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# Integration of thermoelectric generators in a biomass boiler: Experimental tests and study of ash deposition effect

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#### ARTICLE INFO

### ABSTRACT

Keywords: Thermoelectric generator Biomass Boiler Heat transfer Ash Deposition Thermoelectric generators (TEGs) are based on Seebeck effect and produce electricity from a heat flow. They require low maintenance and provide quiet operation. However, they have low efficiency and need suitable temperature values. Accordingly, the integration of TEGs in boilers is a promising approach, since these equipment present high temperature differences between hot gases and water and allows the use of waste heat for water heating. There are experiences of the integration of TEGs in biomass boilers, but almost none with unconventional biomass with a high tendency to ash deposition. The integration of TEG modules in a commercial pellet-fired 25 kW<sub>th</sub> boiler is described and experimental results, working with pine pellets and with an agropellet (70% of vineyard pruning and 30% of barley straw), are presented. Power produced with only 6 modules can reach 60 W. After quantifying the effect of the temperature on both gas and water sides, the study focuses on analyzing the ash deposition on the exchange surfaces and their effect on TEG modules operation (e.g. power reduction of 0.24 W per hour with pine and 0.78 with agropellet). This effect can be minimized by working with high excess of air or by integrating a cleaning system.

# 1. Introduction

Biomass can play a relevant role as an energy source for domestic sector if it is used in small and medium scale boilers ( $<500 \text{ kW}_{th}$ ). Most of these boilers consume wood pellets and they can achieve high efficiency with low emissions by using carefully designed burners and even with extended heat transfer surfaces that allow condensation of part of the water contained in flue gases. Performance of these boilers could even be improved by integrating thermoelectric generators (TEGs), which would produce electricity that could be used for self-consumption of the boiler itself and even be exported for other uses within the house.

Thermoelectric generators are based on Seebeck effect and produce electricity from a heat flow. They have several advantages for their use in domestic applications: they are reliable, operate without noise, need almost no maintenance and can be scalable. Their main disadvantages are low efficiency and high cost [1]. Efficiency is not so relevant when low temperature heat from TEGs cooling is used for another purpose. The cost of TEG modules used in this research has values around 2–3  $\notin$ /W; this means that for an electricity price of 0.25  $\notin$ /kWh, around 10, 000 h of operation would be required only for the recovery of the investment cost of the devices. However, it should be noted that TEGs

prices tend to decrease and, furthermore, the use of TEGs for driving electric self-consumption of boiler allows its application in locations where electric grid is not available (e.g. livestock facilities).

Integration of TEG modules in biomass boilers has been tackled by several authors. A review of TEGs technology and its use for domestic applications was developed by Kutt et al. [2]; the paper presents thermoelectric generators, power electronic converters, applications in domestic boilers (both biomass and gas fired) and use in biomass stoves. Tohidi et al. have made a review of TEGs characteristics and applications that includes fundamentals of TEGs and materials, analysis at both module and system level and applications in several fields: waste heat recovery, photovoltaic thermoelectric hybrid systems, solar thermoelectric generators, off-grid power generation in deprived locations, portable micro-power generation, space applications and integration with fuel cells and in power plants [3]. A review focusing on the integration of TEG modules in biomass stoves for being used in poor regions was made by Najjar and Kseibi [4]. Alanne et al. have developed a numerical simulation of a 20 kWth pellet boiler in which TEGs have been introduced in the combustion chamber and in the convection tubes [5]. A pellet fired stove has been modified by Moser et al. by integrating water cooled TEGs that provided a total maximum electric power of 168

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W while consuming 12 kW of fuel [6]. Höftberger described an improved design that provided 345 W [7]. A stove with heat accumulation capabilities was developed by Sornek et al. which includes a water cooled TEG module that provides 6 W [8]. The same research group compared three different integrations of TEGs in a biomass stove: two of them located in the flat surface (with air and water cooling) and other on the flue gases duct; power produced varied from 45 to 230 W [9]. Brádzil et al. have integrated a flue gas energy harvester in a 25 kW<sub>th</sub> biomass boiler that includes four TEG modules that produced an electric power of 11 W [10,11]. Integration of four water cooled TEG modules in a biomass stove was analyzed by Montecucco et al. who obtained an average power of 27 W [12]. Sakdanupphab and Sakulkalavek modified a stove in order to integrate a TEG which is cooled with water through a heat pipe [13]. Use of TEG modules and a thermosyphon for power generation from waste heat from a biomass engine was developed by Goswami and Das [14]. Li et al. used a heat collector to direct heat from a biomass stove towards TEGs that produced 51 W [15]. Guoneng et al. present a micro-combined heat and power system that combines a stove with 144 TEG modules that provide 200 W electric power [16]. Sornek and Papis-Fraczek performed both numerical and experimental analyses of the integration of a TEG device in a wood stove that provides a maximum power of 15.9 W [17].

In these previous experiences, biomass with low tendency to ash deposition has been used. However, to achieve the objectives set by EU for the use of biomass in the domestic sector it is necessary to draw on others unconventional biomasses: energy crops and residual agricultural biomass (both woody and herbaceous) [18]. Thermal conversion of these biomasses presents clear differences respect to traditional biomass, mainly related with the characteristics of their ashes, both quantity (much greater) and elemental composition which implies greater tendency to sintering and deposition [19–23].

Within these phenomena, the deposition, that is to say the accumulation of part of the ash from the fuel on the heat exchange surfaces of the boiler, is especially important in this case. So, the deposition of ash on the heat exchange surfaces of the TEG modules integrated in the boiler could influence its behaviour: the deposited ash acts as thermal insulator, worsening the transfer of heat [24] and, therefore, decreasing temperature of hot side and, finally, the power produced. Thus, it is very interesting to analyze the effect of ash deposition and find operational strategies, both primary (reduce the severity of the phenomenon, acting on the operating parameters [25,26]) and secondary (removing deposits [27,28]), that improve the operation and optimize the power of TEG modules.

In this paper, an example of integration of TEG modules in a biomass boiler is presented. Only six modules, although there could be many more in a commercial boiler, of three types have been integrated. The facility is aimed at the research of the operation of TEGs under different conditions: power of the boiler, oxygen set-point, water temperature and quality of biomass. In particular, the results obtained with two fuels will be presented: pine pellets with a low tendency to ash deposition and a blended pellet from two residual agricultural biomasses with a high tendency to ash deposition (agropellet).

