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Pyroelectric energy converter using co-polymer P(VDF-TrFE) and Olsen cycle for waste heat energy harvesting

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ABSTRACT

This study was concerned with designing, building, and testing a pyroelectric energy converter to directly convert waste heat into electricity. The pyroelectric effect refers to the flow of charges to or from the surface of a material upon heating or cooling. The device consisted of a teflon cylindrical chamber with a piston oscillating vertically and driving a working fluid back and forth between a heat source and a cold heat exchanger. The Olsen cycle was performed on co-polymer 60/40 poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] thin films sandwiched between metallic electrodes. Their temperature oscillation, charge, voltage and the overall heat input and output were measured experimentally. Then, the electrical power generated and the energy efficiency were computed. The effects of channel width, frequency, and stroke length on temperature swing, heat input, as well as energy and power densities were investigated. Reducing the channel width and increasing the stroke length had the largest effect on device performance. A maximum energy density of 130 J/l was achieved at 0.061 Hz frequency with temperature oscillating between 69.3 and 87.6 °C. Furthermore, a maximum power density of 10.7 W/l was obtained at 0.12 Hz between 70.5 and 85.3 °C. In both cases, the low and high electric fields in the Olsen cycle were 202 and 739 kV/cm. To the best of our knowledge, this is the largest energy density achieved by any pyroelectric energy converter using P(VDF-TrFE). It also matches performances reported in the literature for more expensive lead zirconate stannate titanate (PZST) ceramic films operated at higher temperatures.

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1. Introduction

In the last decades, direct energy conversion devices for medium and low grades waste heat have received significant attention due to the necessity to develop more energy efficient engineering systems. In fact, in 2002 more than 50% of the net primary energy resource consuming in the U.S. was lost mainly in the form of waste heat [1]. To date, the effort has focused on thermoelectric generators based on the Seebeck effect that results in the creation of an electromagnetic force (emf) in the presence of a steady-state temperature difference between two junctions of dissimilar metals or semiconductors. It has been reported that commercial thermoelectric generators have efficiencies that are "*typically around 5%*" [2]. Higher efficiencies have been achieved with expensive nanostructured thermoelectric materials after 50 years of research efforts [3]. Stirling engines can also operate at low temperatures to convert waste heat into mechanical energy but

their performance is hindered by heat losses and an additional step is required to convert mechanical energy into electricity by means of an electrical generator or dynamo. Alternatively, organic Rankine cycles use organic working fluids such as refrigerants and hydrocarbons instead of water to harvest waste heat at temperatures up to 200-300 °C [4].

Pyroelectric energy conversion offers a novel and direct way to convert waste heat into electricity by alternatively heating and cooling a pyroelectric material resulting in electricity generation [5–10]. It makes use of the pyroelectric effect to create a flow of charge to or from the surface of a pyroelectric material as a result of heating or cooling [11]. The fact that very small changes in temperature (~mK) can produce a pyroelectric current on the order of nA or pA [11] has been used extensively in infrared detection for imaging and motion sensing as well as for thermometry applications [12]. The use of pyroelectric materials to harvest waste heat has been explored both experimentally and theoretically. While early theoretical studies predicted low efficiencies [13,14], experimental studies [5,7,8,10,15–18] have achieved significantly better results. Overall, however, little efforts have been dedicated to developing pyroelectric energy conversion.

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Nomenc	lature	t T	time, s				
		$\frac{I_i}{\pi}$	temperature at location 1, °C or K				
A	surface area of the PE, m ²	l _i T T	mean temperature at location i, °C or K				
D	nim thickness, m	I _{c, i} , I _{c, o}	cold heat exchanger inlet and outlet temperatures, °C				
\mathcal{C}_1	capacitance, F	T _{Curie} , ↓	Curie temperature from para- to ferroelectric phase				
c_p	specific heat, J/kg.K	т	change, °C				
D _{PE}	electric displacement of the PE, C/m^2	I _{Curie} , ↑	curie temperature from ferro- to paraelectric phase				
D_h	nydraulic diameter, m		change, °C				
$E_{\rm PE}$	electric field across the PE, V/cm	ΔT_i	temperature swing at location 1, °C				
E_L, E_H	low and high electric fields, V/m	<i>u</i> ₀	amplitude of velocity oscillation, <i>m/s</i>				
F_p	power factor, Eq. (9)	V_1, V_2	measured voltage across C_1 and R_2 , V				
f	frequency, 1/s	V_L, V_H	low and high voltage, V				
h_ω	convective heat transfer coefficient for oscillating flow,	V_m	measured voltage across resistor R_D , V				
	$W/m^2 \cdot K$	$V_{\rm PE}$	voltage across the PEs, V				
I_L	leakage current, A	V _{rms}	Kapton electrical heater voltage, V				
I_p	pyroelectric current, A	A	volume, <i>m</i> ³				
I _{rms}	Kapton electrical heater current, A	W_E	electrical work output, J				
k_f	fluid thermal conductivity, $W/m \cdot K$	W _{cycle}	net electrical power generated over a cycle, W				
L	channel length, <i>m</i>	W_E	generated electrical power, W				
l_0	amplitude of fluid flow displacement, m	W_p	piston pumping power, W				
т _с	cold heat exchanger mass flow rate, <i>kg/s</i>	Ζ	vertical coordinate in (x,z) coordinate system				
$n_{\rm PE}$	number of pyroelectric elements						
N_D	energy density, J/l	Greek symbols					
Nu_{ω}	Nusselt number for oscillating flow	α_f	fluid thermal diffusivity, m^2/s				
p_c	pyroelectric coefficient, <i>C</i> / <i>m</i> ² · <i>K</i>	η	thermodynamic efficiency, %				
P_D	power density, <i>W</i> / <i>l</i>	η_{Carnot}	Carnot efficiency, %				
Pr_f	Prandtl number, Eq. (16)	ω	piston angular velocity, <i>rad/s</i>				
P_S	spontaneous polarization, C/m^2	ν_f	fluid kinematic viscosity, <i>cst</i>				
$q_{ m PE}$	charges on the PE electrodes, C	ρ	density, <i>kg/m</i> ³				
Q _{in}	heat transfer rate into the device, W	τ	period of oscillation $(=1/f)$, s				
Q _{losses}	heat losses, W	τ_t	thermal time constant, s				
Q _{out}	heat transfer rate to the cold heat exchanger, W						
$r_{\rm PE}$	electrical resistivity of the PE, $\Omega.m$		TS .				
R_1, R_2	output and input current resistances, Ω	С	refers to cold fluid				
R_L	load resistance, Ω	H, L	refers to high and low values				
Re_{ω}	Reynolds number for oscillating flow, Eq. (16)	PE	refers to pyroelectric element				
S	piston stroke length, <i>m</i>	min, max refers to minimum and maximum values					
St, St _L	Strouhal numbers, Eq. (15)						

