

# Laminate composites with enhanced pyroelectric effects for energy harvesting

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**Abstract.** Pyroelectric coefficient enhanced 2-2 connectivity laminate composites' energy harvesting credentials have been assessed. The use of the electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) for such assessment has been appraised while the experimental samples are evaluated to show significant improvement in their performance via pyroelectric coefficient enhancement, demonstrative of their great potential in energy harvesting application. Lead zirconate titanate and stainless steel laminate composite with 88% pyroelectric coefficient enhancement is shown to increase its maximum power density, efficiency, and electrothermal coupling factor by 254%, while other material pairings have also been evaluated to exhibit great promise in this application owing to large pyroelectric coefficient enhancement accompanied by reduction in total thermal mass.

**Keywords :** Pyroelectric, laminate composites, energy harvesting

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## 1. Introduction

Recently, there has been a large increase in research being conducted on self-powering systems and energy harvesting applications form an integral part of this. Many different miniature energy harvesting technologies are being considered at present, solar cells, thermoelectricity, Micro fuel cells, and Micro engines to name a few, and this article deals with the one utilizing pyroelectric (PY) effect. This potential use of temporal temperature gradient in energy harvesting has already been studied by various research groups such as Olsen *et al* [1], Sebald *et al* [2], and Xie *et al* [3]. In particular, Sebald *et al* have recently published a number of works on pyroelectric energy harvesting application with emphasis on potential benefits of the non-linear pyroelectricity via Ericsson thermodynamic cycles around the phase transition region. We have recently investigated pyroelectric coefficient enhancement in 2-2 connectivity laminate composites through the improvement in the secondary pyroelectric coefficient [4, 5, 6] under both short and open circuit conditions. This article is intended to demonstrate how this enhanced pyroelectric coefficient can improve the outputs of the pyroelectric energy harvesting application.

By exploiting the symmetries of 2-2 connectivity laminates composites of PY and non-pyroelectric (NP) materials, we have previously reported theoretical pyroelectric coefficient (PY coef) enhancements of up to 800% in PZT5H-Chlorinated polyvinyl chloride thermoplastic (CPVC) composites [5] while experimentally demonstrating substantial PY coef enhancements of more than 100% in PZT-Stainless steel (St)

laminar structures[4]. As it will become evident in section 3, the parameters representative of the energy output in PY energy harvesting application are proportional to  $p_m^2$  where  $p_m$  is the PY coef along the m-axis. Hence this increase in PY coef should result in very large enhancements in the outputs of the PY energy harvesting application and this enhancement will be investigated in laminate composites of PY materials such as Lead zirconate titanate (PZT5H and PZT5A), Barium titanate (BTO), Lithium tantalate (LTO), Lithium niobate (LNO), and Poly-vinylidene fluoride (PVDF), paired with NP materials such as Stainless steel (St), Poly-tetrafluoroethylene (PTFE or Teflon), Chlorinated polyvinyl chloride thermoplastic (CPVC), Aluminium (Al), Zinc (Zn), and Invar 36 (Invar36).

In section 2, a schematics of a simple pyroelectric energy harvesting device which converts a spatial temperature gradient into a temporal one will be exhibited, while the theoretical treatment on its potential performance will be presented in section 3 via the derivation of expected maximum power density, efficiency, and electrothermal coupling factor. The findings from these analyses on PY-NP laminate composites will be presented in section 4, followed by our conclusion in section 5.

## 2. Pyroelectric energy harvesting device

Figure 1 is a schematic diagram of our PY energy harvesting device. It employs one-way Shape memory alloy (SMA)[7] springs with steel ones for returning SMA springs into their original shape and acts as a kind of heat pump transferring heat from hot to cold surfaces/reservoirs. Note that  $\Theta_{PY}$  is the temperature of PY element,  $\Theta_{SMA}$  the temperature of SMA springs.

Cyclic behaviour from Figures 1(a) to 1(f) then back to 1(a) transforms the spatial temperature gradient between  $\Theta_H$  (High temperature reservoir and temperature at which SMA is in austenite phase) and  $\Theta_L$  (Low temperature reservoir and temperature at which SMA is in martensite phase) surfaces into a temporal one. Although the existence of the thermal insulation layer is not essential, it does enable a much simpler phenomenological view of the overall operation while enhancing the actuation behaviour of SMA springs and decreasing the thermal diffusion process. Spring arrangements in figure 1 should maximise the surface contact between the PY element and temperature surfaces, aiding thermal conduction.

## 3. Mathematical treatment of the potential energy output

In this section, we shall derive the mathematical expressions for analyzing the performance of the PY energy harvesting device presented in section 2. This theoretical consideration can be applied to any linear pyroelectric energy harvesting application where the thermal stimulus on PY element can be approximated by sinusoidal temperature variation.

### 3.1. Maximum power density

In order to derive the expression for potential power output from the device depicted in figure 1, we will use similar technique to that employed by Ren *et al* [8] and Shu and Lien[9] in the piezoelectric energy harvesting application. A simple resistive cycle case in Sebald *et al* 's work[2] and standard interface in Lefeuvre *et al* 's work[10] should be analogous to this.

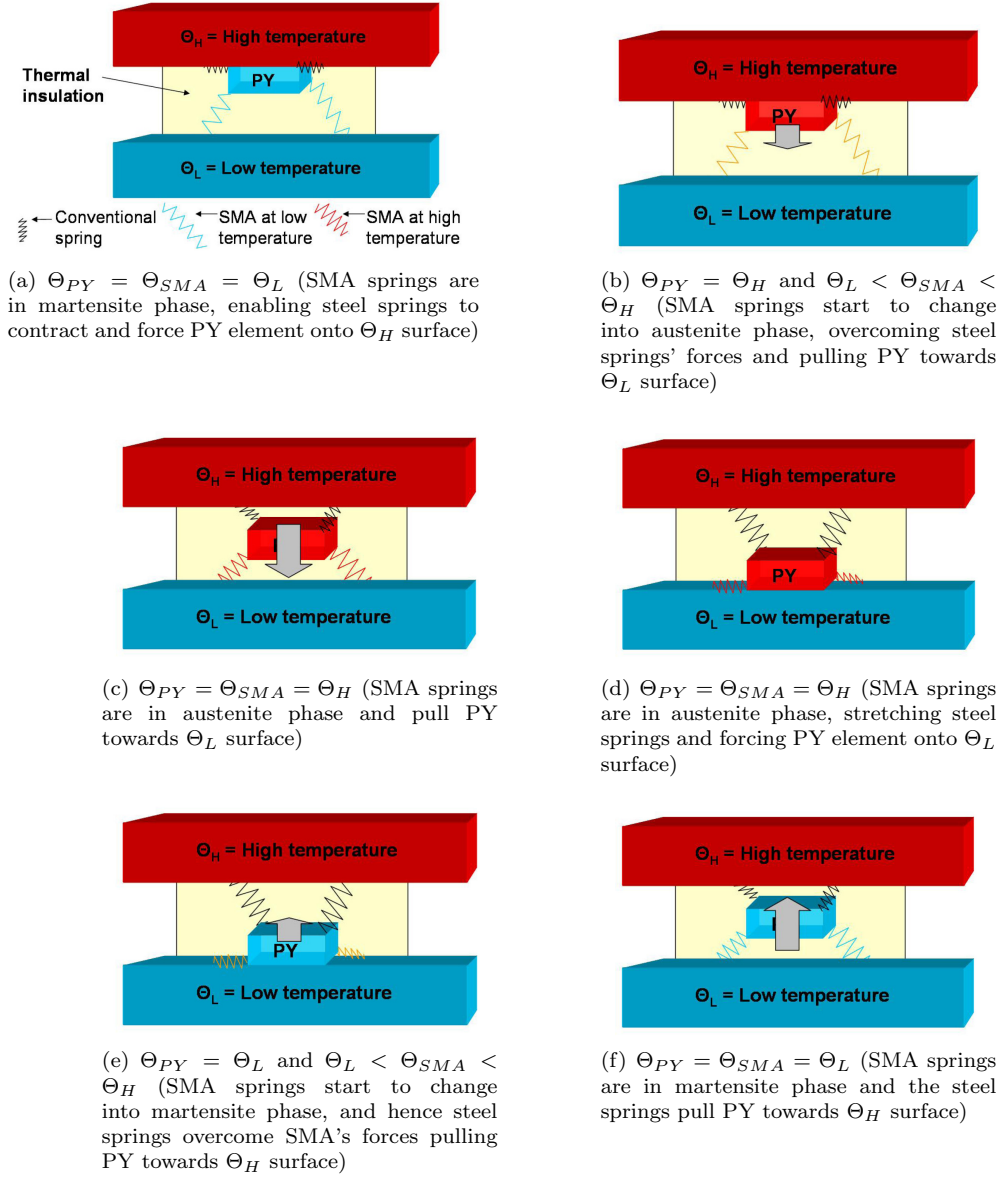


**Figure 1.** Schematics of a pyroelectric energy harvesting device where  $\Theta_{PY}$  and  $\Theta_{SMA}$  are the temperatures of PY element and SMA, respectively.