In the first part of this paper, the experimental facility is described. Then, the influence on TEGs performance of cold and hot side temperatures, as well as the fuel type is described. Afterwards, ash deposition over exchange surfaces when unconventional biomass is used is thoroughly analyzed and two measures to reduce its influence on the power produced by TEGs are presented.

# 2. Material and methods

# 2.1. Boiler and TEGs selection

The boiler used is the model BCH25 manufactured by BioCurve. It is a condensation unit fired by biomass pellet and has a thermal power of 25 kW [29]. It has been chosen because of its high efficiency, easiness of regulation and possibility of TEGs integration (as it will be seen later).

A detailed drawing of the boiler is depicted in Fig. 1. Pellets coming from the feeding bin (not shown) are fed into the combustion plate by a worm screw (1). Combustion air is blown by a fan (2) and introduced to the combustion chamber by several holes located both in the plate and around the flame. Ash from combustion goes to the bottom of the boiler because of the rotation at due time of the combustion plate (3) and the effect of a finger that drags ash out of the plate. Ash is removed from the bottom out of the boiler by a second worn screw (4). Flue gases from the flame go upwards till a piece called flame rebound (5), which direct them downwards and, then, go upwards again till the upper part. Finally, these gases go down through a set of helical ducts (6) surrounded by water and, finally, they leave the boiler at low temperature due to the effect of an induced draft fan. When flue gases flow inside the helical ducts, they transfer heat to the water and gases temperature goes down so that condensation can take place. It should be noted that water is heated not only by heat released by flue gases flowing through the helical ducts but also due to radiation and convection from the flame. Finally, it should be stressed that part of fuel ash is entrained by flue gases and then it can form deposits on heat transfer surfaces.

Two areas have been selected for locating TEG modules in the boiler (see Fig. 1): i) bottom part heated by the flame and cooled down by combustion air blown by the fan and ii) upper part heated by flue gases close to the flame rebound and cooled down by water. Since power produced by TEGs in the bottom part was low [30], only results related to the upper part will be presented here.

Three types of TEG modules have been selected for being located in the upper part. They are manufactured by TecTeg Thermoelectric Gencell Technology and their main characteristics appear in Table 1. The first two rows are the design temperatures for both hot and cold sides, and the other properties correspond to this nominal point (and,



Fig. 1. Scheme of the boiler used. (Source: BioCurve [29]). In the figure: 1. Feeding worm screw; 2. Inlet air fan; 3. Combustion plate; 4. Ash removing worn screw; 5. Flame rebound; 6. Gas helical ducts; 7. Upper cover.

# Table 1

Main characteristics of the TEG modules used [31].

Parameter	TEG1-PB-12611-6.0	TEG1-24111-6.0	TEG1-12611-6.0
Hot side temperature (°C)	350	300	300
Cold side temperature (°C)	30	30	30
Open circuit voltage (V)	9.2	17.7	8.4
Matched load resistance (ohm)	0.97	4.4	1.2
Matched load output voltage (V)	4.6	8.8	4.2
Matched load output current (A)	4.7	2	3.4
Matched load output power (W)	21.7	17.6	14.6
Heat flow across the module (W)	$\approx 310$	$\approx 301$	$\approx 365$

thus, they vary if these temperatures do). Open circuit voltage is the one provided when no electric load is connected to the TEG. Matched load resistance is that one that maximizes power produced: it entails values of voltage, current and power. Finally, it can be seen how heat flow is high compared to electricity produced and, thus, efficiency is around 4–7%. For this reason, integration of TEGs should be done in a fashion that low temperature heat leaving the modules is also used for water heating.

# 2.2. Boiler modification and installation

The aim of this section is to present how the boiler has been modified in order to integrate TEGs in the upper part of the boiler and to monitor different parameters needed to characterize the operation of the facility.

The upper cover of the boiler (see Fig. 1 (7)) made of refractory material was removed and replaced by a new element where TEGs, cooling system and thermocouples are located. Details of this element appear in Fig. 2 and are described next. First, a steel plate is located at the bottom; it will be heated by the flue gases and radiation from the flame and will serve as hot side plate of the TEGs. In order to direct all heat flow towards the TEGs, this plate is covered by a piece of insulation material in which two rectangular and one round perforations are made. TEG modules are located in contact with the steel plate in the rectangular perforations: three modules of the same type in each one, as it can be seen in Fig. 2 a. The figure shows also thermocouples placed for measuring temperatures in the hot side of TEGs and a round hole in the middle that will be used for introducing the deposition probe. TEG modules are cooled by two water coolers that are located just over them, as it can be seen in Fig. 2 b. Water for this cooling can come from two alternative ways: i) water entering the boiler to be heated in it (thus, the cooler acts as a preheater) or ii) tap water that is being heated while cooling the TEGs (this would correspond to a preheater if the boiler were used for sanitary hot water); the change from one mode to the other is made by manually operated valves. Temperature of cold side is measured by thermocouples located on the coolers. Finally, the system is covered by a steel plate, as it can be seen in Fig. 2 c.

During the first tests, power produced by TEG modules was low because temperature of the high temperature plate was significantly lower than the design temperature of the hot side of the modules. For this reason, changes were made in order to increase heat exchange between flue gases and high temperature plate [30]. The main modification was the use of fins, which can be seen in Fig. 2 d; in this photo, the plate is seen outside the boiler from the side which is in contact with flue gases. Additionally, the cover of the flame rebound (see Fig. 1 (5)), which is made of refractory material, was removed, what allowed higher flow of gases towards the finned plate and also to increase heat transfer from the flame by radiation. As it will be seen in the results section, these changes allowed the plate to achieve high temperature that enabled TEG modules to produce more power.

In the duct for flue gases evacuation, a gas analyser is located. Next to the boiler there is a bin that contains the pellets; this bin is placed on a scale in order to monitor the weight and, thus, the mass flow rate of pellets that are fed into the boiler. The facility also includes a water circuit with fan cooler and a pump to cool down water heated by the boiler in order to work in a closed circuit. The monitoring system registers temperatures (water, flue gases, entering air, hot and cold sides of the TEGs), mass of pellets in the bin, flue gases composition and current and voltage of each one of the TEG modules. Besides the computer, a box was located to contain electronic boards required for monitoring as well as electric resistances that serve as load for the TEGs. Each one of these resistances has been tuned so that the resistance plus the electric cable linking it with the module correspond to the matched load value.