The objectives of the present study are: (1) to design and assemble a fully instrumented pyroelectric energy converter performing the Olsen cycle on co-polymer poly(vinylidene fluoride—trifluoroethylene) [P(VDF-TrFE)], (2) to experimentally measure temporal temperature and pressure evolutions at various locations throughout the device, and (3) to improve the energy and power densities of the pyroelectric energy converter built by Olsen and co-workers [5,10].

2. Current state of knowledge

2.1. Material considerations

Pyroelectric materials possess a spontaneous polarization in the absence of an applied electric field. The spontaneous polarization is strongly dependent of temperature due to the pyroelectric material's crystallographic structure [11]. The displacement of the atoms from their equilibrium positions upon heating and cooling results in the pyroelectric effect. The electric current I_p generated by pyroelectric materials is given by $I_p = Ap_c dT/dt$ where A is the surface area of the pyroelectric material, p_c is the pyroelectric coefficient, and dT/dt is the heating or cooling rate [15]. Note that this is not valid for large temperature changes due to non-linear material behavior [19].

Moreover, ferroelectricity is the ability of a material to have a spontaneous polarization $-P_S$ which can be reversed to $+P_S$ by applying an electric field. Ferroelectric materials have a history dependent response to an applied electric field which results in a hysteresis loop. The electric displacement in the absence of an applied electric field is called the spontaneous polarization P_S while the electric field that is needed to reach zero displacement is called the coercive field. The maximum electric field that the material can withstand without being conductive is called the electric breakdown field or dielectric strength while the maximum electric displacement that can be achieved by the cycle is called the displacement saturation [20]. Some pyroelectric materials such as P (VDF-TrFE) are also ferroelectric.

Paraelectricity is the ability of a material with no spontaneous polarization to become polarized under an applied electric field [21]. Paraelectricity takes place in crystal structures where the electric dipole moments are unaligned, but have the potential to be aligned when subjected to an external electric field.

Curie phase transition corresponds to the material phase change from ferroelectric to paraelectric or vice versa, upon heating or cooling, respectively. The Curie temperature at which the transition from ferro- to paraelectric occurs is denoted by $T_{\text{Curie}, \uparrow}$ and the Curie temperature at which the material transitions from para- to

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ferroelectric is denoted by $T_{\text{Curie, }\downarrow}$. Note that the phase transition from ferroelectric to paraeletric is accompanied by a large discharge of electrical energy. It is, thus, advantageous to heat and cool ferroelectric materials across $T_{\text{Curie, }\uparrow}$ [22].

2.2. Polymeric pyroelectric materials

PVDF and its co-polymer P(VDF-TrFE) have received considerable attention due to their large pyroelectric coefficient used in biomedical and industrial applications [20]. Unlike PVDF, co-polymer P(VDF-TrFE) crystallizes into the ferroelectric β -phase in absence of external stresses [23]. Thus, re-poling P(VDF-TrFE) requires only cooling below $T_{\text{Curie}, \downarrow}$ and under electric poling. On the contrary, re-poling PVDF requires not only electric poling but also mechanical deformation.

Parameters that affect $T_{\text{Curie},\uparrow}$ and $T_{\text{Curie},\downarrow}$ of P(VDF-TrFE) include VDF/TrFE ratio, magnitude of the applied electric field, and polymer processing techniques [24]. It has been established that $T_{\text{Curie},\uparrow}$ increases as VDF/TrFE ratio increases due to an increase in the disorder of the co-polymer domains [20]. In addition, $T_{\text{Curie},\uparrow}$ was shown to increase with the applied electric field [25]. For example, $T_{\text{Curie},\uparrow}$ for 60/40 P(VDF-TrFE) is approximately 66 °C at zero field [26] and increases to 120 °C when the field is 527 kV/cm [25].

2.3. Pyroelectric energy conversion

2.3.1. Olsen cycle

The so-called Olsen cycle is the electric analog of the Ericsson heat engine in the charge–voltage diagram (*q*-*V*) as opposed to the pressure-volume (*p*-*v*) diagram. The cycle was developed by Olsen and co-workers between 1978 and 1985 [10,11,15,20,22,23,27,28]. Fig. 1 shows the *q*-*V* diagram of a typical pyroelectric element (PE) where the Olsen cycle is represented by the 1-2-3-4 loop. A pyroelectric element consists of a pyroelectric thin film and its metallic electrodes deposited on both faces. The Olsen cycle begins by charging the PE at low temperature T_L by increasing the voltage from V_L to V_H (process 1-2). Next, the PE is discharged by heating the element from T_L to T_H at constant voltage V_H where ideally $T_H > T_{\text{Curie}, \uparrow}$ (process 2-3). The material is further discharged by reducing the voltage from V_H to V_L at constant temperature T_H (process 3-4). Finally, the PE is recharged by cooling it from T_H to $T_L < T_{\text{Curie}, \downarrow}$ at constant voltage V_L (process 4-1) [23]. The shaded



Fig. 1. Charge versus voltage for a typical pyroelectric material at different temperatures. The Olsen power cycle is represented by the area between 1-2-3-4.

area in Fig. 1 corresponds to the electrical work output W_E expressed in J/l and given by,

$$W_{\rm E} = \oint V_{\rm PE} dq_{\rm PE} \tag{1}$$

Note that heating and cooling the pyroelectric element above and below the Curie temperatures are not required during the cycle, but doing so could result in a larger electrical energy production as previously discussed.

