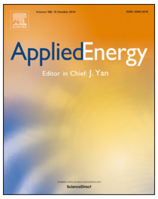
Applied Energy 226 (2018) 1208–1218

Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy



Towards a comprehensive model for characterising and assessing thermoelectric modules by impedance spectroscopy

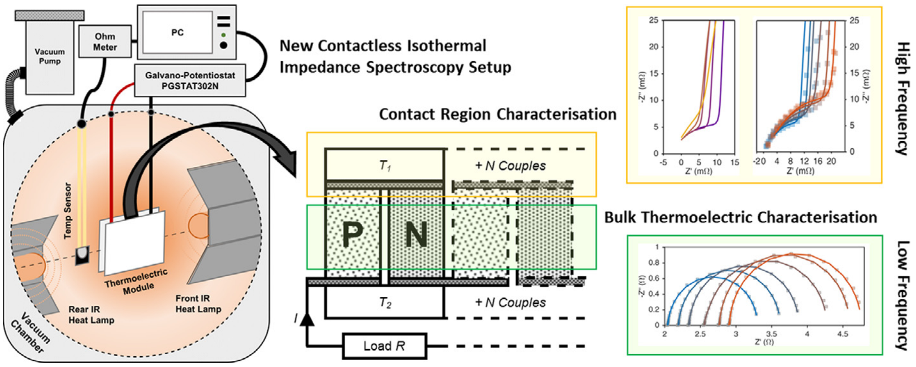
Ramy Mesalam[a,*], Hugo R. Williams[a], Richard M. Ambrosi[b], Jorge García-Cañadas[c], Keith Stephenson[d]

[a] Department of Engineering, University of Leicester, Leicester LE1 7RH, UK
[b] Department of Physics & Astronomy, University of Leicester, Leicester LE1 7RH, UK
[c] Department of Industrial Systems Engineering and Design, Universitat Jaume I, 12071 Castellón, Spain
[d] European Space Agency, ESTEC TEC-EP, Keplerlaan 1, 2201AZ Noordwijk, The Netherlands

HIGHLIGHTS

- New impedance model developed including all key phenomena.
- A complete parametric analysis of each phenomena's influence.
- Complete module characterisation of all key temperature dependent properties.
- Convection coefficient measured accurately with a thermoelectric module.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Thermoelectric module
Impedance spectroscopy
Thermoelectric characterisation
Thermoelectric generator
Thermoelectric cooling

ABSTRACT

Thermoelectric devices have potential energy conversion applications ranging from space exploration through to mass-market products. Standardised, accurate and repeatable high-throughput measurement of their properties is a key enabling technology. Impedance spectroscopy has shown promise as a tool to parametrically characterise thermoelectric modules with one simple measurement. However, previously published models which attempt to characterise fundamental properties of a thermoelectric module have been found to rely on heavily simplified assumptions, leaving its validity in question. In this paper a new comprehensive impedance model is mathematically developed. The new model integrates all relevant transport phenomena: thermal convection, radiation, and spreading-constriction at junction interfaces. Additionally, non-adiabatic internal surface boundary conditions are introduced for the first time. These phenomena were found to significantly alter the low and high frequency response of Nyquist spectra, showing their necessity for accurate characterisation. To validate the model, impedance spectra of a commercial thermoelectric module was experimentally measured and parametrically fitted. Technique precision is investigated using a Monte-Carlo residual resampling approach. A complete characterisation of all key thermoelectric properties as a function of temperature is validated with material property data provided by the module manufacturer. Additionally, by firstly characterising the module in vacuum, the ability to characterise a heat transfer coefficient for free and forced convection is demonstrated. The model developed in this study is therefore a critical enabler to potentially allow impedance spectroscopy to

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[*] Corresponding author.
E-mail address: rm467@le.ac.uk (R. Mesalam).

https://doi.org/10.1016/j.apenergy.2018.05.041
Received 31 January 2018; Received in revised form 17 April 2018; Accepted 6 May 2018

characterise and monitor manufacturing and operational defects in practical thermoelectric modules across multiple sectors, as well as promote new sensor technologies.

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1. Introduction

Thermoelectric (TE) materials can directly convert heat into electricity. The TE efficiency in materials is evaluated using a dimensionless figure-of-merit $ZT = S^2T/\rho\lambda$, which comprises of absolute temperature T and three fundamental TE properties: Seebeck coefficient S, electrical resistivity ρ (or electrical conductivity σ) and thermal conductivity λ. While literature shows maturity in measuring TE properties on a material level [1], there is not an international test standard for measuring these properties at a device level. Solving this widely recognized issue [2] is crucial, as device level characterisation is needed to validate the implementation of an ever-growing number of novel TE materials [3], guide system design [4], direct quality control and monitor device longevity. It is therefore critical for the development and application of TE technology to develop standardised, accurate and repeatable high-throughput characterisation methods.