# 2.3. Fuels

Tests have been carried out with two different fuels.



Fig. 2. Modifications of the upper part of the boiler for installing TEG modules.

# Table 2

Fuels properties.

Fuels properties				Ash properties			
		Pine	Agropellet			Pine	Agropellet
Proximate analysis (% m/m d.b)	Volatile matter <sup>a</sup>	84.2	72.4	Chemical ash composition (% m/m d.b.) <sup>h</sup>	Al <sub>2</sub> O <sub>3</sub>	2.1	2.7
	Fixed carbón <sup>g</sup>	15.4	21.7		CaO	24.0	45.8
	Ash <sup>b</sup>	0.4	5.9		Fe <sub>2</sub> O <sub>3</sub>	1.8	2.2
					K <sub>2</sub> O	13.0	14.9
Total Moisture (% m/m w.b.) <sup>c</sup>		7.2	9.1		MgO	9.9	8.6
Ultimate analysis (% m/m d.b.)	Carbon <sup>d</sup>	50.9	46.36		Na <sub>2</sub> O	1.8	0.4
	Hydrogen <sup>d</sup>	6.1	5.77		P2O5	8.5	4.5
	Nitrogen <sup>d</sup>	0.09	0.56		SO <sub>3</sub>	4.4	2.3
	Sulfur <sup>e</sup>	< 0.01	0.055		SiO <sub>2</sub>	15.0	17.7
	Chlorine <sup>e</sup>	0.01	0.047		TiO <sub>2</sub>	0.08	0.2
	Oxygen <sup>g</sup>	42.49	41.29		Cl	-	0.2
HHV (d.b. at p=constant) $(MJ \cdot kg^{-1})^{f}$		20.43	18.54	Ash melting temperatures oxidant conditions (°C) <sup>i</sup>	Initial deformation temperature (DT)	>1400	1130
LHV (w.b. at p=constant) (MJ·kg <sup>-1</sup> ) <sup>f</sup>		17.55	15.48		Flow temperature (FT)	>1400 >1400	1310 1370
<ul> <li><sup>a</sup> EN-ISO 18123:2016.</li> <li><sup>b</sup> EN-ISO 18122:2016.</li> <li><sup>c</sup> EN-ISO 18134:2016.</li> <li><sup>d</sup> EN-ISO 16948:2015.</li> <li><sup>e</sup> EN-ISO 16994:201.</li> <li><sup>f</sup> EN-ISO 14918:2011.</li> <li><sup>g</sup> Calculated.</li> <li><sup>h</sup> EN-ISO 16967:2015.</li> <li><sup>i</sup> CEN/TS 15370–1:2006.</li> </ul>							

- Pine pellet: Pellet A1 according to the UNE-EN ISO 17225-2 standard [32]. It will be used as a reference fuel to study the differences from fuels with high tendency to ash deposition.
- Agropellet: Agricultural residual pellets. In this case the pellet is a blend of 70% of vineyard pruning (woody) and 30% of barley straw (herbaceous).

The used fuels characteristics, as well as their ash properties are shown in Table 2.

# 2.4. Test features

In the installation that has just been described, a test campaign has been carried out with the objective of analyzing the influence of the temperature of the cold and hot side as well as the fuel used on the power generated by the TEGs.

Prior to each test, a general checking of the boiler and ancillary devices is made. If needed, TEGs are changed and their electric load resistances adjusted to the suitable value for maximum power. Afterwards, boiler is switched on and its parameters (e.g. oxygen set point or power level) are fixed according to the test to be performed. Once stable operation is achieved, the test itself is performed. Once it is finished, boiler is switched off. More specific details of each test are presented along section 3.

# 2.5. Methodology for the analysis of the influence of temperatures and heat transfer on power produced by TEG

A method based on linear regression and mobile mean will be applied in order to separate the influence that flue gas temperature, cooling water temperature and heat transfer in both hot and cold sides have on power produced by TEGs. Data used for the analysis are temperature of flue gases (Tg), average temperatures of hot and cold sides of TEGs (T<sub>h</sub> and T<sub>c</sub>), average temperature of cooling water (T<sub>w</sub>) and power produced by TEGs (W). Since this power is small compared to heat flows entering and leaving them (around 4-7%), it can be roughly considered that heat flow is constant and, thus, a thermal circuit can be defined with three thermal resistances; i) R<sub>h</sub>, from T<sub>g</sub> to T<sub>h</sub>, (taking into account hot side convection and radiation, resistance due to ash deposition and, finally, conduction in the hot plate), ii) Rk (thermal resistance of TEGs themselves as well as some effect of contact resistance) and iii) R<sub>c</sub>, which includes cold plate conduction as well as convection to cooling water. According to this thermal circuit, the following relations for the heat transferred (Q) can be written:

$$Q = \frac{T_g - T_h}{R_h} \approx \frac{T_h - T_c}{R_k} \approx \frac{T_c - T_w}{R_c}$$

The relative values of  $R_h$  and  $R_c$  compared to  $R_k$  can be calculated as function of measured temperatures:

$$rac{R_h}{R_k}pprox rac{T_g-T_h}{T_h-T_c}$$
 $rac{R_c}{R_k}pprox rac{T_c-T_w}{T_h-T_c}$ 

Since  $R_k$  is constant, the previous quotients between resistances are indicators of thermal resistances of hot and cold sides. By introducing these thermal resistances, W is a function of  $T_g$ ,  $T_w$ ,  $R_h/R_k$  and  $R_c/R_k$ . Since the variations of these variables are small, the dependence will be considered as linear:

$$W \approx a_{T_g} \cdot T_g + a_{T_w} \cdot T_w + a_{R_h} \cdot \left(\frac{R_h}{R_k}\right) + a_{R_c} \cdot \left(\frac{R_c}{R_k}\right) + b$$

Coefficients  $a_i$  and b of the previous relation can be obtained by applying linear regression from measured data. Finally, impacts that temperatures and resistances have on W are calculated as:

$$egin{aligned} & H_{T_g} = a_{T_g} \cdot \left(T_g - T_{g,ref}
ight) \ & H_{T_w} = a_{T_w} \cdot \left(T_w - T_{w,ref}
ight) \ & H_{R_h} = a_{R_h} \cdot \left[\left(rac{R_h}{R_k}
ight) - \left(rac{R_h}{R_k}
ight)_{ref}
ight] \ & H_{R_c} = a_{R_c} \cdot \left[\left(rac{R_c}{R_k}
ight) - \left(rac{R_c}{R_k}
ight)_{ref}
ight] \end{aligned}$$

where the subscript ref indicates reference values. In order to check the accuracy of the method, it is interesting to compute the error due to linearization:

$$Error = W - \left( a_{T_g} \cdot T_{g,ref} + a_{T_w} \cdot T_{w,ref} + a_{R_h} \cdot \left(\frac{R_h}{R_k}\right)_{ref} + a_{R_c} \cdot \left(\frac{R_c}{R_k}\right)_{ref} + b \right)$$