According to the first and second law of thermodynamics, the reversible change  $dU$  in the internal energy  $U$  of an elastic dielectric subjected to a small change of the strain  $dS$ , electric displacement  $dD$ , and entropy  $d\sigma$  is given by;

$$dU = \Theta d\sigma + T_{kl} dS_{ij} + E_n dD_m \quad (3.1)$$

where  $\Theta$  is the temperature of the material.

If one wishes to investigate systems under isothermal conditions, and use electric field,  $E$ , and stress,  $T$ , as the independent variables, a Legendre transformation of  $U$

has to be performed by adding the expression  $-S_{ij}T_{kl} - D_m E_n - \sigma\Theta$  to  $U$ . This results in the following free energy function, which is also known as the Gibbs free energy,  $G$ , of a piezoelectric crystal[11, 12];

$$G = U - S_{ij}T_{kl} - D_m E_n - \sigma\Theta \quad (3.2)$$

where  $i, j, k, l, m, n = 1..3$ .

Any natural process occurs if and only if the associated change in  $G$  of the process is negative. Likewise, a system reaches an equilibrium when the associated change in  $G$  is zero. We now assume constant external electric field, i.e.  $dE_n = 0 \forall n$ , from the definition of short circuit condition[5] and choose the temperature ( $\Theta$ ), stress ( $T_{ij}$ ) and electric field ( $E_m$ ) as the independent variables. By considering the conventional nine components of the second order strain and stress tensors, while the magnetic effect is ignored as usual, we get the expression for the change in electric displacement under short circuit condition (3.3).

$$\begin{aligned} dD_m &= \sum_k \sum_l \left( \frac{\partial D_m}{\partial T_{kl}} \right)_{E, \Theta} dT_{kl} + \sum_n \left( \frac{\partial D_m}{\partial E_n} \right)_{T, \Theta} dE_n \\ &\quad + \left( \frac{\partial D_m}{\partial \Theta} \right)_{T, E} d\Theta \\ &= d_{mkl}^{E, \Theta} dT_{kl} + \varepsilon_{mn}^T dE_n + p_m^{T, E} d\Theta \end{aligned} \quad (3.3)$$

Assuming the PY element is free to deform and even 2-2 connectivity laminate composites behave in a homogeneous manner in this freedom ( $dT_{kl} = 0$ ), 3.3 can be simplified.

$$dD_m = \varepsilon_{mn}^T dE_n + p_m^{T, E} d\Theta \quad (3.4)$$

Let thermal stimulus, namely temperature variation inside PY element, be  $\Theta$ ,  $f$  frequency ( $\omega = 2\pi f$ ),  $t$  time, and  $\Theta_0$  amplitude of temperature variation ( $\Theta_0 = (\Theta_H - \Theta_L)/2$ ).

$$\begin{aligned} \Rightarrow \quad \Theta &= \Theta_0 \exp(i\omega t) + \frac{\Theta_L + \Theta_H}{2} \\ \therefore \frac{d\Theta}{dt} &= i\omega\Theta_0 \exp(i\omega t) \\ &\quad \text{with thermal variation assumed to be sinusoidal} \end{aligned} \quad (3.5)$$

Now,  $dQ_m = AdD_m$  and  $dE_n = dV_n/PY t$  where  $Q_m$  is the charge on the surface,  $A$  surface area,  $V_n$  potential difference across the thickness of PY element, and  $PY t$  the thickness of PY element.

From 3.4:

$$\begin{aligned} \Rightarrow \quad dQ_m &= AdD_m = A (\varepsilon_{mn}^T dE_n + p_m^{T, E} d\Theta) \\ &= \frac{A}{PY t} \varepsilon_{mn}^T dV_n + Ap_m^{T, E} d\Theta \end{aligned} \quad (3.6)$$

From 3.5:

$$\begin{aligned}
\Rightarrow I_m &= \frac{dQ_m}{dt} = \frac{A}{PYt} \varepsilon_{mn}^T \frac{dV_n}{dt} + Ap_m^{T,E} \frac{d\Theta}{dt} \\
&= \frac{A}{PYt} \varepsilon_{mn}^T \frac{d\{V_0 \exp(i(\omega t + \theta))\}}{dt} \\
&\quad + Ap_m^{T,E} (i\omega \Theta_0 \exp(i\omega t)) \\
&= \frac{A}{PYt} \varepsilon_{mn}^T \{i\omega V_0 \exp(i(\omega t + \theta))\} \\
&\quad + i\omega \Theta_0 Ap_m^{T,E} \exp(i\omega t)
\end{aligned} \tag{3.7}$$

where  $I_m$  = Current generated from PY element

$V_0$  = Amplitude of the potential difference generated across PY element

$\theta$  = Phase difference between the sinusoidal thermal variation and the potential difference across PY element

Equation 3.7 implies the generated potential difference or voltage across a resistor  $R_0$  (total external load resistance) will be:

$$\begin{aligned}
V &= dV_n = V_0 \exp(i(\omega t + \theta)) = R_0 I_m \\
&= R_0 \left[ \frac{i\omega A \varepsilon_{mn}^T}{PYt} V_0 \exp(i\theta) + i\omega \Theta_0 Ap_m^{T,E} \right] \exp(i\omega t) \\
\Rightarrow V_0 \exp(i\theta) &= \frac{i\omega R_0 \Theta_0 Ap_m^{T,E}}{1 - i\omega \frac{R_0 A \varepsilon_{mn}^T}{PYt}}
\end{aligned} \tag{3.8}$$

Hence power dissipation averaged over time in the load resistor  $R_0$  is:

$$\begin{aligned}
\text{Power} = P &= \frac{V_0^2}{2R_0} = \frac{\omega^2 R_0 \Theta_0^2 A^2 (p_m^{T,E})^2}{2 \left[ 1 + \frac{\omega^2 R_0^2 A^2 (\varepsilon_{mn}^T)^2}{PYt^2} \right]} \\
&= \frac{\omega^2 R_0 \Theta_0^2 A^2 (p_m^{T,E})^2}{2 [1 + \omega^2 R_0^2 C_0^2]}
\end{aligned} \tag{3.9}$$

where  $C_0$  = Capacitance of PY element =  $\frac{\varepsilon_{mn}^T A}{PYt}$

Note that this power dissipation in 3.9 is the power generated from PY element, which in turn gets dissipated by the external load resistor  $R_0$ . This expression reaches its maximum when  $R_0 = 1/(\omega C_0)$ , i.e. when  $R_0$  matches the impedance of the voltage source.

$$\begin{aligned}
\Rightarrow P_{max} &= \text{Maximum power dissipation} \\
&= \frac{\omega \Theta_0^2 A^2 (p_m^{T,E})^2}{4C_0} \\
&= \frac{\omega \Theta_0^2 A (p_m^{T,E})^2}{4 \frac{\varepsilon_{mn}^T}{PYt}} = \frac{PYt \omega \Theta_0^2 A (p_m^{T,E})^2}{4 \varepsilon_{mn}^T}
\end{aligned} \tag{3.10}$$