Measurement methods currently used mainly rely on either a heat flow meter method or some variation of the Harman technique [5]. While these two methods have gained popularity [6,7] they each have clear disadvantages limiting their use. These include, (*i*) the need for intricate and expensive experimental setups for accurately measuring heat flows with minimal thermal losses, (*ii*) requires multiple measuring systems to be able to characterise all fundamental TE properties [5], (*iii*) are known to potentially feature large uncertainties which can reach up to 50% in some cases [8]. Recently, impedance spectroscopy (IS) analysis has been explored to not only lower uncertainties in the calculation of TE module zT [9,10] but also gain additional information on the three fundamental TE properties without the need of multiple measuring systems [11,12]. In fact, commonplace laboratory equipment such as a lock-in amplifier or a galvano-potentiostat equipped with a frequency-response analyser, is sufficient for room temperature characterisation. IS achieves this by measuring the voltage-current ratio (impedance) across the system as a function of frequency (impedance spectrum) when a small amplitude ac signal is applied under initial steady state conditions. To evaluate device characteristics, a theoretically derived impedance function, representing a collection of physical processes occurring in the system (equivalent circuit), is typically fitted to the experimental impedance spectrum for property extraction. Currently, literature reports various theoretical models which attempt to characterise TE devices under suspended conditions. Initially, Downey et al. proposed the use of a simple RC one-port circuit analogy and a transmission line model [13]. On the other hand, De Marchi et al. [14,15] and García-Cañadas et al. [16,17] later analysed the one-dimensional heat balance equation and performed a more detailed impedance analysis in the complex plane (Nyquist plot). The latter group were the first to explore the potential of IS for providing a complete characterisation of TE modules [11] although neglecting several key effects which take place in TE devices.

Other models, however, were also developed to more specifically analyse the influence of these important effects on the impedance response of a TE module. First Casalegno et al. [18] introduced the effect of spreading-constriction on heat fluxes [19] due to area differences between the TE legs and the external ceramic layers. Later, De Marchi et al. [14] added convection at the outer surfaces of the ceramics in conjunction with the spreading-constriction effect but did not analyse the effect of convection in the impedance response. This has been recently performed by Beltran-Pitárch et al. [20], although only considering free convection. Radiative losses are also important and were first considered by Downey et al. on a single TE element, correcting previously underestimated ZT values by up to 40% [13]. Consequently, large uncertainties as similarly experienced with the Harman method [21] could be present under certain characterisation conditions. While all these models improved the analysis of the impedance response and in some cases the ability to estimate ZT, there is yet a single comprehensive model which includes all these relevant effects with the capability to provide a complete characterisation of TE modules. Consequently, the underlying effect these additional phenomena have on characterising all the fundamental TE properties of a module have yet to be outlined.

The aim of this work is to develop a new single impedance model that can characterise all fundamental TE properties of a module as a function of temperature with accuracy and confidence suitable for practical use. This is made possible, for the first time, by integrating all relevant phenomena: thermal spreading-constriction at layer interfaces and internal-external surface heat losses via convection and radiation, under one easily implementable analytical model. The variations produced in the impedance response due to the introduction of each phenomena are analysed and more importantly their underlying effect on characterising TE properties is reported. In particular, the sensitivity of the impedance response to convective effects is focused upon and the possible use of TE modules as convective heat transfer coefficient sensors, for free and forced fluid flows, is demonstrated using the new model. Such a sensor would be highly desirable and beneficial to a variety of thermofluidic systems which require the characterisation or monitoring of convective heat transfer [22,23]. Validation of the model is performed by experimentally measuring the impedance spectra of a commercial Bi_2Te_3-based TE module over various isothermal temperatures using a new contactless infrared heating arrangement. A comparison of fitted TE properties with material property data provided by the module manufacturer is reported and fitting errors are investigated using a Monte-Carlo residual resampling approach (bootstrapping). Good agreement and relatively low errors indicates that IS characterisation is a viable technique for the assessment of TE devices in energy applications and more.

2. Theoretical model

2.1. Mathematical formulation

A typical TE module formed of $2N$ legs (N couples) of lengths L with cross-sectional area A, connected electrically in series and thermally in parallel, and sandwiched by two electrically isolating ceramic layers is considered (see Fig. 1a). It is assumed that the device is suspended vertically (TE legs rest horizontally) in air, the ambient temperature T_∞ does not vary with time and the initial homogenous temperature of the device T_i is in thermal equilibrium with its environment ($T_i = T_\infty$). The total voltage difference across the device is therefore given by the contribution of the total ohmic resistance R_Ω (comprising of the intrinsic electrical resistance of the TE legs R_i, and parasitic resistances R_p, which includes contributions from the copper strips, wires and contact resistance R_c) and the total Seebeck voltage from $2N$ TE legs when subjected to a temperature difference $\Delta T = T_L - T_0$, with T_L and T_0 being the temperatures at positions $x = L$ and $x = 0$, respectively. Therefore, the measured electrical impedance $Z = V/I$ in the time domain t can be summarised as $Z(t) = R + 2NS\Delta T/I$ [11].

Assuming the module is excited in the small signal regime (≤ 10 mA), the contribution of residual Joule heat can be ignored in comparison to the generated Peltier heat [10]. Therefore, under suspended conditions, the temperature at the half length L_h of the TE legs does not change (see Fig. 1b), consequently $\Delta T = -2(T_0 - T_i)$.