2.3.2. Pyroelectric energy converters based on olsen cycle

To the best of our knowledge, only five prototypical pyroelectric converters have been built to date. Table 1 summarizes the pyroelectric material and the working fluid used as well as the optimum operating frequency and performance reported in previous studies. For example, Olsen et al.[5] assembled a regenerative and multistage device using lead zirconate stannate titanate (PZST) thin films with various compositions in which Ti⁴⁺ was substituted by Sn⁴⁻ Heat input into the device was provided by a resistive stainless steel heating band while cooling was achieved via a copper cooling coil. A ceramic stack consisting of 100 layers of alumina sheets separated by microchannels filled with working fluid was placed vertically between the heat source and the cold heat exchanger. A piston oscillated the working fluid back and forth between heat source and cold heat exchanger at 185 and 145 °C, respectively. In addition, four PZST pyroelectric elements were placed in thermal contact with the working fluid. The low voltage V_L applied to the PEs was kept constant at 100 V or an electric field of 4 kV/cm while the high voltage V_H was varied between 100 and 700 V (28 kV/cm). The maximum output power density achieved was 33 W/l of PZST at 0.26 Hz for V_H equal to 700 V corresponding to an efficiency of 0.56% [5].

Although PZST has attractive pyroelectric properties for power generation applications, its cost, estimated as ~\$10,000/Watt, makes its use impractical [10]. Therefore, Olsen *et al.*[10] proposed to use inexpensive 73/27 P(VDF-TrFE) thin films 30–70 μ m in thickness. Instead of using a ceramic stack, the PEs were embedded into a spiral stack constructed by alternate layers of plastic sheets and nylon separator screens [10]. The low and high electric field were 230 and 530 kV/cm and the device was able to generate a maximum energy density of 30 J/I [10].

2.3.3. Performance of pyroelectric energy converters

In order to assess the performance of the pyroelectric converter, the thermodynamic energy efficiency of the system over a cycle is defined as [29],

$$\eta = \frac{\dot{W}_{\text{cycle}}}{\dot{Q}_{\text{in}}} = \frac{n_{\text{PE}}\dot{W}_{\text{E}} - \dot{W}_{\text{p}}}{\dot{Q}_{\text{in}}}$$
(2)

where \dot{Q}_{in} (in W) is the heat transfer rate provided to the converter, \dot{W}_{cycle} (in W) is the net power produced by the device, and n_{PE} is the number of pyroelectric elements used for power generation. The electrical power produced by a single PE and the power consumed for pumping the working fluid are denoted by \dot{W}_E and \dot{W}_p , respectively.

2.3.4. Leakage current

A pyroelectric element can be represented by a current generator, a capacitor, and a resistor in parallel. The direction of generated current reverses upon heating and cooling. Ideally, the resistance of the pyroelectric element should be infinite which corresponds to zero leakage across the PE. In reality, however, the resistance is finite and decreases as the applied electric field and H. Nguyen et al. / Applied Thermal Engineering 30 (2010) 2127-2137

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Table 1

	-								
Summ	harv of	design	and	performance	e of	pyroelectric	converters	built to date.	

Ref.	Material	Thickness	Fluid	T_{cold}/T_{hot} (°C)	f (Hz)	Electrodes	$V_L/V_H(V)$	Electrical output	η(%)
[5]	PZST	250 μ m flat stack	Silicone oil (50cst)	145/185	0.14	Ni (sputtered)	100/1100	1.37 W	1.05
[6]	PZST	250 μ m flat stack	Silicone oil (200cst)	146/159	0.14	Ni (sputtered)	0/700	100 J/l/cycle	N/A
[7]	PZST	250 μ m flat stack	Silicone oil (100cst)	145/178	0.128	Ni (sputtered)	300/800	130 J/l/cycle	0.42
[9]	PZST	250 μ m flat stack	Silicone oil (200cst)	110/170	0.04	Ag (fired-on)	0/700	0.36 J/l/cycle	N/A
[10]	P(VDF-TrFE) 73/27	30–70 μ m spiral stack	Silicone oil (200cst)	20/90	0.08-0.5	Al	200/550	30 J/l/cycle	N/A

temperature increase. As a result, a so-called leakage current denoted by I_L flows through the PE, thus, reducing the electrical energy produced [10,15,22,23]. Leakage current increases with increasing temperature and increasing voltage [25].

3. Experimental setup and procedures

A pyroelectric converter was designed and fabricated to directly convert thermal energy into electrical energy based on the Olsen cycle [10]. Fig. 2 shows a cross-section of the device along with a photograph. The device consisted of a thermal and an electrical sub-systems. The reader is referred to Ref.[30] for detailed description and discussion of the device and its operation.

3.1. Thermal sub-system

The thermal sub-system consisted of five major parts, namely (i) the piston-cylinder assembly, (ii) the pyroelectric elements, (iii) the heating source, (iv) the cold heat exchanger, and (v) the working fluid. The piston oscillated the working fluid vertically between the heater located at the top of the device and the cold heat exchanger located at the bottom. Enclosure cylinders made of Teflon were used to guide the motion of the working fluid and acted also as thermal insulation and as a support on which parts were mounted. The cylinder had inner and outer diameters of 38.10 and 57.15 mm,

respectively. Two 3.20 mm diameter holes were drilled onto the part where they were offset from the center by 25 mm. Two 25 cm in length M3 threaded rods were inserted into these holes in order to hold the device together. Teflon was used for outer cylinders because of its low density and thermal conductivity which minimized the device's weight and heat losses. In fact, the density and thermal conductivity of Teflon at room temperature are 357 kg/m³ and 0.25 W/mK, respectively. Teflon can also operate at temperature up to 260 °C [31]. Additionally, Teflon can be easily machined into the desired shapes unlike its ceramic counterparts such as Al₂O₃ used by Olsen *et al.*[5]. Silicone gaskets were placed between enclosing cylinders to prevent the working fluid from leaking out to the environment.