Finally, it is possible to filter the effects of  $T_g$ ,  $T_w$  and  $R_c$  and to calculate what would be the power produced if only the effect of  $R_h$  would be used. This value is very interesting because it quantifies the effect of deposition that vary during the test.

$$W_{only\ Rh} = W - I_{T_g} - I_{T_w} - I_{R_c}$$

The method presented does not provide suitable results when applied directly to the measured variables because of the inertia of the system (especially the thick steel layer and the fins located in the hot side of TEGs). This makes that when temperature of gases increases, temperature of hot side of TEGs needs several minutes to increase. To solve this problem, mobile means of the measured variables are used. In particular,  $T_h$ ,  $T_c$ ,  $T_w$  and W are averaged between -15 and 15 min whereas  $T_g$  is averaged between -20 and 10 min (what allows better fitting due to the delay between  $T_g$  and  $T_h$ ). Afterwards, the calculation of  $R_h/R_k$  and  $R_c/R_k$  and the adjustment of the linear regression is made based on these moving means instead of the instant measured variables.

# 2.6. Ash analysis

In addition to what has been described in section 2.2, the facility includes a deposition probe, with a removable sampling ring in the upper part of the combustion chamber. This is a common device used to quantify fly ash deposition in furnace pipes and heat exchangers [22, 33]. Prior to the experiment, the removable sampling ring is cleaned, dried, measured and weighed. During a stable combustion period, the deposition probe is inserted inside boiler and the ring is cooled by compressed air, keeping its surface at an appropriate temperature for studying deposition [34]. For the tests presented here, compressed inlet air was adjusted to keep an average temperature of  $325 \pm 25$  °C. Once extracted, the dirty ring is dried and weighed again to determine the mass of deposits, allowing deposition rate (DR, g·m<sup>-2</sup>·h<sup>-1</sup>) to be calculated.

Once deposits had been weighed and DR calculated, a sample was taken from the removable sampling ring. A sample of bottom ash were also collected and crushed in a mortar to obtain a homogeneous mixture with an adequate particle size to be analyzed. Each one of these samples (deposits and bottom ash) were glued onto metal plates with carbon tape and then coated with carbon before being analyzed by SEM-EDS method. The equipment used was a Carl Zeiss Merlin electronic field emission microscope equipped with Gemini Column, with acceleration voltages between 0.02 and 30 kV, fitted with an EDS X-MAS detector by Oxford Instruments with a window of 20 mm<sup>2</sup> and energy resolution between 127 eV and 5.9 keV. For each sample, three 1 mm<sup>2</sup>-zones were selected, and images taken with the retro-dispersed detector (asb). Average elemental composition was obtained through EDS, using a voltage of 15 kV. INCA software was used to process the results. Major participating elements in the most important ash transformation processes —namely Na, Mg, Al, Si, P, S, Cl, K, Ca and Fe— were included in the analysis.

# 3. Tests results

# 3.1. Influence of cold side temperature

A test has been performed in the facility previously presented in order to compare the performance of TEG modules depending on whether they are cooled by boiler water or by tap water. Three modules of TEG1-PB-12611-6.0 type and three modules of TEG1-24111-6.0 type are used (see Table 1). Boiler is fed by pine pellet and it is set to the maximum power (level 5). The test starts with boiler start-up with TEGs cooled by boiler water; actually, water coming from the fan cooler flows through TEGs cooler before entering the boiler to be heated again. Once a quite stable value is achieved, TEGs cooling system is changed to tap water. Finally, boiler is shut down and, during this process TEGs are cooled again by water of the boiler cooling circuit.

First of all, power produced by the different modules is presented.

Afterwards, values of temperatures at both hot and cold side are presented in order to show how they are the main cause of variation of TEGs power.

TEG 1 to 3 are TEG1-PB-12611-6.0 type and the power they produced is plotted in Fig. 3 (left). It can be seen how power of TEG 1 and 3 is quite similar, whereas that of TEG 2 is lower. This lower value may be due because of poor thermal contact of the module, perhaps due to its location in the middle of the row, although more research is needed to confirm this idea. In all cases, it can be seen how power is zero at the beginning of the test when boiler is switched on and increases till reaching an almost steady state value because of the heating of the hot plate due to radiation and convection from flue gases. As said before, cooling during this first stage is done by water coming from the fan cooler before it enters the boiler. After about 30 min, cooling mode is changed and now is done by using tap water. During the change of cooling mode, there is a very short period of time when cooling disappear, what makes a strong reduction of power. Afterwards, power increases suddenly and then it decreases slightly to reach a stable value higher than the previous one. Finally, boiler is switched off after about one hour and 50 min, and power decreases as temperature of hot side also does. It should be noted that, in order to save tap water, cooling mode is changed again to boiler water just after the boiler is switched off, what causes worse cooling and a decrease in power produced by TEGs. It is far from its nominal value of 21.7 W, what is mainly due to the fact that temperatures of both hot and cold side are not the same as the design ones.



Fig. 3. Power (W) produced by TEG 1 to 3 (TEG1-PB-12611-6.0) (left) and TEG 4 to 6 (TEG1-24111-6.0) (right).

# Table 3

Summary of power produced during the test.

		TOTAL	TEG1	TEG2	TEG3	TEG4	TEG5	TEG6
Boiler water cooling	Max. power (W)	59.6	7.71	6.05	8.08	13.08	12.34	13.24
	Mean power (W) <sup>a</sup>	56.91	7.50	5.01	7.48	12.59	11.84	12.46
Tap water cooling	Max. power (W)	69.54	9.56	6.27	9.61	15.41	14.18	15.52
	Mean power (W) <sup>b</sup>	68.10	9.15	5.85	8.96	15.11	13.92	15.11

<sup>a</sup> During 10 min before change in cooling mode.

<sup>b</sup> From change in cooling mode until boiler stop.