Also from 3.9:

$$\begin{aligned}
\Rightarrow P_{den} &= \text{Power density} = \frac{\text{Power}}{\text{Volume}} \\
&= \frac{\omega^2 R_0 \Theta_0^2 A^2 (p_m^{T,E})^2}{2 [1 + \omega^2 R_0^2 C_0^2] \times A (N^P t + P^Y t)} \\
&= \frac{\omega^2 R_0 \Theta_0^2 A (p_m^{T,E})^2}{2 [1 + \omega^2 R_0^2 C_0^2] (N^P t + P^Y t)} \\
\Rightarrow P_{denMax} &= \text{Maximum power density} \\
&= \frac{\omega \Theta_0^2 A (p_m^{T,E})^2}{4 C_0 (N^P t + P^Y t)} \\
&= \frac{\omega A \Theta_0^2 (p_m^{T,E})^2}{4 \frac{\varepsilon_{mn}^T A}{P^Y t} (N^P t + P^Y t)} \\
&= \frac{\omega \Theta_0^2 (p_m^{T,E})^2}{4 \varepsilon_{mn}^T \left( \frac{N^P t}{P^Y t} + 1 \right)} \\
&= \frac{2\pi f \Theta_0^2 (p_m^{T,E})^2}{4 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right)} = \frac{\pi f \Theta_0^2 (p_m^{T,E})^2}{2 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right)} \\
&= \frac{\pi f (p_m^{T,E})^2 (\Theta_H - \Theta_L)^2}{2 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right) \times 4} \\
&= \frac{\pi f (p_m^{T,E})^2 (\Theta_H - \Theta_L)^2}{8 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right)}
\end{aligned} \tag{3.11}$$

where  $R = P^Y t / N^P t$  is the thickness ratio,  $\omega = 2\pi f$  with  $f$  being frequency in  $Hz$ , and the unit of Power density being  $Watts m^{-3}$  or  $W m^{-3}$ .

### 3.2. Efficiency and electrothermal coupling factor

Equation 3.11 means:

$$\begin{aligned}
\Rightarrow W &= \text{Maximum electrical energy output} \\
&= P_{denMax} \times \tau_{H-L} \times Vol \\
&= \frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)^2}{8 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right)} \times (f \times \tau_{H-L}) \times Vol \\
&= \frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)^2}{16 \varepsilon_{mn}^T \left( \frac{1}{R} + 1 \right)} \times Vol
\end{aligned} \tag{3.12}$$

where  $\tau_{H-L} = 1/(2f)$  is the time taken for  $\Theta_{PY}$  to vary from  $\Theta_H$  to  $\Theta_L$  (this would be half of the period of this sinusoidal wave),  $Vol$  is the volume, and  $W$  is the maximum electrical energy output measured in *Watts*.

Note that  $W$  is evaluated over a period of half a thermal variation cycle, i.e. from figures 1(b) to 1(e), since the next half cycle will have exactly the same electrical characteristics except the direction of polarisation/current/voltage being reversed,

which can be taken care of using AC-DC converter circuit incorporating rectifier bridge.

In order to make comparisons with various other PY energy harvesting arrangements, techniques employed in Sebald *et al* 's work[2], namely evaluation and comparison of the “Efficiency ( $\eta$ )” and “Electrothermal coupling factor ( $k^2$ )”, will need to be carried out. Note that  $\eta$  is a more application specific version of our “Efficiency (*Eff*)” expressions in our previous publication[5].

For the time period of  $\tau_{H-L} = 1/(2f)$  one can also calculate the amount of thermal energy input used to bring about the temperature change of PY element from  $\Theta_H$  to  $\Theta_L$ .

$$\begin{aligned}
 Q_h &= \text{Heat taken from hot reservoir} \\
 &= \text{Thermal energy input} \\
 &= Vol \times \bar{c}_E \int_0^{\tau_{H-L}} \left( \frac{d\Theta}{dt} \right) dt \\
 &= Vol \times \bar{c}_E [\Theta]_{t=0}^{t=\tau_{H-L}} = Vol \times \bar{c}_E \times 2\Theta_0 \\
 &= \frac{R^{PY}c_E + {}^{NP}c_E}{R+1} \times (\Theta_H - \Theta_L) \times Vol
 \end{aligned} \tag{3.13}$$

where  $\bar{c}_E = \frac{A({}^{PY}c_E {}^{PY}t + {}^{NP}c_E {}^{NP}t)}{A({}^{PY}t + {}^{NP}t)} = \frac{R^{PY}c_E + {}^{NP}c_E}{R+1}$  is the volumetric heat capacity of the whole PY element and  $c_E = c_{vol}$  volumetric heat capacity of each constituent.

Equations 3.12 and 3.13 leads to optimal  $\eta$  (Efficiency) expression for simple resistive load PY energy harvesting case, namely  $\eta_{Res}$ .

$$\begin{aligned}
 \Rightarrow \quad \eta_{Res} &= \frac{W}{Q_h} = \frac{\frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)^2}{16\varepsilon_{mn}^T (\frac{1}{R} + 1)} \times Vol}{\frac{R^{PY}c_E + {}^{NP}c_E}{R+1} \times (\Theta_H - \Theta_L) \times Vol} \\
 &= \frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)}{16\varepsilon_{mn}^T (\frac{R+1}{R}) \left( \frac{R^{PY}c_E + {}^{NP}c_E}{R+1} \right)} \\
 &= \frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)}{16\varepsilon_{mn}^T \left( {}^{PY}c_E + \frac{{}^{NP}c_E}{R} \right)}
 \end{aligned} \tag{3.14}$$

It is apparent from 3.14 that where  $\eta_{Res}$  is concerned there is a trade off between the increased PY coef and denominator of  $\eta_{Res}$  expression as  $R$  gets smaller. Hence for every 2-2 connectivity laminate composite, there will be an optimal  $R$  which maximises the efficiency ( $\eta_{Res}$ ). In addition it also suggests that enhancing PY coef is a very effective route for improving PY energy harvesting efficiency since  $\eta_{Res}$  is proportional to  $(p_m^{T,E})^2$ .

In the literature[2] “Electrothermal coupling factor ( $k^2$ )” is defined as:

$$\begin{aligned}
 k^2 &= \text{Electrothermal coupling factor at } \Theta_H \\
 &= \frac{(p_m^{T,E})^2 \Theta_H}{\varepsilon_{mn}^T \bar{c}_E} = \frac{(p_m^{T,E})^2 \Theta_H (R+1)}{\varepsilon_{mn}^T (R^{PY}c_E + {}^{NP}c_E)}
 \end{aligned} \tag{3.15}$$

Using 3.15 and noting the Carnot efficiency is defined as [13, 2];  $\eta_{Carnot} = 1 - \frac{\Theta_L}{\Theta_H} = \frac{\Theta_H - \Theta_L}{\Theta_H}$  with temperatures measured in absolute temperature scale, one can also make comparison between  $\eta_{Carnot}$  and  $\eta_{Res}$  (3.14), ideally optimised energy harvesting cycle (Carnot cycle)'s efficiency and that of simple resistive load case respectively.

$$\begin{aligned}
 \Rightarrow \eta_{Res} &= \frac{\pi (p_m^{T,E})^2 (\Theta_H - \Theta_L)}{16 \varepsilon_{mn}^T \left( R^{PY} c_E + \frac{N^P c_E}{R} \right)} \\
 &= \frac{\pi}{16} \times \frac{(p_m^{T,E})^2 \Theta_H (R+1)}{\varepsilon_{mn}^T (R^{PY} c_E + N^P c_E)} \times \frac{\Theta_H - \Theta_L}{\Theta_H} \\
 &\quad \times \frac{(R^{PY} c_E + N^P c_E)}{R+1} \times \frac{1}{R^{PY} c_E + \frac{N^P c_E}{R}} \\
 &= \frac{\pi}{16} \times k^2 \eta_{Carnot} \times \frac{(R^{PY} c_E + N^P c_E) R}{(R+1) (R^{PY} c_E + N^P c_E)} \\
 &= \frac{\pi}{16} \left[ \frac{R}{R+1} \right] k^2 \eta_{Carnot}
 \end{aligned} \tag{3.16}$$

Comparison in 3.16 enables comparison between various PY energy harvesting systems operating in the same environment, namely defined available temperature gradient or hot/cold reservoirs and evaluated maximum possible energy conversion efficiency (Carnot efficiency).

