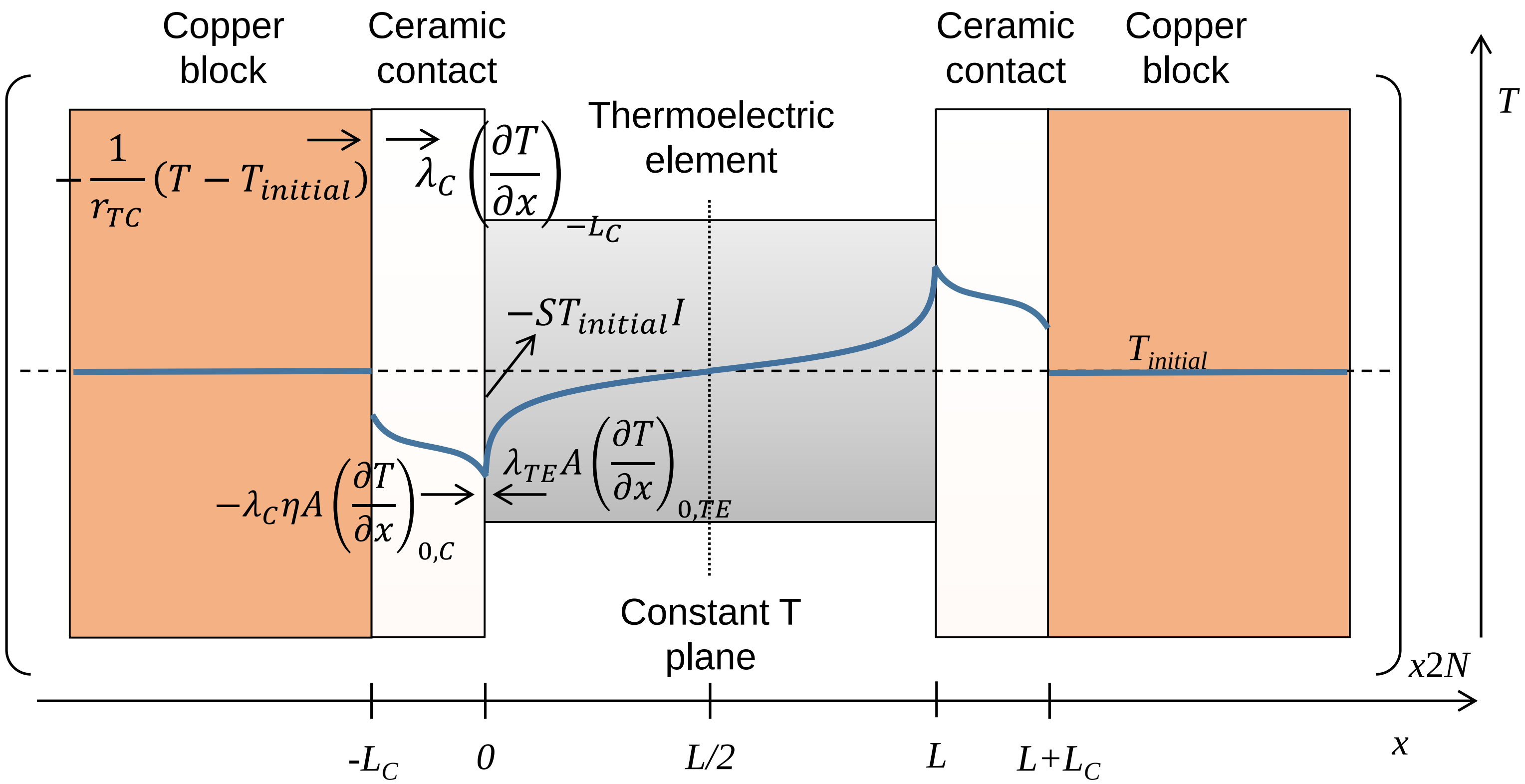


B. Beltrán-Pitarch and J. García-Cañadas. Influence of convection at outer ceramic surfaces on the characterization of thermoelectric modules by impedance spectroscopy. **J. Appl. Phys.** 123 (2018) 084505.

5. Thermal contact resistance determination



Model considerations



$$R_{TC} = \frac{4NS^2 T_{initial} r_{TC} \eta}{A}$$

$$Z_{C_{TC}} = \frac{4NS^2 T_{initial} \eta}{\lambda_C \rho_C C_{p,C} A r_{TC}} \frac{1}{j\omega}$$