3.1.1. Piston-cylinder assembly

A piston and cylinder sleeve assembly of the Evolution 35GT2 two stroke gasoline engine were purchased from Evolution Engines [32]. The sleeve's inner diameter was 3.5 cm while its height was 6.4 cm. Both the sleeve and the piston were made of stainless steel and the fluid located within the cylinder sleeve was sealed from leaking via the piston ring mounted onto the piston head. A metal rod connected the piston to a flywheel powered by an electric motor model 2709K18 by McMaster-Carr. The piston stroke length was varied from 2.5 to 4.7 cm by changing the mounting radius of the connection between rod and the flywheel.



Fig. 2. (a) Schematic of the device cross-section and its corresponding components and (b) photograph of the actual device (all units are in millimeters).

3.1.2. Pyroelectric assembly and stack

Each pyroelectric element in the pyroelectric assembly was sandwiched between two mica plates to prevent it from moving horizontally and vertically. Fig. 3 shows the pyroelectric assembly and stack integration process. First, a 310 μ m thick mica plate was laid flat on a smooth surface where a 10 mm \times 10 mm window was cut away in the plate's center using a knife. The length of the mica plates was 40.6 mm while their width varied from 20.8 to 38.1 mm so they could be stacked and fit into the Teflon cylinder.

Then, the PE was placed onto the plate and four pieces of 89 μ m thick Kapton tape were used to tape the edges of the PE to the mica plate. Afterwards, an electrical wire was connected to each electrode of the PE using 89 μ m thick copper tape. The wires were then guided outside of the device and connected to the electrical circuit described in Section 3.2. Next, an identical mica plate was pressed down on the edges of the PE. The two plates were bonded to one another via a thin layer of thermal epoxy at four corners. Two pieces of 3.18 mm wide and 330 μ m thick double sided Teflon tape were then adhered to the vertical edges of the mica plate where they bonded to another pyroelectric assembly to create a 330 μ m wide microchannel. Finally, the process was repeated in order to form a stack consisting of 38 pyroelectric elements. Note that the microchannel width can be made wider by using multiple layers of Teflon tape.

P(VDF-TrFE) was chosen as the pyroelectric material for power generation for its lower Curie temperature compared with typical ceramics (e.g., PZST) which makes it attractive for low grade waste heat harvesting applications. In addition, P(VDF-TrFE) has an electric breakdown field that is 25 times greater than that of PZST [10]. The material can be easily processed and costs ten times less than PZST [10]. Mica, on the other hand, was chosen as the PE supporting plates due to its high melting point, high dielectric strength, and its mechanical stiffness. Note that Teflon sheets of 381 and 508 μ m in thickness were also tested to support the PEs. However, the sheets

deformed upon the application of an electric field and/or heating, thus obstructing the microchannels and blocking the working fluid flow.

3.1.3. Heat source and cold heat exchanger

A flexible 50 W AC Kapton electrical heater was used as the heat source. It was taped to a thin copper sheet of dimensions 12.7 cm \times 2.54 cm \times 0.20 mm. The ensemble was subsequently bent to fit inside the Teflon cylinder and placed above the PE stack. The power input could be continuously increased up to a maximum of 50 W by increasing the voltage across the heater. On the other hand, the cold heat exchanger was made of a copper tube with inner and outer diameters equal to 1.59 mm and 3.18 mm (1/16" and 1/8"), respectively. The pipe was 34.3 cm long and was bent into a helical shape before being mounted onto the cold heat exchanger's outer cylinder. Cold water was fed into the coil via a DC electrical pump model W309-11 by Hargraves. The flow rate was adjusted in order to achieve the desired operating conditions.

3.1.4. Working fluid

Dow Corning 50 cst silicone oil was used as the working fluid by analogy with previous studies [5–10]. This oil is a dielectric fluid compatible with electronic components. Its relatively large viscosity ensures that the fluid flow was laminar in the microchannels. Furthermore, the oil's high dielectric strength of 15.7×10^6 V/m at 25 °C [33] and its boiling point of 200 °C at atmospheric pressure make it attractive for pyroelectric power generation.

3.2. Electrical sub-system

Fig. 4 shows the electrical sub-system used in this study to both prepole the pyroelectric film and perform the Olsen cycle. The electrical sub-system controlled the voltage applied to the



Fig. 3. Schematic of the pyroelectric stack assembling process showing (a) a mica plate with a 10×10 mm window, (b) a mica plate with PE, (c) PE sandwiched between two mica plates, (d) pyroelectric assembly with Teflon strips, (e) mating of two mica plates, (f) entire PE stack, and (g) photograph of the actual stack (all dimensions are in millimeters).

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Fig. 4. Schematic of the electrical circuit used to prepole and measure the electrical resistivity r_{PE} of the pyroelectric element and to perform the Olsen cycle.

pyroelectric element and imposed V_L or V_H at the appropriate time during the Olsen cycle. It also measured the power generated by the pyroelectric element by measuring the voltage $V_{\rm PE}$ and charge $q_{\rm PE}$. The triple DC output power supply provided input voltages V_L^* and V_H^* to the high voltage power supply. Two rocker switches were connected to the high voltage power supply terminals to provide either voltage V_L^* or V_H^* . The latter converted the input voltage $V_{L/H}^*$ to output voltage such that $V_{L/H} = 250V_{L/H}^*$. At all times, only one switch was permitted to be opened while the other was closed. The resistor R_L (7.80 M Ω) acted as a voltage divider to scale down the voltage across the resistor R_2 (21.8 k Ω) in order to match the maximum voltage input of 10 V of the data acquisition system (DAQ). Voltage V_1 across capacitor C_1 (1.0 μ F) and voltage V_2 across resistor R_2 were measured via the DAQ and used to calculate the magnitude of the PE's electric displacement D_{PE} and electric field $E_{\rm PE}$. The voltage V_1 was measured through a custom built electrometer to minimize the discharge of capacitor C_1 in the DAQ. The magnitude of the electric displacement D_{PE} was defined as,

$$D_{\rm PE} = \frac{q_{\rm PE}}{A} = \frac{C_1 V_1}{A} \tag{3}$$

where *A* is the surface area of the pyroelectric element. The magnitude of the electric field across the pyroelectric element was computed via Ohm's and Kirchhoff's laws such that,

$$E_{\rm PE} = \frac{V_{\rm PE}}{b} = \frac{V_2(1 + R_L/R_2) - V_1}{b}$$
(4)

where b is the pyroelectric film's thickness. Then, the D-E plot (equivalent to q-V plots) for the pyroelectric element undergoing the Olsen cycle can be generated.