It should also be noted that all expressions, 3.11  $\sim$  3.16 converges to that of non-composite PY material only case presented by Sebald *et al* as  $R \rightarrow \infty$ .

#### 4. Results and discussion

A few of the most widely used PY materials [5], such as PZT (PZT-5H and PZT-5A), BTO, LTO, LNO, and PVDF, were paired with six different NP materials with wide ranging thermal and elastic properties [5], namely, St, PTFE, CPVC, Al, Zn, and Invar36, to analyze the resulting laminate composite's energy harvesting credentials. Although all the 36 pairs were examined, in this communication the conclusions of only selected few with the most interesting results are presented.

It must also be noted that in figure 1, although  $\Theta_H$  and  $\Theta_L$  are shown as the temperatures of hot and cold reservoirs/heat sources/drains respectively, as long as the resultant force from steel and SMA springs changes direction at these temperatures owing to SMA springs temperature dependent spring constant (Elastic stiffness), the actual temperatures of the hot and cold reservoirs ( $\Theta_{hot}$  and  $\Theta_{cold}$ ) can be anything as long as they satisfy  $\Theta_{hot} > \Theta_H$  and  $\Theta_{cold} < \Theta_L$ . This facilitates the possibility of having potentially huge spatial temperature gradient when PY element is in contact with the surfaces of hot and cold reservoirs, which can improve the thermal conductivity even further leading to higher available frequency ( $f$ ), and hence ultimately greater Maximum power density ( $P_{denMax}$ ). The use of SMA, however, means the thermal (heating/cooling) cycle frequency ( $f$ ) achievable from such arrangements would still be rather limited. This is one of the main reasons why the frequency range considered in section 4 is constrained to those below or equal to 1 Hz, dampening the potential energy output available. One way of overcoming this

may be the use of ferromagnetic SMA[14], which can produce actuation frequency ranges of 200  $Hz$  or more, although the use of magnetic stimuli means more design complications.

#### 4.1. Pyroelectric energy harvesting potentials of 2-2 connectivity laminate composites

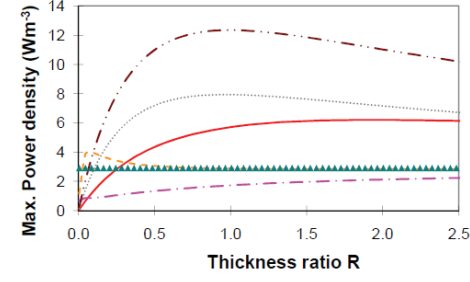
Although PZT-5A's energy harvesting credentials were also considered, the trend was very similar to that of PZT-5H with smaller numerical values. Hence only the outcomes of the latter will be presented as an example of perovskite materials' energy harvesting potential. BTO did not demonstrate interesting enough results, while LNO behaved very similarly to LTO with only smaller numerical values. Hence only LTO's results are presented.

*4.1.1. PZT-5H pairs* From figures 2(a) and 2(b), it is evident that despite PTFE and CPVC introducing extreme PY coef enhancements, owing to their high thermal expansion coefs, and possessing very low volumetric heat capacities, it is in fact Zn, Al, and St with their reasonably high thermal expansion coefs and middle-range volumetric heat capacities that demonstrate the most promise in PY energy harvesting (cf. table A3). This is traced back to their high Young's moduli, which provides for sufficient enhancement even at relatively high  $R$  values, i.e. even when only small mass of NP material attached. This is a very good example of a case where pure enhancement in PY coef alone is not enough for good performance in a particular application of PY effect. Energy harvesting characteristics of PZT5H-Zn, PZT5H-Al, and PZT5H-St pairs indicate their great potential in PY energy harvesting applications. In particular, noting that all these values are evaluated for the simplest resistive cycle case means should much better energy harvesting circuits and storage technologies be used, there indeed is great potential in these 2-2 connectivity laminate composites to be exploited in PY energy harvesting application based on both linear and non-linear pyroelectricity.

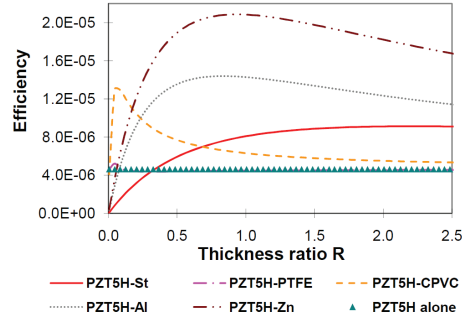
*4.1.2. LTO pairs* LNO and LTO's small PY coef enhancement[5] meant that their 2-2 connectivity laminate composites actually reduced their PY energy harvesting abilities as illustrated by Figures 3(a) and 3(b). However, LTO and LNO single crystals by themselves are expected to show the highest  $P_{denMax}$  of all the materials and composites considered in this dissertation, implying to its potential employment in energy harvesting applications. The reason for this is their relatively high PY coefs coupled with low dielectric constants resulting in high voltage response.

*4.1.3. PVDF pairs* Figures 4(a) and 4(b) insinuates that although PVDF pairs also fared rather badly when it comes to PY energy harvesting abilities, at higher  $R$  values some actually showed slight improvement, PVDF-Invar36 and PVDF-St in particular. This suggests that where PY energy harvesting application is concerned, thin coating of Invar36 or St on PVDF (even to act as electrodes) can improve electrical energy output ( $P_{denMax}$  and  $\eta_{Res}$ ) of PVDF PY element.

*4.1.4. Comparison with other pyroelectric materials* In order to make comparison between PY materials and laminate composites considered in this work and various PY



(a) Maximum power density ( $Wm^{-3}$  or  $\mu Wcm^{-3}$ ) vs R



(b) Efficiency ( $\eta_{Res}$ ) vs R

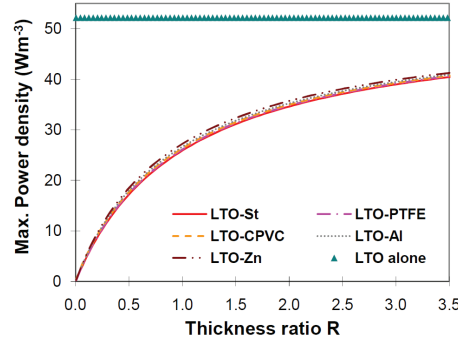
**Figure 2.** Pyroelectric energy harvesting potentials of PZT5H pairs with ideal interfacial bonding layer,  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$  assumed.

elements assessed by Sebald *et al* [2], table 1 was created with materials such as PMN-PT (Lead magnesium niobate-Lead titanate single crystals), PLZT (Lead-lanthanum-zirconate-titanate), and PVDF-HFP (Poly(vinylidene fluoride-hexafluoropropylene) copolymer).