Fig. 4. Temperature (°C) of hot side of TEG 1 to 3 (left) and TEG 4 to 6 (right).



Fig. 5. Mean temperature (°C) of cold side of TEG.

Fig. 3 (right) shows the evolution of power produced by TEG 4 to 6, which are of TEG1-24111-6.0 type. It can be seen how the power is higher than in the previous case, and no so far from to its nominal value (17.6 W). Besides, there are no big differences among the three modules, although the one located in the middle (number 5 in this case) also has the lower values. The general trends in the evolution are the same as those commented for TEG 1 to 3 but, additionally, other effects that were negligible in that figure can now be appreciated more clearly. First, about 20 min from the test start pump driving water in the circuit connecting boiler, fan cooler and, in this case, also TEG cooler, turns on, what causes an improvement in cooling and, thus, a sudden increment in power produced. Second, at the end of the cooling process, the same pump turns on and off alternatively (depending on the value of water temperature), what causes changes in cooling and in power produced.

A summary of power produced in both situations is shown in Table 3. It can be seen how by using tap water for cooling maximum power produced is 10 W higher. Since operation is quite stable, average values are close to maximum power values.

In order to understand the variation in power produced presented, it is worth to analyze the evolution of temperatures at both sides of TEG modules. Fig. 4 (left) shows the evolution of temperatures in hot side of TEG 1 to 3. It should be noted that this temperature is measured very close to the TEGs but in the hot plate, what means that the actual temperatures of the TEGs hot faces will be lower due to contact thermal resistance. It can be seen how, even in the best situations, these measured temperatures ranges from around 240 to 310  $^\circ$ C; since the actual temperature will be lower, hot side of TEGs will be well below the value of 350 °C of design conditions, what is a main cause for producing power lower than the value of design. The graph shows how temperature increases during boiler heating and then tends to be stable. When cooling mode is changed, it also affects slightly to temperature of hot side, but due to the inertia of the system (the hot plate is thick) it takes several minutes to have the stable value corresponding to tap water cooling. Finally, when boiler is switched off, temperature decreases slowly. At the end of the cooling process, there is a period when temperature almost does not decrease; this effect appears because during this period the pump is switched off.

Evolution of temperature at hot side of TEG 4 to 6 is plotted in Fig. 4 (right). Again, this value is measured in the hot plate and the actual values of TEGs face would be lower. It can be seen a similar trend as that of hot side temperature of the other TEGs, but now values are higher. This effect would be mainly due to an asymmetry of the boiler, what would direct hotter gases and/or radiation from the flame to the area where TEG 4 to 6 are located. These higher temperatures combined with the fact that design hot side temperature of these TEGs is lower, would justify that the power provided by TEG 4 to 6 is much closer to the design value than that of TEG 1 to 3.

Finally, cold side temperature appears in Fig. 5. Two measures are plotted, one for the cooler of TEG 1 to 3 and the other for the cooler of TEG 4 to 6. Since thermocouples are located close to TEGs cold side but in the surface of the cooler, the actual temperature of the TEGs cold side would be higher. As it can be seen, these temperatures have relevant variations due to changes in the cooling mode and to switching on and off of the pump. At the beginning, temperatures increase fast during 20 min, when they drop suddenly because pump is switched on. Afterwards, they increase again till reaching almost stable values and, then, they decrease fast due to the change in cooling mode. Since the decrease of cold side temperatures is faster than the decrease of hot side temperatures (the latter have higher thermal inertia than the former), peaks in power produced appear (see Fig. 3). Just when boiler is switched off, cooling mode is changed again, what causes an increment of temperatures. Finally, temperatures decrease gradually as boiler cools down, except during a short period at the end when they increase because pump is stopped. The graph shows that temperatures are around 25 °C lower when tap water is used, which is the cause of the higher power produced.

The test results show how the change of cooling system affects power produced by TEG modules (a variation of more than 10 W). This result supports the importance of using the coldest flow available in order to maximize produced power.

# 3.2. Influence of hot side temperature

In order to analyze the influence of the hot side temperature in the power generated by the TEG modules, several tests were carried out with both fuels (pine pellet and agropellet). In this test, as well as in all test presented in the following sections, three TEG1-PB12611-6.0 modules and three TEG1-12601-6.0 modules are used (see Table 1), cooling is made by boiler water and it is set to the medium power (level 3).

To vary the gas temperature, and with it the temperature of the hot side of the TEG modules, two O<sub>2</sub> set-points were selected in the chimney: 7% and 10%. A total of 5 tests were carried out (T1 to T5), in three of which we worked with two different conditions, varying the set-point during the test, thus making the result obtained independent of other factors, mainly ambient temperature. For each condition, an average duration of 1 h, in a stable situation, was considered. The mean results obtained are shown in Table 4. It can be verified that the temperature of the hot side of the TEGs is lower when the percentage of oxygen is higher, as expected, which implies a decrease in the power generated as reflected in tests 3, 4 and 5.

Summary o	ummary of power produced during the test.								
Pine Pellet			Agropellet						
	O <sub>2</sub> (%)	Mean Temp Hot Side (°C)	Mean Power (W)		O <sub>2</sub> (%)	Mean Temp Hot Side (°C)	Mean Power (W)		
T1	6.3	342.0	44.0	T4	7.0	307.3	40.1		
T2	9.6	324.4	39.8		10.2	295.0	36.4		
T3	6.8	330.0	41.4	T5	7.4	265.2	29.9		
	9.3	320.6	40.3		10.1	260.9	28.9		



Fig. 6. Mean Temperature (°C) of hot side of TEG in long-term test with pine pellet and agropellet.



Fig. 7. Total power in long-term test with pine pellet and agropellet.

Despite the fact that the temperatures reached with the agropellet are lower than those of the pine pellet, mainly due to the different settings of the boiler to adapt to the characteristics of each of the fuels (bulk density, PCI and ash percentage), it can be verified how there is a statistically relevant correlation between the mean temperature of the hot side and the total power of the TEGs of the 8 periods analyzed ( $R^2 = 0.9634$ ).

Therefore, taking into account the location of the TEG modules, working with less excess of air means more power generated for these modules for both fuels, due to the increase in the temperature of the hot side.