3.3. Power generation analysis

3.3.1. Resistivity

The pyroelectric element was prepoled and its resistivity was measured as a function of time before performing the Olsen cycle. The PE was held at a constant temperature of 85 °C while a constant voltage was applied across the PE. The leakage current I_L was expressed as,

$$I_L = C_1 \frac{\mathrm{d}V_1}{\mathrm{d}t} \tag{5}$$

where V_1 is the registered voltage across capacitor C_1 (Fig. 4). Moreover, the poling electric field applied across the PE computed from Eq. (4) was found to be 201 kV/cm.

Then, the film electrical resistivity, expressed in Ω .m, was determined as,

$$p_{\rm E} = \frac{V_{\rm PE}A}{I_L b} = \frac{E_{\rm PE}A}{I_L} \tag{6}$$

3.3.2. Power generation

Voltages V_1 and V_2 along with temperature T_{PE} were measured and recorded simultaneously as the Olsen cycle was performed. The energy density N_D of one cycle is represented by the enclosed area within the electric displacement versus electric field (*D*-*E*) curve and is given by,

$$N_D = \oint E_{\rm PE} dD_{\rm PE} = \frac{W_E}{\forall} \tag{7}$$

where the volume of the PE is $\forall = Ab$. In practice, the integral was estimated numerically using the trapezoid rule. The power density for a single pyroelectric element, denoted by P_D , is the power generated per liter of pyroelectric material. It is expressed as,

$$P_D = \frac{N_D}{\tau} = \frac{W_E}{\forall} \tag{8}$$

where τ is the period of a cycle and \dot{W}_E is the power generated by the PE.

3.4. Instrumentation

3.4.1. Piston frequency measurements

A transmissive sensor model HOA 0866-N55 by Honeywell was used to measure the frequency of the oscillating piston. It consisted of an infrared emitting diode and a bipolar negative-positivenegative (NPN) silicon phototransistor.

3.4.2. Temperature measurements

In order to determine the local temperatures within the device, type J thermocouples were securely mounted at various locations within the working fluid. Fig. 2 shows the location of the different thermocouples. The temperature T_1 was measured at the center of the cold heat exchanger while T_2 , T_3 , and T_4 were the pyroelectric element' bottom, middle, and top temperatures, respectively. On the other hand, temperature T_5 was measured using a type J thermocouple placed at the center of the heater cylinder. If one defines the *z*-axis along the axis of the device with origin z = 0 set at its base, then, the temperatures T_1 , T_2 , T_3 , T_4 , and T_5 were measured at location *z* = 7.9, 10.8, 11.3, 11.8, and 15.2 cm, respectively. Moreover, the inlet and outlet temperatures of the cold heat exchanger, denoted by $T_{c, i}$ and $T_{c, o}$, were measured with type K thermocouples placed within the feeding pipes. All thermocouples were connected to the DAQ for data recording and were calibrated for temperatures between 0 and 100 °C. Temperatures T_1 , T_2 , T_3 , T_4 , T_5 , $T_{c, i}$, and $T_{c, o}$ were measured at a time interval of 0.05 s.

3.4.3. Heat input and output measurements

A variac model 1010B by Staco Energy Products was used to control the voltage applied to the Kapton electrical heater. Its resistance and output voltage were adjusted to set the device heat input. The input power \dot{Q}_{in} was expressed as,

$$\dot{Q}_{\rm in} = V_{\rm rms} I_{\rm rms} F_p \tag{9}$$

where F_p is the power factor and V_{rms} and I_{rms} are respectively the root mean-square voltage and current, simultaneously measured using a digital multimeter model 34401A by Agilent. The power factor was taken to be unity due to the fact that the heater's alternating current and voltage were in phase.

The mass flow rate \dot{m}_c from the DC electrical pump used to flow cold water through the cold heat exchanger was calibrated as a function of voltage [30]. The voltage across the pump was adjusted and recorded while the Olsen cycle was performed. The heat output removed by the cold heat exchanger was then computed as $\dot{Q}_{out} = \dot{m}_c c_{p,c} (T_{c,o} - T_{c,i})$ where $c_{p,c}$ is the specific heat of water at the mean temperature $(T_{c,o} + T_{c,i})/2$.

Finally, the overall energy balance of the device allows us to determine the performance of the cold heat exchanger and quantify the heat losses to the surroundings according to,

$$\dot{Q}_{\text{losses}} = \dot{Q}_{\text{in}} - n_{\text{PE}}\dot{W}_E - \dot{Q}_{\text{out}}$$
(10)

Where n_{PE} is the number of pyroelectric elements in the device.

4. Results and discussion

4.1. Preliminary tests

4.1.1. Temperature measurements

Fig. 5 shows the temporal evolution of the fluid temperature near the cold heat exchanger T_1 for piston frequency of 0.025 Hz, piston stroke length of 3.4 cm, channel width of 330 μ m, and heat input of 19 W. The minimum and maximum values of temperature T_1 were 21.4 and 49.7 °C, respectively. Note that the temperature oscillations featured a frequency of 0.025 Hz in good agreement with the piston frequency measured independently. Similar results were obtained with the pressure at the bottom of the heat exchanger [30].