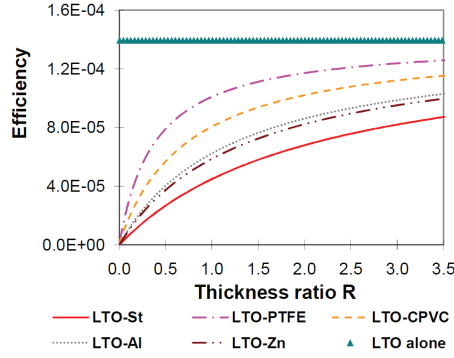
Note that our PZTs have much lower  $k^2$  since the literature used much smaller  $\varepsilon_{33}^T$  and  $c_E = c_{vol}$  values than ours. In case  $k^2$  was not fully representative of potential  $P_{denMax}$  particular PY element might be capable of,  $k_{Lam}^2 = \{R/(R+1)\} k^2$  from 3.16 were also evaluated as illustrated by table 1. Equation 3.15 can be used to derive an expression for  $k_{Lam}^2$ .

$$\begin{aligned}
 k_{Lam}^2 &= \frac{R}{R+1} k^2 = \left( \frac{R}{R+1} \right) \frac{(p_m^{T,E})^2 \Theta_H (R+1)}{\varepsilon_{mn}^T (R^{PY} c_E + {}^{NP} c_E)} \\
 &= \frac{(p_m^{T,E})^2 \Theta_H R}{\varepsilon_{mn}^T (R^{PY} c_E + {}^{NP} c_E)} = \frac{(p_m^{T,E})^2 \Theta_H}{\varepsilon_{mn}^T \left( {}^{PY} c_E + \frac{{}^{NP} c_E}{R} \right)}
 \end{aligned} \tag{4.1}$$

This new electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) should be used when assessing PY energy harvesting potentials of 2-2 connectivity laminate composites, which suggests that when considering various composites for their



(a) Maximum power density ( $Wm^{-3}$  or  $\mu Wcm^{-3}$ ) vs  $R$



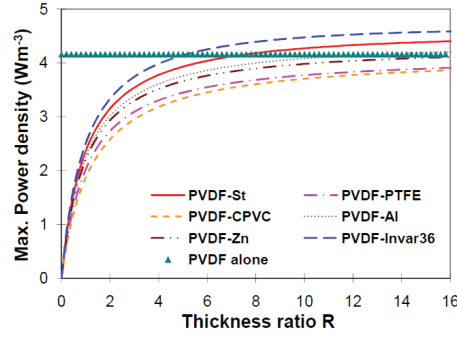
(b) Efficiency ( $\eta_{Res}$ ) vs  $R$

**Figure 3.** Pyroelectric energy harvesting potentials of LTO pairs with ideal interfacial bonding layer,  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$  assumed.

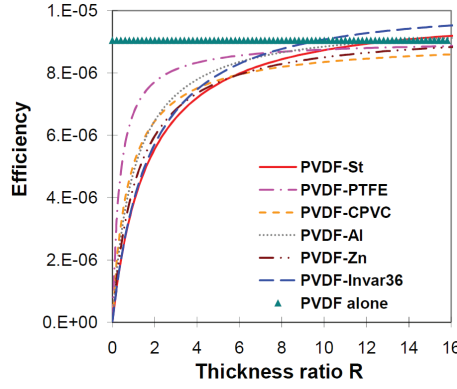
energy harvesting credentials, as suggested by Sebald *et al* [2], their particular connectivity symmetry must be taken into account before carrying out comparisons with their material counterparts. Figure 5 illustrates this point very well by closely approximating the trends depicted in figure 2(a).

Good  $k_{Lam}^2$  values predicted for PZT5H-Zn, PZT5H-Al, and to a less extent PZT5H-St in figure 5 owes largely to NP materials' ability to generate significantly large enough PY coef enhancement even at quite high  $R$  values while having relatively low  $c_{vol}$ , leading to less additional thermal mass. PTFE and CPVC pairs do have a peak at low  $R$  due to their extreme PY coef enhancements at such  $R$  values and their extremely low  $c_{vol}$ . (cf. table A3)

It is also evident from table 1 that LTO single crystal is a very promising PY material for energy harvesting application. Although PMN-PT single crystal from the literature exhibits the highest  $k^2$  it is expensive and fragile, while PZT/PVDF-HFP composite, another PY element from the literature with great promise, has  $k^2$  evaluated from electrothermal coupling factor expression not yet adapted for composites. With that in mind, LTO single crystal, PZT5H-Zn ( $R = 1.005$ ), and PZT5H-CPVC ( $R = 0.045$ ) 2-2 connectivity laminate composites show extreme promise in PY energy harvesting application. In particular, PZT5H-Zn ( $R = 1.005$ ) composite's  $P_{denMax}$  of  $12.35 Wm^{-3}$  at  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$ ,



(a) Maximum power density ( $Wm^{-3}$  or  $\mu Wcm^{-3}$ ) vs  $R$



(b) Efficiency ( $\eta_{Res}$ ) vs  $R$

**Figure 4.** Pyroelectric energy harvesting potentials of PVDF pairs with ideal interfacial bonding layer,  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$  assumed.

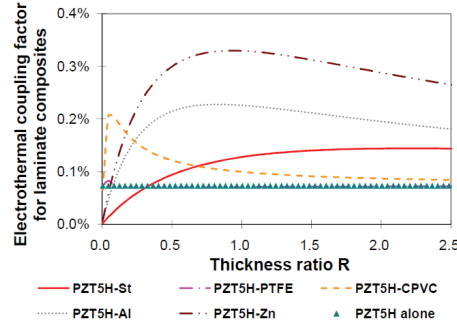


**Figure 5.** Electrothermal coupling factor for laminate composites ( $k_{Lam}^2 = \frac{R}{R+1}k^2$ ) for PZT5H pairs with ideal interfacial bonding layer,  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$  assumed.

is very respectable, which can easily be further improved by increasing  $\Theta_H - \Theta_L$  and  $f$ , when compared to that of a typical thermoelectric module, i.e.  $30 Wm^{-3}$ [2].

Considering the important role the frequency plays in determining the maximum

**Table 1.** Comparison between enhanced 2-2 connectivity laminate composites and pyroelectric elements considered by Sebal *et al* [2] for energy harvesting application.

| PY element                                    | p      | $\varepsilon_{33}^T$ | $P_{denMax}$ | $\eta_{Res}$ | $k^2$ | $k_{Lam}^2 = \frac{R}{R+1}k^2$ |
|-----------------------------------------------|--------|----------------------|--------------|--------------|-------|--------------------------------|
| Selected PY elements from the literature[2]   |        |                      |              |              |       |                                |
| 111 PMN-0,25PT                                | -17.90 | 961                  | N/A          | N/A          | 4.79  | N/A                            |
| PZT                                           | -5.33  | 1116                 | N/A          | N/A          | 0.37  | N/A                            |
| PLZT 0.5/53/47                                | -3.60  | 854                  | N/A          | N/A          | 0.22  | N/A                            |
| PVDF                                          | -0.33  | 9                    | N/A          | N/A          | 0.14  | N/A                            |
| PZT/PVDF-HFP 50/50                            | -4.50  | 85                   | N/A          | N/A          | 4.28  | N/A                            |
| PY materials considered in this work          |        |                      |              |              |       |                                |
| PZT5H                                         | -5.00  | 2874                 | 2.92         | 4.63         | 0.073 | N/A                            |
| PZT5A                                         | -3.00  | 1803                 | 2.22         | 3.52         | 0.056 | N/A                            |
| BTO                                           | -2.00  | 168                  | 1.04         | 1.64         | 0.026 | N/A                            |
| LTO                                           | -2.30  | 45                   | 52.14        | 139.41       | 2.20  | N/A                            |
| LNO                                           | -0.83  | 30                   | 10.18        | 17.44        | 0.28  | N/A                            |
| PVDF                                          | -0.274 | 7.75                 | 4.16         | 9.05         | 0.14  | N/A                            |
| Selected 2-2 connectivity laminate composites |        |                      |              |              |       |                                |
| PZT5H-CPVC ( $R = 0.005$ )                    | -44.68 | 2874                 | 1.16         | 4.11         | 13.06 | 0.065                          |
| PZT5H-Zn ( $R = 1.005$ )                      | -14.53 | 2874                 | 12.35        | 20.86        | 0.66  | 0.33                           |
| PZT5H-CPVC ( $R = 0.045$ )                    | -27.68 | 2874                 | 3.85         | 13.05        | 4.78  | 0.21                           |
| PZT5H-PTFE ( $R = 0.005$ )                    | -34.16 | 2874                 | 0.68         | 4.63         | 14.69 | 0.073                          |