# 3.3. Influence of fuel type (ash deposition tendency)

Both fuels used in the test present a different tendency to deposition as it has been demonstrated in previous tests carried out in a lab-scale reactor, being significantly worse the behaviour of the agropellet (see Ref. [26]). To analyze the influence on the TEG modules performance of the two fuel types, a long test (about 8 h in stable condition) with each of them were carried out. In both cases, we started from a situation in which the lower plate of the location of the TEG modules in which the fins are located (see Fig. 2 d) was cleaned with compressed air. The oxygen set-point in both cases was 7%.

Fig. 6 shows the evolution of the mean temperature on the hot side of the TEGs. It can be seen how this temperature was again lower in the test carried out with the agropellets, as had already happened in the short tests. In addition, the figure shows how the mean temperature decreases progressively in the agropellet test, while it is more stable in the test with pine pellet. Taking into account the greater tendency to ash deposition of the first, the decrease could be directly related to the formation of a thermally insulating layer of deposits on the gas side, which decreases heat transfer, and therefore the temperature reached on the hot side of the TEGs. Related to it, it can also be verified as the power produced by the TEG modules decreased progressively in both tests (see Fig. 7), diminishing more pronouncedly in the one carried out with agropellet (approximately 0.78 W per hour compared to 0.24 W per hour with pine pellets).

To quantify ash deposition, the DR was also measured with each one of the fuels during these tests. In the test with pine pellets, the probe was inserted for 5 h in order to collect a sufficient sample (in previous preliminary tests it was verified that with a short time of insertion of the probe the amount of deposits collected was insufficient to calculate the DR). In the case of the agropellet test, the probe was introduced five times with different durations. Table 5 shows the results obtained for deposited mass per unit area and DR. For the agropellet, the deposited mass increases proportionally with time as it has also been detected in other experiences [24], which could be indicative that the thermally insulating layer of ash is increasing, making its effect progressively greater. In addition, the amount deposited is much greater than in the case of pine pellets (the amount deposited per unit area for 2 h is similar to that deposited during 5 h when the fuel used is pine pellet), which accelerate the negative effect of ash deposition, as shown in Figs. 6 and 7. DR with agropellets was on average 2.4 times higher than with pine pellets, which would confirm again its greater tendency to deposition. From the five values obtained with this fuel, it was possible to verify that the DR did not vary throughout the test, nor was it influenced by the time that the probe was in the boiler, which may indicate that there are no symptoms of saturation as if they have been detected in other experiences with similar fuels [35].

It was verified that, in addition to the power of the TEG modules, throughout the long-term tests the temperatures of the water and gases also varied. Both values can influence temperatures of cold and hot side of TEG, as it has been seen in previous sections and, therefore, they can cause variations in the power produced by the TEGs. Thus, it is not immediate to determine which part of the power loss detected is due to ash deposition and which to other factors. For all these reasons, it was decided to analyze the effect of ash deposition in greater depth. This analysis is shown in the next section.

# 4. Ash deposition analysis

# 4.1. Quantification of ash deposition effect

The methodology presented in section 2.5 has been applied to analyze how temperatures of gases and water and thermal resistances in the hot and cold side affect the evolution of power produced in both

DR in long-term test.

Pine Pellet	Agropellet					
5	0.5	1	1.5	2	2.5	Mean
1.11	2.43	2.63	2.45	2.84	2.76	2.62
5.55	1.25	2.67	3.66	5.67	6.88	-
	Pine Pellet 5 1.11 5.55	Pine Pellet         Agropellet           5         0.5           1.11         2.43           5.55         1.25	Pine Pellet         Agropellet           5         0.5         1           1.11         2.43         2.63           5.55         1.25         2.67	Pine Pellet         Agropellet           5         0.5         1         1.5           1.11         2.43         2.63         2.45           5.55         1.25         2.67         3.66	Pine Pellet         Agropellet           5         0.5         1         1.5         2           1.11         2.43         2.63         2.45         2.84           5.55         1.25         2.67         3.66         5.67	Pine Pellet         Agropellet           5         0.5         1         1.5         2         2.5           1.11         2.43         2.63         2.45         2.84         2.76           5.55         1.25         2.67         3.66         5.67         6.88



Fig. 8. Impact of  $T_g,\,T_w,\,R_h$  and  $R_c$  in the power produced by TEGs during the agropellet test.

agropellet and pine pellet tests. The analysis has considered only the stable operation period avoiding boiler heating and boiler shut-off.

First, the case of agropellet is analyzed. Regression provides the following equation:

$$W \approx 0.11793 \cdot T_g - 0.32344 \cdot T_w - 28.952 \cdot \left(\frac{R_h}{R_k}\right) + 271.18 \cdot \left(\frac{R_c}{R_k}\right) + 5.7710 \left(R^2 = 0.9980\right)$$

Fig. 8 shows the impact on power produced by TEGs that is due to each one of the different causes as well as the error. The reference values have been chosen as round values close to the average of them during the analyzed period:  $T_{g,ref} = 570 \degree C$ ,  $T_{w,ref} = 36.5 \degree C$ ,  $(R_h/R_k)_{ref} = 1$ ,  $(R_c/R_k)_{ref} = 1$ ,  $R_k$ )<sub>ref</sub> = 0.2. As explained in section 2.5, impact is the variation of power that appears because the value of each one of the causes (temperature of gases, temperature of water, resistance of hot side and resistance of cold side) is different from the reference value. For instance, according to the previous regression equation, an increment of one degree of flue gases temperature causes an impact of 0.11793 W in the power produced by TEG, whereas an increment of one degree of water temperature causes an impact of -0.32344 W. Fig. 8 shows how at the beginning impact due to T<sub>g</sub> is negative because this temperature is lower than the reference value, then impact is small because temperature is stable and close to the reference value and finally impact is negative because Tg decreases again. Impact of temperature of water is positive at the beginning and decreases becoming negative at the end (because this temperature increases during the test and the coefficient is negative). Due to deposition, thermal resistance of the hot side (R<sub>h</sub>) increases along the test and causes power to decrease from the beginning to the end of the test (a total reduction of around 5 W). It should be noted that Rh does not only



Fig. 9. Evolution of power during the agropellet test.

take into account resistance due to deposition but also radiation and convection heat transfer from flue gases and conduction through the hot plate ( $T_h$  is measured in the side of this plate which is in contact with TEG modules); however, resistance due to deposition is the only one of them that has a relevant long time variation along the test (effect of flue gases temperature on radiation and convection is of short term), so that it can be said that long term impact due to  $R_h$  variation is almost completely due to the effect of ash deposition. Finally, effect of thermal resistance in the cold side is negligible and error of the method is smaller than the main impacts.