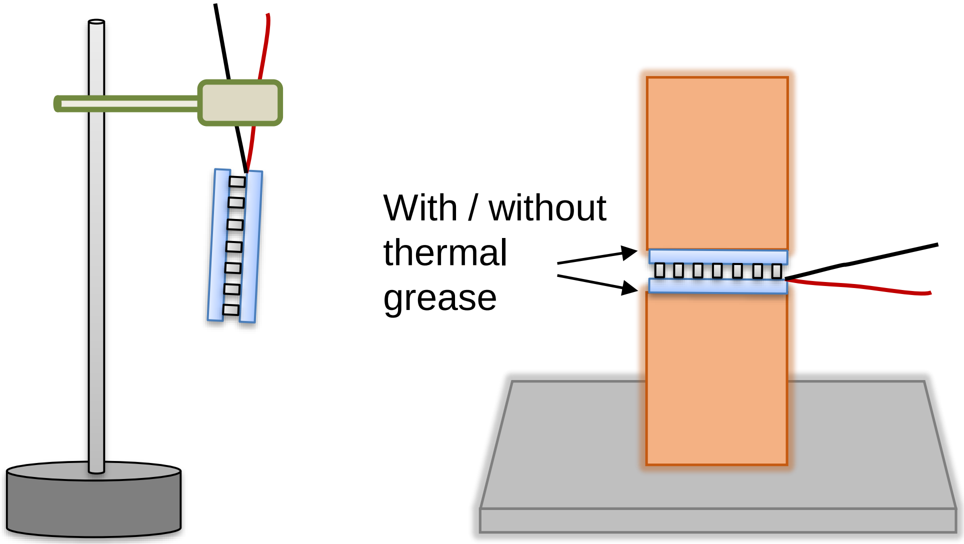
Thermal contact resistance

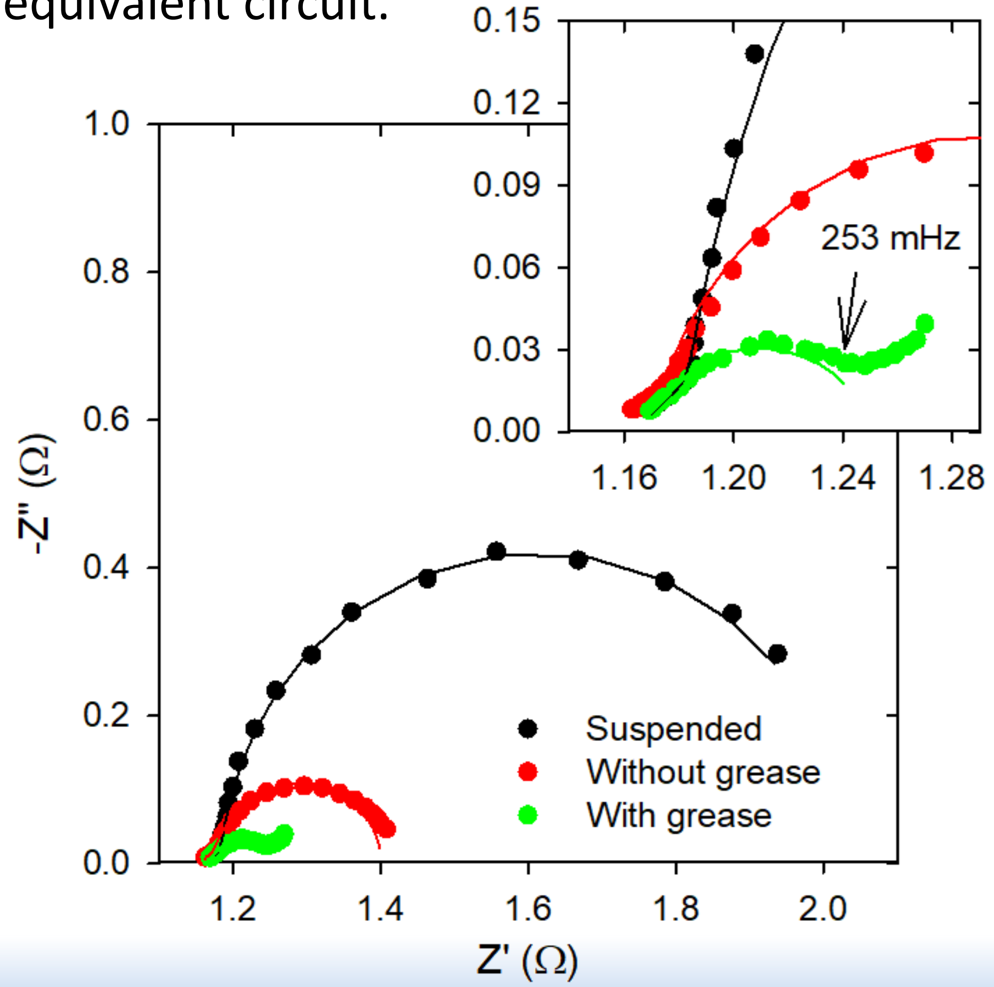
$$Z_{WCT,C} = \frac{4NS^2 T_{initial} L_C \eta}{\lambda_C A} \left(\frac{j\omega}{\omega_C}\right)^{-0.5} tanh\left[\left(\frac{j\omega}{\omega_C}\right)^{0.5}\right]$$

5. Thermal contact resistance determination

Experimental results

The impedance response significantly **changes with the thermal contact resistance** value. Good fittings (lines) to the equivalent circuit.





5. Thermal contact resistance determination

Extracted parameters

The R_{TC} can be obtained from a fitting to the **module in vacuum and in the contacted module setup**. The **thermal contact resistance** can be obtained from R_{TC} **if the Seebeck coefficient** of the module **is known** (S=186.42 µV/K).

	R_{Ω} (Ω)	R_{TE} (Ω)	ω_{TE} (rad s^{-1})	R_C (Ω)	ω_C (rad s^{-1})	R_{TC} (Ω)	r_{TC} (m^2mKW^{-1})
Suspended in vacuum	1.16	0.869	0.392	0.0812	5.48	---	---
Without thermal grease	1.15	---	---	---	---	0.259	0.311
With thermal grease	1.16	---	---	---	---	0.012	**0.014**

The obtained value is **in agreement with literature** results.
Impedance is an **excellent tool to monitor** and evaluate the influence of the **thermal contact**.

B. Beltrán-Pitarch, J. García-Cañadas. Characterization of the thermal contacts between heat exchangers and a thermoelectric module by impedance spectroscopy (under preparation).

6. Acknowledgements

Spanish Agencia Estatal de Investigación under the Ramón y Cajal program (RYC-2013-13970).

Generalitat Valenciana and the **European Social Fund** under the ACIF program (ACIF/2018/233).

Universitat Jaume I under the project UJI-A2016-08.

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