Units: - p (PY coef);  $\times 10^{-4} Cm^{-2}K^{-1}$  -  $\varepsilon_{33}^T$ ; No unit -  $\eta_{Res}$ ;  $\times 10^{-6}$  (No unit)  
 -  $P_{denMax}$ ;  $Wm^{-3}$  or  $\mu Wcm^{-3}$  (evaluated at  $f = 0.01Hz$ ) -  $k^2$ ; %

power density (cf. 3.11), it must be noted that the choice of  $0.01Hz$  frequency was not entirely arbitrary. In Appendix A.1, “Time constant” expression in A.3 was used for the largest sample experimentally investigated (a  $267 \mu m$  PZT5H and two  $250 \mu m$  St), with the maximum time constant being evaluated to be approximately  $1.87 \times 10^{-1}s$ , giving maximum thermal variational frequency of  $f_{max} = 1/(\text{Maximum time constant}) \approx 5.35Hz$  for  $1K$  temperature variation. However, for  $\Theta_H = 310K$  and  $\Theta_L = 300K$  considered in our work, total temperature change of PY element during one full thermal variation cycle in figure 1 is  $\Delta\Theta = 2 \times 2\Theta_0 = 2 \times (\Theta_H - \Theta_L) = 20K$ . In addition, the movement of PY element from one surface to another is not instantaneous. The time taken for this translation from figures 1(a) to 1(d) would depend on the exact configurations of the SMA and Steel springs.

Hence for a given thermal cycle, the total time it takes for the translation to take place would be  $\tau_{tran} = 1 \times 2 = 2s$ . For  $\Delta\Theta = 20K$  temperature variation, the time used for changing  $\Theta_{PY}$  would be  $\tau_{PY} = 0.187 \times 20 = 3.74s$ , assuming that SMA and Steel springs’ temperature changing times are a lot less than PY element’s due to their volume being much smaller. Hence a single thermal cycle process should take

a maximum of  $\tau_{total} = \tau_{trans} + \tau_{PY} = 5.74s$ . This means the maximum thermal variational frequency a 2-2 connectivity laminate composite of PZT5H-St can achieve is at least  $f_{Max} = 1/(\tau_{total}) = 0.175Hz$ , which is much larger than  $0.01Hz$  used for our analysis. The fact that Xie *et al* [3] uses heating rate of  $15^\circ Cs^{-1}$ , effectively  $0.75Hz$  frequency for  $\Delta\Theta = 20K$  cycles, on a  $150\mu m$  thick PZT-5A demonstrates that this should be achievable, implying  $0.01Hz$  is a conservative and reasonable choice.

#### 4.2. Pyroelectric energy harvesting potentials of measured laminate samples

We now move on to the results of similar analyses performed on the experimental samples whose PY coefs were measured to be enhanced. Should the same samples be measured for their power output with impedance matching circuitry, following outcomes in tables 2 ~ 4 are expected.

The frequencies investigated in these tables are;  $f = 0.0017Hz$  representing  $2^\circ Cmin^{-1}$  heating rate (used in the experimentation for testing the samples) applied to a  $\Delta\Theta = 20K$  thermal cycle, and  $f = 0.07$  for being the frequency at which some of the tested experimental samples start to achieve maximum power densities ( $P_{denMax}$ ) larger than that of a typical thermoelectric module, i.e.  $30 Wm^{-3}$ . Where the efficiencies are concerned,  $\eta_{Carnot} = 0.0323$  for  $\Theta_H = 310$ , while  $\eta_{Carnot} = 0.0625$  for  $\Theta_H = 320$ .

Table 2 illustrates energy harvesting parameters bonded 2-2 connectivity laminate composites of PZT5H-St can produce. Even after taking the differences in frequencies investigated into account, the composites in table 2 that possess  $R > 1$  outperform the typical PZT5H's parameters presented in table 1. Although smaller  $R$  leads to higher PY coef enhancement, it also means larger additional thermal mass from NP layer, resulting in larger  $\bar{c}_E$  which in turn has a negative effect on all the energy harvesting parameters,  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$ . As expected, this suggests maximising the PY coef alone is not enough to optimise the energy harvesting system and other issues such as the additional thermal mass needs to be considered.

Where the electrothermal coupling factor for laminate composite ( $k_{Lam}^2$ ) is concerned Sample XIR7 with  $R = 1.068$  has the highest value, leading to highest maximum power densities of  $32.2$  and  $129.0 Wm^{-3}$  at  $f = 0.07Hz$  for  $\Theta_H = 310$  and  $\Theta_H = 320$  respectively. All the samples with  $R > 1$  depict a rather large  $P_{denMax}$ , larger than  $100 Wm^{-3}$  at  $f = 0.07Hz$  for  $\Theta_H = 320$ , insinuating their potential deployment in energy harvesting application. Bearing in mind that the frequency and temperature variations used for these calculations are all viewed as reasonably conservative values, there is a good chance that these composites, or similar composites with different PY or NP materials such as Zn, Al, or CPVC (cf. figure 5) could well find their use in PY energy harvesting.

Figures 6, 7, and 8 summarizes the findings from table 2. Interfacial factor  $k_{i1}$  (a ratio of strain loss across the PY and NP layers' interface)[4, 5] is employed to describe the effect of imperfect bonding layer. It is clear from these figures that as long as the thickness ratio ( $R$ ) is larger than certain value and the bonding quality is reasonably good, 2-2 connectivity laminate composites of PZT-5H will outperform stand alone PZT-5H in the energy harvesting application.

In figure 6(a), a theoretical  $P_{denMax}$  should  $f = 1Hz$  be possible is displayed. With PY coef enhancement of over 100% predicted, theoretically over  $500 Wm^{-3}$  or  $0.5 mWcm^{-3}$  is shown to be possible. Considering the fact that phase transition independent PY effect (linear PY effect)'s energy harvesting credentials

**Table 2.** Energy harvesting potentials of the enhanced experimental samples from our other publications[4, 5].

| Name   | $R$   | $p$   | $\Theta_H$ | $P_{denMax}^{0.07}$ | $P_{denMax}^{0.0017}$ | $\eta_{Res}$ | $k_{Lam}^2$ |
|--------|-------|-------|------------|---------------------|-----------------------|--------------|-------------|
| XI3    | 0.254 | -9.18 | 310        | 13.9                | 0.33                  | 2.65         | 0.042       |
|        |       |       | 320        | 55.7                | 1.33                  | 5.30         | 0.043       |
| XII4   |       | -9.02 | 310        | 13.5                | 0.32                  | 2.56         | 0.040       |
|        |       |       | 320        | 53.8                | 1.28                  | 5.12         | 0.042       |
| XI5    | 0.382 | -9.22 | 310        | 19.2                | 0.46                  | 3.71         | 0.059       |
|        |       |       | 320        | 76.8                | 1.83                  | 7.41         | 0.060       |
| XIR5   |       | -7.37 | 310        | 12.3                | 0.29                  | 2.37         | 0.037       |
|        |       |       | 320        | 49.1                | 1.17                  | 4.74         | 0.039       |
| XI8=X3 | 0.534 | -7.95 | 310        | 18.0                | 0.43                  | 3.52         | 0.056       |
|        |       |       | 320        | 71.8                | 1.71                  | 7.04         | 0.057       |
| XI7    | 1.068 | -8.56 | 310        | 30.9                | 0.74                  | 6.28         | 0.099       |
|        |       |       | 320        | 123.7               | 2.94                  | 12.56        | 0.102       |
| XIR7C  |       | -8.68 | 310        | 31.8                | 0.76                  | 6.46         | 0.102       |
|        |       |       | 320        | 127.2               | 3.03                  | 12.91        | 0.105       |
| XIR7   |       | -8.74 | 310        | 32.2                | 0.77                  | 6.55         | 0.103       |
|        |       |       | 320        | 129.0               | 3.07                  | 13.09        | 0.107       |
| XI1    | 1.270 | -7.96 | 310        | 29.0                | 0.69                  | 5.94         | 0.094       |
|        |       |       | 320        | 116.0               | 2.76                  | 11.88        | 0.097       |
| XIR1   |       | -7.55 | 310        | 26.1                | 0.62                  | 5.34         | 0.084       |
|        |       |       | 320        | 104.2               | 2.48                  | 10.68        | 0.087       |
| XI4    | 1.910 | -7.49 | 310        | 30.1                | 0.72                  | 6.31         | 0.100       |
|        |       |       | 320        | 120.5               | 2.87                  | 12.62        | 0.103       |
| XIR4   |       | -7.14 | 310        | 27.3                | 0.65                  | 5.72         | 0.090       |
|        |       |       | 320        | 109.4               | 2.60                  | 11.45        | 0.093       |
| XIR4C  |       | -7.17 | 310        | 27.5                | 0.66                  | 5.77         | 0.091       |
|        |       |       | 320        | 110.2               | 2.62                  | 11.54        | 0.094       |
| XI6=X2 | 2.670 | -6.72 | 310        | 26.8                | 0.64                  | 5.71         | 0.090       |
|        |       |       | 320        | 107.4               | 2.56                  | 11.43        | 0.093       |