Moreover, Fig. 6 shows the temporal evolution of temperatures T_2 , T_3 , and T_4 along the pyroelectric assembly. The temperature swing ΔT_i at location "*i*" is defined as,

$$\Delta T_i = T_{i,\max} - T_{i,\min} \tag{11}$$

where $T_{i,\max}$ and $T_{i,\min}$ are the maximum and minimum temperatures reached by T_i during one cycle. In addition, the arithmetic mean temperature $\overline{T_i}$ at location "*i*" is defined as,



Fig. 5. Temporal temperature oscillation near the cold heat exchanger T_1 . The piston oscillating frequency and stroke length, the stack channel width, and the heat input were constant and equal to 0.025 Hz, 3.4 cm, 330 μ m, and 19 W respectively.



Fig. 6. Temporal evolutions of the working fluid temperatures at the bottom (T_2) , middle (T_3) , and top (T_4) of the pyroelectric element. The piston oscillating frequency and stroke length, stack channel width, and the heat heat input were constant and equal to 0.025 Hz, 3.4 cm, 330 μ m, and 19 W, respectively.

$$\overline{T}_i = \frac{T_{i,\max} + T_{i,\min}}{2} \tag{12}$$

Fig. 6 indicates that T_3 , measured in the middle of the PE, oscillated between 90.7 and 66.7°C, resulting in a temperature swing $\Delta T_3 = 24$ °C. Similarly, T_2 oscillated between 85.9 and 62.9 °C while T_4 oscillated between 94.6 and 71.6 °C, resulting in a temperature swing $\Delta T_2 = \Delta T_4 = 23$ °C. As expected, the maximum temperature occurred when the piston was at the bottom of its stroke while the minimum temperature occurred when the piston was at the top of its stroke. Here also, the temperature oscillations had the same frequency as that of the piston. In the remaining of this study, the temperature T_3 along with \overline{T}_3 and ΔT_3 will be used as representative of the PE thermal response to oscillation of the working fluid.

4.1.2. Power generation

A pyroelectric element made of P(VDF-TrFE) with VDF-TrFE weight ratio of 60/40 was prepoled in situ before performing the Olsen cycle. Fig. 7 plots the electrical resistivity r_{PE} of a 1 cm × 1 cm and 45.7 μ m thick P(VDF-TrFE) film measured as a function of time. The film was poled at $E_{PE} = 201$ kV/cm and at $T_3 = 85$ °C while the piston and the working fluid were at rest. The film resistivity r_{PE} increased rapidly during the first 10 min of poling. After 140 min, the resistivity was $4.54 \times 10^{10} \Omega$.m and did not change noticeably as poling continued. Thus, the poling process was stopped after 180 min. Similar poling procedure was followed for every runs reported in this study.

Once the poling process was completed, the Olsen cycle was performed on 45.7 μ m thick PEs. Fig. 8 shows the corresponding electrical displacement D_{PE} versus electric field E_{PE} . Here, the device had a stroke length of 4.7 cm and 330 μ m wide channels while operating at 0.061 Hz. The value of the low and high electric fields imposed in the Olsen cycle were $E_L = 202 \text{ kV/cm}$ and $E_H = 379 \text{ kV/cm}$, respectively. The applied low electric field E_L was set at 202 kV/cm as suggested by Olsen *et al.*[10,25] in order to keep the film properly poled during process 4–1 of the Olsen cycle. The heat input provided by the heater was 30.9 W. The minimum and maximum temperatures measured at the center of the PE, $T_{3, \min}$ and $T_{3, \max}$, were 66.4 and 83.0 °C, respectively corresponding to $\Delta T_3 = 16.6 \text{ °C}$ and $\overline{T}_3 = 74.7 \text{ °C}$. The corresponding energy and

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Fig. 7. Logarithmic of electrical resistivity of a 1 cm \times 1 cm and 45.7 μ m thick 60/40 P (VDF-TrFE) film as a function of time during poling at 201 kV/cm and $T_3 = 85$ °C.

power densities computed using Eqs. (7) and (8) were 130 J/l and 8.12 W/l of pyroelectric material, respectively.

4.1.3. Effect of temperature

Fig. 9 shows the energy density of a pyroelectric element as a function of arithmetic mean temperature at the center of the pyroelectric element \overline{T}_3 . Here, the high field E_H was set to either 257 or 329 kV/cm. The channel width, stroke length, and frequency were 1 mm, 3.4 cm, and 0.025 Hz, respectively. In addition, the applied low electric field E_L was set at 202 \pm 1 kV/cm. The heat input provided by the electrical heater was adjusted in order to maintain \overline{T}_3 at the desired temperature. Fig. 9 shows that a maximum energy density of 83 J/l of pyroelectric material was



Fig. 8. Electric displacement versus electric field (*D-E*) for a 1 cm \times 1 cm and 45.7 μ m thick pyroelectric element. The piston stroke length, frequency, and channel width were 4.7 cm, 0.061 Hz, and 330 μ m, respectively.



Fig. 9. Energy density as a function of \overline{T}_3 . The channel width, piston stroke length and frequency, and low electric field were constant and equal to 1 mm, 3.4 cm, 0.025 Hz, and 202 kV/cm, respectively while the heat input by the electrical heater was adjusted to maintain \overline{T}_3 at the desired temperature.

achieved when \overline{T}_3 was about 74 \pm 1 °C and E_H was 329 kV/cm. Exposing the center of the PE to a temperature \overline{T}_3 larger than 74 °C decreased the energy density of the pyroelectric element due to an increase in leakage current. On the other hand, exposing the center of the PE to a temperature \overline{T}_3 smaller than 74 °C resulted in smaller energy density due to a smaller temperature swing and to the fact that the PE may not undergo the ferro- to paraelectric phase transition. Indeed, the Curie temperature $T_{\text{Curie}, \uparrow}$ for 60/40 P(VDF-TrFE) was reported to be equal to 90 and 93 °C for 257 and 329 kV/cm electric field, respectively [25].