Evolution of three different values of power produced by TEGs during the stable part of the agropellet test is plotted in Fig. 9. First, the measured value of power (W\_measured) shows oscillation due to the short term variation of conditions (mainly  $T_g$ ). This oscillation is minimized by filtering the measurement by moving average (W\_filtered). Finally, a corrected value of W removing all effects different from deposition is also plotted (W\_onlyR\_h); this corrected value corresponds to what would be the evolution of W\_filtered if only deposition would take place (keeping  $T_g$ ,  $T_w$  and  $R_c$  constants). It can be seen how during the first half of the test W\_filtered is close to W\_onlyR\_h, what means that almost all power reduction is due to deposition. However, in the second half W\_filtered and W\_onlyR\_h diverge and at the end the former is around 2 W lower than the latter. This means that, from all power reduction, 2 W are due to other reasons (Fig. 8 shows that it is due to temperature of gases and of water).

The same methodology has been applied to the pine pellet test in order to quantify how temperatures of gases and water and thermal resistances in hot and cold sides affect power produced. In this case, regression equation is:

$$W \approx 0.11558 \cdot T_g - 0.49820 \cdot T_w - 30.891 \cdot \left(\frac{R_h}{R_k}\right) + 41.069 \cdot \left(\frac{R_c}{R_k}\right) + 18.523 \ (R^2 = 0.9892)$$

Again, reference values of the variables have been chosen close to their average values during the test, which are not always the same as in the agropellet test:  $T_{g,ref} = 650$  °C,  $T_{w,ref} = 35$  °C,  $(R_h/R_k)_{ref} = 0.98$ ,  $(R_c/R_k)_{ref} = 0.2$ . Impacts of the four variables are plotted in Fig. 10. It can be seen how temperatures of gases and water have the main impacts on power produced. At the beginning of the test, temperature of gases is close to reference value and, thus, impact is small. Afterwards, this temperature increases (what causes a positive impact on power, because the correlation coefficient is positive) and then decreases again close to the reference value (impact between hours 3 and 5 is small). Finally, flue gases temperature of water is lower than the reference value at the beginning and higher at the end; since its coefficient is negative



Fig. 10. Impact of  $T_g$ ,  $T_w$ ,  $R_h$  and  $R_c$  in the power produced by TEGs during the pine pellet test.



Fig. 11. Evolution of power during the pine pellet test.

(-0.49820 W/°C), this cause a positive impact on power at the beginning and negative impact at the end. Effect of deposition (R<sub>h</sub>) is smaller and is positive at the beginning (low deposition and higher power) and negative at the end (the opposite). This effect is much lower than in the agropellet test (which is in accordance with its lower tendency to deposition). Finally, effect of heat transfer at cold side (R<sub>h</sub>) is negligible and error is smaller than the main impacts.

Finally, Fig. 11 shows the evolution of three values of power during the pine pellet test: measured power (W measured), the same power but filtered by using moving mean (W filtered) and what would be the W filtered if only effect of deposition were present (W onlyR<sub>h</sub>). It can be seen how power (W filtered) is high at the beginning, then decreases and finally increases again but up to a lower value. However, the smaller variation of W\_onlyRh shows that deposition is the cause of a small part of the big variation of W\_filtered and that the main part is due to other causes (Fig. 10 shows how these causes are temperature of gases and temperature of water). Besides, W\_onlyRh shows that power reduction due to deposition along the test is lower than this power reduction in the agropellet test.

The methodology developed has been able to disaggregate the influence that temperatures of gases and water and thermal resistances at hot and cold side have on power produced or, in other words, to quantify the impact of each one of these causes. In particular, the analysis has allowed to quantify more precisely the impact that ash deposition has on power produced and to see clearly how this effect is much higher when agropellet is used.

# Table 6

Results of the tests carried out to analyze the deposition phenomer	non
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In order to better understand the phenomenon of deposition with the agropellet, and thus be able to improve the operating conditions that help to minimize it, the deposits collected in each of the five sampling rings of the test shown in section 3.3 are analyzed.

Deposits can be separated into two fractions, depending on the mechanisms by which the ash leaves the bed:

- The first fraction corresponds to the deposits by condensation, associated with the ash that leaves the bed by vaporization and which includes the deposition by thermophoresis and diffusion. Vaporization is closely related to the temperature in the bed (influenced by the combustion temperature) [35].
- The second fraction includes the deposits by inertial impact which are associated with the ash that leaves the bed by entrainment, which depends on the velocity of the air in the bed and the degree of sintering (particle fineness) of the bottom ash [35]. In this sense, it should be noted that sintering phenomena were not detected in bottom ash in any of the tests.

In order to quantify these two deposition fractions, the methodology developed in Ref. [35], will be applied. This methodology is based on the elemental composition of the ash (deposits and non-sintered bottom ash fraction) and basically assumes that 1) the composition of the deposits by inertial impact is similar to the non-sintered bottom ash fraction and 2) that Si can only leave the bed by entrainment. Ash samples of each one of the five sampling rings were obtained from the removable sampling rings and were analyzed by SEM-EDS method (section 2.6). A single bottom ash sample was also analyzed, which was used for the 5 sampling rings due to the difficulty of obtaining bottom ash samples that correspond to a certain moment of the test. From the results of these analyses and applying the abovementioned methodology, the DR percentages that are due to condensation (DR<sub>Cond</sub>) and inertial impact deposition (DR<sub>Imp</sub>) mechanisms were obtained for each of the five periods in which the deposition probe was inserted in the boiler. Table 6 shows the operating conditions of these five periods and the results obtained.

The first thing that can be observed is that, with the time ranges used and as it happened with DR, it is also not possible to find relationships between the time of introduction of the deposition probe with the composition of the deposits, the  $\text{DR}_{\text{Imp}}$  or the  $\text{DR}_{\text{Cond}}$ , so this variable is not will take into account.