Units: -  $R$  ; No unit -  $p$  = PY coef ;  $\times 10^{-4} \text{ } Cm^{-2}K^{-1}$   
-  $\Theta_H$  ;  $K$  ( $\Theta_L = 300K$ )  
-  $P_{denMax}^{0.07}$  = Maximum power density at  $f = 0.07$  ;  $Wm^{-3}$  (or  $\mu Wcm^{-3}$ )  
-  $P_{denMax}^{0.0017}$  = Maximum power density at  $f = 0.0017$  ( $2^\circ Cmin^{-1}$ ) ;  $Wm^{-3}$   
-  $\eta_{Res}$  = Efficiency in resistive cycle case ;  $\times 10^{-6}$  (No unit)  
-  $k_{Lam}^2$  = Electrothermal coupling factor for laminate composites ; %

were previously assumed to be rather limited, this is certainly a respectable value. In particular, with PZT5H-St pair not being the best performing 2-2 connectivity laminate composite pair in PY coef enhancement, there is a good chance that even this value can be exceeded by substituting the NP or PY materials.

Figures 6(b) ~ 6(d) demonstrate more realistic cases where the frequency and temperature variations are well within the value the previously described PY energy harvesting system in Figure 1 can deliver.  $P_{denMax}$  of over  $160 \text{ } mWcm^{-3}$  is predicted to be possible at  $f = 0.07Hz$  and  $\Theta_H = 320K$  with the best performing experimental sample exhibiting about  $130 \text{ } Wm^{-3}$  under the same condition. This again is a considerable amount of power. For example, a  $1 \text{ } cm^3$  PY element of this composite should be able to provide the maximum of  $130 \text{ } \mu W$  of power, which is enough to power

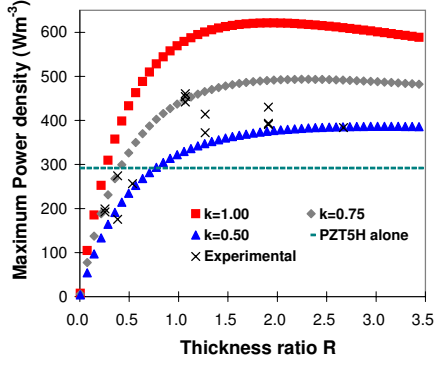
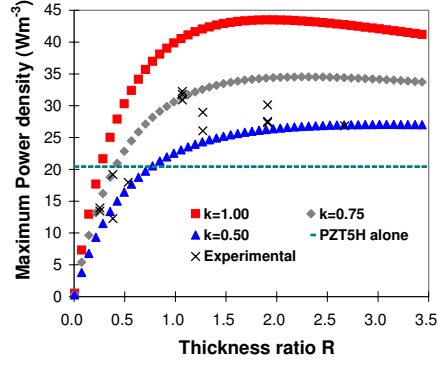
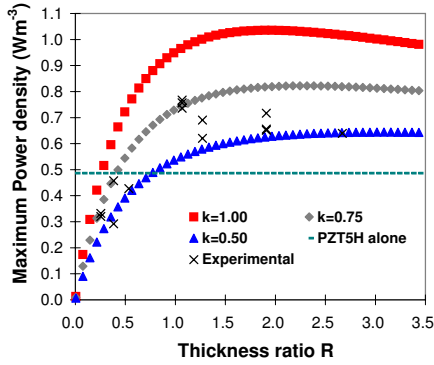
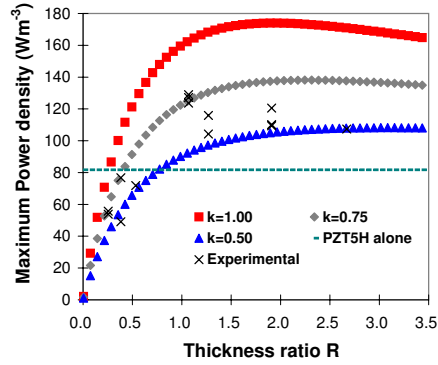

 (a)  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 1Hz$ 

 (b)  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.07Hz$  for around  $30 \mu W cm^{-3}$  or  $W m^{-3}$ 

 (c)  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.0017Hz$  (Experimental frequency)

 (d)  $\Theta_L = 300K$ ,  $\Theta_H = 320K$ , and  $f = 0.07Hz$  (Increasing temperature variation amplitude)

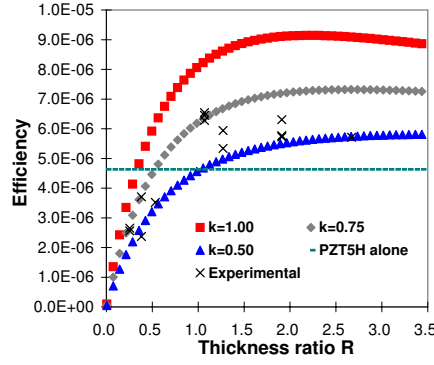
**Figure 6.** Maximum power density for PZT5H-St pairs and the samples from the experiment in our previous publications[4, 5] (with  $k=1$  interfacial factor).

a Radio Frequency IDentification (RFID) tag or a hearing aid[2].

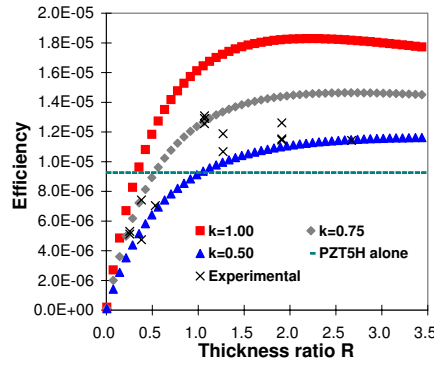
Figure 7 predicts an improvement in the efficiency at around  $R > 1$  or  $R > 1.5$ . Maximum of near doubling of the efficiency ( $\eta_{Res}$ ) are predicted via PY coef enhancement, although experimental samples were only able to demonstrate up to around 40% improvement. It should be noted that  $\eta_{Res}$  is independent of frequency, as the expression in 3.14 suggests.

Electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) and its relationship with  $R$  is depicted in figure 8. This measure for PY energy harvesting credentials was developed to assess a PY material or composite's potential in PY energy harvesting application. As such, it is independent of frequency and temperature variation amplitude (cf. 4.1). It is evident from figure 8 that it predicts PZT5H-St laminate composite's performance in both  $P_{denMax}$  and  $\eta_{Res}$  quite well, with up to doubling of  $k_{Lam}^2$  under optimal conditions.

Changes in energy harvesting parameters as a PZT-5H is evaluated, then as it is bonded to St forming a laminate composite (cf. table 2), are displayed in Table



(a)  $\Theta_L = 300K$ ,  $\Theta_H = 310K$ ,  $f = 0.07Hz$ , and  $\eta_{Carnot} = 0.0323$



(b)  $\Theta_L = 300K$ ,  $\Theta_H = 320K$ ,  $f = 0.07Hz$ , and  $\eta_{Carnot} = 0.0625$  (Increasing temperature variation amplitude)

**Figure 7.** Efficiency ( $\eta_{Res}$ ) for PZT5H-St pairs and the samples from the experiment in our previous publications[4, 5] (with  $k_{-1}$  interfacial factor).