4.1.4. Preliminary conclusion

Preliminary tests showed that, as expected, the device's local temperatures and pressure oscillated sinusoidally at the same frequency as the piston. Moreover, electrical power was successfully generated by performing the Olsen cycle on 60/40 P(VDF-TrFE) films. The following sections report a systematic study of the effect of different parameters on the device's temperature swing, heat input, and electrical power generated. In all cases, the heat input was adjusted to maintain the arithmetic mean temperature at the center of the pyroelectric element \overline{T}_3 at 74 ± 1 °C found to achieve the best compromise between large temperature swing and minimum leakage current.

4.2. Effect of channel width

Fig. 10 compares the temperature swing ΔT_3 at the center of the PE for channel width equal to 330 μ m and 1 mm at frequency ranging from 0.025 to 0.16 Hz. The stroke length was equal to 3.4 cm. Fig. 10 indicates that the temperature swing ΔT_3 decreased with increasing frequency. This can be attributed to thermal inertia of both the working fluid and the pyroelectric element. As the frequency increased, there was less time for thermal energy to be exchanged between the heating band and the oscillating working fluid and between the working fluid and the pyroelectric elements. This limitation can be addressed by lowering the thermal time constant of the pyroelectric element defined as [34],



Fig. 10. Temperature swing ΔT_3 at the center of the pyroelectric element as a function of frequency measured for stack channel width equal to 330 μ m and 1 mm. The stroke length and the mean temperature \overline{T}_3 were 3.4 cm and 74 \pm 1 °C, respectively.

$$\tau_t = \frac{\rho_{\rm PE} c_{p,\rm PE} \forall}{h_w A} \tag{13}$$

where h_{ω} is the oscillatory flow convective heat transfer coefficient. The thermal time constant can be reduced by increasing h_{ω} and the pyroelectric element surface area *A* and/or by decreasing the thickness of the pyroelectric element $b = \forall / A$.

Moreover, Fig. 10 shows that a larger temperature swing was achieved with 330 μ m wide channels compared with 1 mm wide channels for the same \overline{T}_3 value at all frequencies considered. Ozawa *et al.*[35] developed a correlation between the Strouhal, Reynolds, and Nusselt numbers for laminar oscillating flow in a circular channel given by,

$$Nu_{\omega} = \frac{h_{\omega}l_0}{k_f} = 0.92St^{-0.2}St_L^{-0.26}Pr_f^{0.4}Re_{\omega}^{0.44}$$
(14)

where l_0 and k_f are the amplitude of fluid displacement, and the thermal conductivity of the working fluid, respectively. The Strouhal numbers are denoted by *St* and *St*_L and defined as [35],

$$St = D_h/S$$
 and $St_L = L/S$ (15)

Where D_h is the channel hydraulic diameter, *S* is the piston stroke length, and *L* is the channel length. In addition, the Reynolds and Prandtl numbers are denoted by Re_{ω} and Pr_f and defined as [35],

$$Re_{\omega} = \frac{u_0 l_0}{v_f} = \frac{u_0^2}{v_f \omega}$$
 and $Pr_f = \frac{v_f}{\alpha_f}$ (16)

where v_f , $\omega = 2\pi f$, $u_0 = l_0 \omega$, and α_f are the fluid kinematic viscosity, piston angular frequency, amplitude of velocity oscillation, and fluid thermal diffusivity, respectively. As the channel width decreased, the hydraulic diameter decreased while the velocity of the working fluid increased thus, increasing the Strouhal *St* and Reynolds Re_{ω} numbers. This, in turns, resulted in larger Nusselt number and heat transfer coefficient between the working fluid and the pyroelectric material, as suggested by Eq. [14] leading to larger temperature swing in the PE.



Fig. 11. (a) Temperature swing ΔT_3 at the center of the PE and (b) the associated heat input \dot{Q}_{in} as a function of frequency for piston stroke lengths of 2.5, 3.4, and 4.7 cm. The channel width was 330 μ m and the mean temperature \overline{T}_3 was 74 ± 1 °C.

4.3. Effect of frequency and stroke length

Fig. 11a compares the temperature swing ΔT_3 at the center of the PE as a function of frequency for piston stroke length equal to 2.5, 3.4, and 4.7 cm. It indicates that the temperature swing decreased as the frequency increased regardless of the stroke length, as previously discussed. Moreover, for a given frequency, the temperature swing increased as the stroke length increased from 2.5 to 4.7 cm. Increasing the stroke length to 4.7 cm allowed a fluid particle located at the heating band when the piston was at its highest position to carry the thermal energy down to the bottom of the PE as the piston reached its lowest position. As a result, a larger temperature swing was observed in the PE.

Fig. 11b shows the heat input \dot{Q}_{in} provided by the electrical heater to the working fluid as a function of piston frequency for stroke length equal to 2.5, 3.4, and 4.7 cm. Data shown in Fig. 11b correspond to temperature swing ΔT_3 shown in Fig. 11a. It establishes that heat

input required to ensure the PE temperature $\overline{T}_3 = 74 \pm 1$ °C increased with frequency and stroke length. Indeed, as the frequency increased, the time during which thermal energy can be exchanged between the heating band and the working fluid decreased. This required a larger heat input to maintain \overline{T}_3 at its prescribed value.

4.4. Energy and power densities

4.4.1. Effect of high electric field

Fig. 12 compares the energy density N_D as a function of high electric field E_H between 220 and 415 kV/cm for frequencies equal to 0.035, 0.061, and 0.12 Hz. The piston stroke length, channel width, and low electric field were 4.7 cm, 330 μ m, and 202 kV/cm, respectively. It shows that a maximum energy density of 130 J/l of pyroelectric material was obtained at 0.061 Hz with a high electric field E_H of 379 kV/cm. For E_H above 379 kV/cm, the energy density decreased due to increasing leakage current. On the other hand, applying E_H smaller than 379 kV/cm decreased the difference in electric field span (E_H – E_L) and reduced the energy density as suggested by Eq. (7). Thus, the optimum value of E_H to maximize the energy density was found to be 379 kV/cm.