It can be verified that, in average, more than 86% of the deposits

		Period 1	Period 2	Period 3	Period 4	Period 5	Mean values	
T gas	(°C)	523	518	535	518	517	522	
02	(%)	7.11	7.22	7.29	7.30	7.58	7.30	
Air Exc.	(%)	50.14	51.43	52.17	52.29	55.39	52.28	
SEM-EDS ana	lisys results							
		Deposits						Bottom Ash
Na	(%)	0.47	0.48	0.52	0.55	0.58	0.52	0.26
Mg	(%)	0.15	0.25	0.32	0.32	0.32	0.27	3.92
Al	(%)	0.27	0.29	0.25	0.26	0.25	0.26	1.40
Si	(%)	0.59	1.20	1.60	1.54	1.55	1.30	18.61
Р	(%)	0.08	0.18	0.15	0.22	0.20	0.17	2.26
S	(%)	8.33	6.41	12.08	9.72	9.83	9.27	1.13
Cl	(%)	35.25	36.06	27.02	31.25	30.57	32.03	0.44
К	(%)	53.07	52.46	54.71	52.81	53.47	53.30	26.39
Ca	(%)	1.66	2.48	3.14	3.19	3.04	2.70	43.71
Fe	(%)	0.11	0.19	0.22	0.14	0.21	0.17	1.88
Deposition R	ates							
DR	(g/m2·h)	2.45	2.43	2.76	2.63	2.84	2.62	
DRImp	(%)	6.6	11.0	13.1	13.4	13.0	11.42	
DR <sub>Cond</sub>	(%)	93.4	89.0	86.9	86.6	87.0	88.58	



Fig. 12. Influence of cleaning system in the total power produced by TEG modules.

came from vaporization in the bed (and subsequent condensation on the exchange surfaces; see  $DR_{Cond}$  values in Table 6) and therefore less than 14% came from entrainment (and subsequent deposit by inertial impact; see  $DR_{Imp}$  values in Table 6).

Therefore, given the predominance of deposition by condensation, it is foreseeable that the most critical parameter in deposition is the bed temperature, with air velocity being less important.

For this, it can be concluded that in order to limit the deposition phenomena when using unconventional fuels in terms of their ashes and therefore reduce the negative influence on heat transfer in the exchange surfaces, including those that provide heat to TEGs, it is interesting to work with high excesses of air, which reduce the temperature of the combustion chamber. Nevertheless, this has two negative effects.

- First, it affects the boiler's performance. In any case, it is not especially significant for condensing boilers, such as the one used in this project; thus, when using the agropellet, going from an excess of air of 50–75% means reducing the boiler's efficiency over LHV by only 1 point (from 101.4 to 100.4%).
- Second, it means a decrease in the gases temperature, which negatively affects the power of the TEG modules (see section 3.2). Since the temperature of these gases is much higher than the optimum temperature of their hot side, this decrease in power could be minimized by further improving heat transfer between the gases and the TEGs.

Anyway, for each case it will be necessary to find an excess of air that represents a compromise between an acceptable deposition and a reduced decrease in the boiler's and TEGs performances.

# 4.3. Strategies to reduce the effect of ash deposition on TEGs performance

Through medium/long term tests, it has been verified that using a biomass with a high tendency to ash deposition means a decrease in the power produced by the TEGs over time, due to ash deposition on the hot exchange surface. To mitigate this effect both primary (reduce the severity of the phenomenon) and secondary (removing deposits) strategies should be analyzed.

Fistly, it is possible to act by modifying the operating parameters and working with high excess of air that reduces deposition by condensation (which is, in this case, the most important fraction). However, this solution, as already explained in the previous section, is limited, so it is necessary to find additional options. One of them could be to integrate a cleaning system in the boiler. These types of systems are commonly used to remove the thermally insulating layer of deposits and with an optimized blowing sequence, they turn out to be a very suitable solution [27, 36].

To analyze the influence that cleaning the hot side can have over TEG modules operation, a simple system with compressed air was designed and fabricated. To verify its usefulness, a new test using agropellet was carried out ( $O_2$  set-point was 7% and cooling with boiler water). As shown in Fig. 12, once the different parameters were stabilized, the power produced by the TEGs progressively decreased (period A), most probably due to ash deposition on the hot side associated with the use of agropellets as fuel, which implies a loss of heat transfer.

After this initial period, a hot cleaning of the exchange surface was carried out using the designed system. An average increase in power of 9% was obtained after it (see Fig. 12, period B; corresponding to the new period with the boiler parameters stabilized again).

Results obtained in this test allow to conclude that a blowing system correctly integrated into the boiler and with an optimal cleaning sequence would allow the achievement of adequate operating conditions for the TEGs in the boiler, and thereby increase the power generated, using unconventional biomasses as fuel.

# 5. Conclusions

A lab facility has been developed in order to demonstrate and test the integration of thermoelectric modules in a biomass boiler. The upper part of a 25 kW<sub>th</sub> pellet-fired condensation boiler was modified in order to integrate six TEGs of different types.

The influence of the cold side temperature has been verified, testing two different TEGs integration configurations. Likewise, the influence of the temperature of the hot side has been analyzed. In this sense, it has been verified that when working with a biomass with a high tendency to ash deposition, there is a progressive decrease in the heat transferred and, therefore, in the power produced by the TEG modules. To minimize this fact, it is necessary to adopt an adequate operating strategy related with excess of air which, together with an adequate integration of the TEG modules, means reaching a compromise between a reduction in ash deposition without significantly affecting the performance of the boiler or the TEG modules themselves. In any case, it seems that in order to work with these types of biomass it is essential to have an adequate cleaning system for the exchange surfaces.

Due to only 6 TEGs were integrated, and therefore only a small fraction of heat provided by the boiler (less than 10%) was directed towards TEG modules, the amount of power in this experience was small (no more than 70 W). Accordingly, there is a strong potential for increasing this power by introducing bigger changes in the design of the boiler that would allow to integrate more TEG modules. This could be useful for driving ancillary devices (fans, pump and worm screw), what eventually would lead to a boiler that could be used in an isolated facility with no electric grid.

According to results obtained in this research and with the aim of

continuing ahead in the integration of TEGs in biomass boilers that use non conventional biomass, it would be interesting to continue the research in several directions such as: i) additional locations for including more TEGs in the boiler to produce more power, ii) ash deposition with other types of agricultural biomass or iii) optimization of cleaning cycles.

# **CRediT** authorship contribution statement

Sergio Usón: Conceptualization, Formal analysis, Data curation, Investigation, Methodology, Project administration, Writing - original draft, Writing - review & editing. Javier Royo: Conceptualization, Formal analysis, Data curation, Investigation, Methodology, Writing original draft, Writing - review & editing. Paula Canalís: Conceptualization, Formal analysis, Data curation, Investigation, Methodology, Writing - original draft, Writing - review & editing.

# Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

# Data availability

Data will be made available on request.

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