4.4.2. Effect of frequency

Fig. 13 plots the maximum energy and power densities as a function of frequency. The piston stroke length, stack channel width, and arithmetic mean temperature at the center of the pyroelectric element \overline{T}_3 were constant and equal to 4.7 cm, 330 μ m, and 74 \pm 1 °C, respectively. The low and high electric fields were $E_L = 202 \text{ kV/cm}$ and $E_H = 379 \text{ kV/cm}$ as suggested by previous results. Even though the temperature swing at 0.035 Hz was larger than that at 0.061 Hz, the associated energy density was smaller. Indeed, the larger cycle time at 0.035 Hz provided an opportunity for charges at the surface of the pyroelectric element to conduct through its body (leakage current) thus dissipating the energy as Joule heating. On the other hand, operating the device at a frequency larger than 0.061 Hz decreased the energy density of the pyroelectric material since the temperature swing was smaller as indicated in Fig. 11. Therefore, achieving the maximum energy density of the pyroelectric element was a compromise between



Fig. 12. Comparison of energy density for different frequencies as a function of high electric field E_H . The channel width, stroke length, low field, and mean temperature \overline{T}_3 were constant and equal to 330 μ m, 4.7 cm, 202 kV/cm, and 74 \pm 1 °C, respectively.



Fig. 13. Maximum energy and power densities as a function of frequency. The channel width, stroke length, and mean temperature \overline{T}_3 were constant and equal to 330 μ m, 4.7 cm, and 74 \pm 1 °C, respectively.

large temperature swing and small cycle time and leakage current. Fig. 13 also indicates that a maximum power density P_D of 10.7 W/l of pyroelectric material was obtained at 0.12 Hz. Although the energy density of the pyroelectric element was smaller at 0.12 Hz compared with 0.061 Hz, the fact that the cycle time was smaller resulted in a larger power density. Operating the device at frequency larger than 0.12 Hz, on the contrary, resulted in smaller power output due to a large reduction in output energy density caused by smaller temperature swing ΔT_3 .

4.5. Heat losses to the surroundings

Efforts have been made to reduce the device heat losses to the surrounding by using Teflon for the device outer cylinders and fiberglass wool to entirely insulate the device. The device heat losses were estimated by performing an overall energy balance for the device. Heat losses were found to range from 15 to 20% of the total heat input \dot{Q}_{in} and increased as the operating temperature and heat input increased. For example, at 0.025 Hz, the heat input by the electrical heater was 26.6 W and the heat losses were 4.4 W. However, at 0.082 Hz, the heat input was 34.6 W and the heat losses were 6.7 W. In both cases, the stroke length, \overline{T}_3 , and channel width were 4.7 cm, 74 \pm 1 °C, 330 μ m, respectively. Because heat losses decrease the efficiency of the device, additional care must be taken to lower this parasitic losses. One solution is to use better insulating materials such as pyrogel whose thermal conductivity is half that of fiberglass wool [36].

4.6. Efficiency

The device efficiency increased as the high electric field increased but reached a maximum at 379 kV/cm. The maximum efficiency for 0.061 and 0.12 Hz were 0.045% and 0.053%, respectively. Applying an electric field E_H smaller than 379 kV/cm decreased the power output and therefore the efficiency due to smaller field span and charge displacement. Moreover, applying an electric field E_H larger than 379 kV/cm decreased the efficiency due to larger current leakage through the PE. Note that the device was not optimized to achieve maximum efficiency but to maximize the energy and power densities per unit volume of PE. In particular, the efficiency was smaller than that reported by Olsen *et al.*[5] due to

(1) a smaller number of pyroelectric elements n_{PE} in the stack (38 compared to 100) and (2) smaller surface area of the pyroelectric element used for power generation.

5. Conclusion

This study was concerned with the design, assembly, and operation of a pyroelectric converter for harvesting waste heat by performing the Olsen cycle on 60/40 P(VDF-TrFE) thin films. First, the underlying physical principles were presented. Then, the pyroelectric device assembled and the associated thermal and electrical sub-systems were described in details. Finally, the experimental results were reported and discussed. The following conclusions can be drawn:

- 1. Temperature swing at the pyroelectric element decreased as frequency increased for constant arithmetic mean temperature at the center of the pyroelectric element \overline{T}_3 .
- 2. The greatest increase in the device's energy and power densities was caused by reducing the channel width and increasing the piston stroke length.
- 3. For the same frequency, stroke length, and \overline{T}_3 , the temperature swing ΔT_3 increased as the channel width decreased.
- 4. ΔT_3 also increased as stroke length increased. The optimum stroke length was theoretically determined to be 4.7 cm for the device assembled in this study.
- 5. In order to maximize the pyroelectric element energy and power densities, it was best to operate the device where the arithmetic mean temperature at center of the pyroelectric element \overline{T}_3 was equal to 74 \pm 1 °C identified as the best compromise between large temperature swing and excessive leakage current.
- 6. Heat input required to maintain the mean temperature \overline{T}_3 at 74 \pm 1 °C increased as stroke length and frequency increased.
- 7. The optimum high electric field E_H in the Olsen cycle was 379 kV/cm while E_L was set at 202 kV/cm.
- The largest energy density achieved by the device was 130 J/l of pyroelectric material at 0.061 Hz while the largest power density was 10.7 W/l of pyroelectric material at 0.12 Hz. To the best of our knowledge, this is the largest energy density achieved by any pyroelectric energy converter using polymeric pyroelectric material P(VDF-TrFE).

Such a device could be used for waste heat recovery from internal combustion engines or various electronic devices. Future research should focus on (i) further reducing the channel width to increase the temperature swing and the number of pyroelectric elements in the stack to increase the device power density and the efficiency and on (ii) synthesizing inexpensive pyroelectric thin films with large surface area which feature large spontaneous polarization, electric breakdown field, and electrical resistivity at all operating temperatures.

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