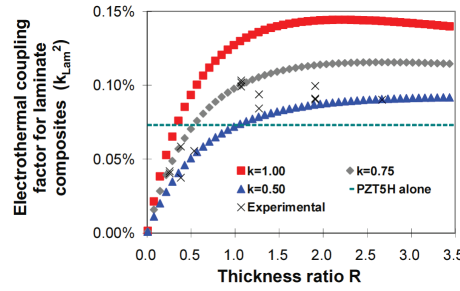


**Figure 8.** Electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) evaluated for PZT5H-St pairs and the samples from the experiment in our previous publications[4, 5] (with  $k_{-1}$  interfacial factor).

3. This compares the parameters of a stand alone PY material with its laminate composite counterpart, which possesses larger volume owing to the introduction of

NP layer. Samples up to  $R < 0.534$  show reduction in their parameters despite large Percentile  $\Delta p$  of up to 88.3% owing to increased thermal mass from the introduction of NP layer. XI6 ( $R = 2.670$ ) showed the best improvement in all the parameters despite its relatively low Percentile  $\Delta p$  of 63.8%. In fact, near doubling of  $P_{denMax}$  (95.1% increase), and both  $\eta_{Res}$  and  $k_{Lam}^2$  (83.1% increase) indicates that attaching a thin layer of St may be the best route for improving PY energy harvesting with PZT-5H. One of the main reasons behind this is St's large  $c_{vol}$  ( $3.91 \times 10^6 \text{ Jm}^{-3}\text{K}^{-1}$ ), which is even larger than that of PZT-5H ( $3.15 \times 10^6 \text{ Jm}^{-3}\text{K}^{-1}$ ). Use of other NP materials such as Al ( $c_{vol} = 2.40 \times 10^6 \text{ Jm}^{-3}\text{K}^{-1}$ ), or even PTFE and CPVC with  $0.72$  and  $1.40 \times 10^6 \text{ Jm}^{-3}\text{K}^{-1}$  respectively, should aid in reducing this hinderance to improvement. Table 3 formed the basis for figure 9.

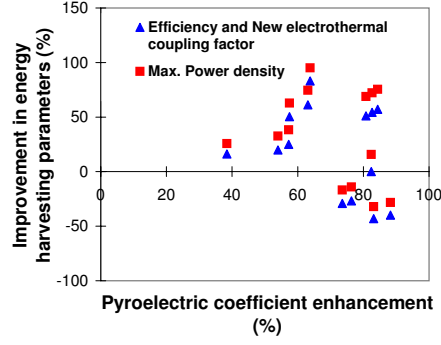
**Table 3.** Experimental samples before (without NP layer) and after (with NP) bonding.

| Name   | $R$   | Perc $\Delta p$ | Perc $\Delta P_{denMax}$ | Perc $\Delta \eta_{Res}$ | Perc $\Delta k_{Lam}^2$ |
|--------|-------|-----------------|--------------------------|--------------------------|-------------------------|
| XI3    | 0.254 | 83.1            | -32.1                    | -43.0                    |                         |
| XII4   | 0.254 | 88.3            | -28.2                    | -39.8                    |                         |
| XI5    | 0.382 | 76.3            | -14.0                    | -26.8                    |                         |
| XIR5   | 0.382 | 73.6            | -16.7                    | -29.1                    |                         |
| XI8=X3 | 0.534 | 82.4            | 15.8                     | 0.08                     |                         |
| XI7    | 1.068 | 84.4            | 75.5                     | 57.2                     |                         |
| XIR7C  | 1.068 | 80.8            | 68.8                     | 51.2                     |                         |
| XIR7   | 1.068 | 82.6            | 72.3                     | 54.3                     |                         |
| XI1    | 1.270 | 54.0            | 32.6                     | 19.9                     |                         |
| XIR1   | 1.270 | 57.3            | 38.4                     | 25.1                     |                         |
| XI4    | 1.910 | 38.4            | 25.8                     | 16.2                     |                         |
| XIR4   | 1.910 | 57.5            | 62.8                     | 50.4                     |                         |
| XIR4C  | 1.910 | 63.1            | 74.6                     | 61.2                     |                         |
| XI6=X2 | 2.670 | 63.8            | 95.1                     | 83.1                     |                         |

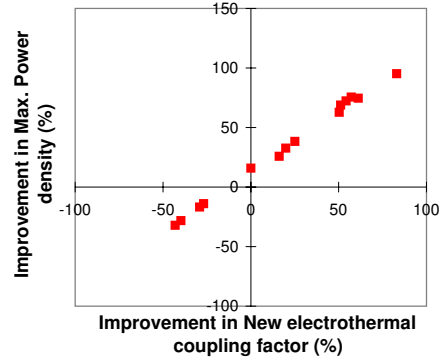
Units: -  $R = \frac{PY_t}{NP_t}$  = thickness ratio ; No unit

- Perc = Percentile
- Perc  $\Delta p$  = Percentile PY coef enhancement after bonding ; %
- Perc  $\Delta P_{denMax}$  = Percentile maximum power density ( $P_{denMax}$ ) change after enhancement ; %
- Perc  $\Delta \eta_{Res}$  = Percentile efficiency ( $\eta_{Res}$ ) change after enhancement ; %
- Perc  $\Delta k_{Lam}^2$  = Percentile electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) change after enhancement ; %

Figure 9 describes how the percentile change in energy harvesting application specific parameters ( $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$ ) vary with the percentile PY coef enhancement. By comparing the percentile improvements in these parameters between stand alone PZT-5H and PZT5H-St composite (with additional NP layer bonded, and hence with larger volume), one can investigate the effect of trade-off between the enhanced PY coef and increased thermal mass. Figure 9(a) depicts a somewhat confusing picture where rather unexpectedly high PY coef enhancement leads to a reduction, which can be attributed to increased thermal mass from NP overtaking the improvement from the enhanced PY coef. However, figure 9(b) describes a proportional correlation between Percentile  $\Delta k_{Lam}^2$  and Percentile  $\Delta P_{denMax}$ , demonstrating the suitability of  $k_{Lam}^2$  as potential indicators for PY energy harvesting performance in laminate composites.



(a) Percentile PY coef enhancement vs improvements in  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$  in percentage



(b) Percentile improvement in  $k_{Lam}^2$  vs Percentile improvement in  $P_{denMax}$

**Figure 9.** Percentile improvements in  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$  (from Table 3).

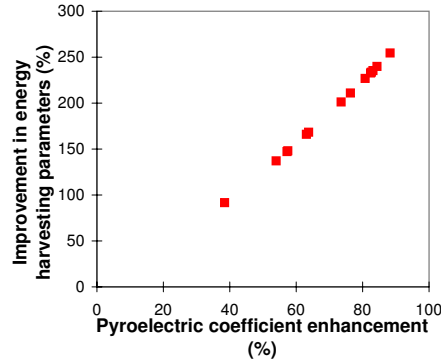


**Figure 10.** Percentile pyroelectric coefficient enhancement versus improvements in  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$  in percentage (from Table 4).

**Table 4.** Experimental samples before and after the bonding (same volume).

| Name   | $R$   | Perc $\Delta p$ | $\Delta P_{denMax}$ , $\Delta \eta_{Res}$ , and $\Delta k_{Lam}^2$ (Perc) |
|--------|-------|-----------------|---------------------------------------------------------------------------|
| XI3    | 0.254 | 83.1            | 235.3                                                                     |
| XII4   | 0.254 | 88.3            | 254.4                                                                     |
| XI5    | 0.382 | 76.3            | 211.0                                                                     |
| XIR5   | 0.382 | 73.6            | 201.2                                                                     |
| XI8=X3 | 0.534 | 82.4            | 232.7                                                                     |
| XI7    | 1.068 | 84.4            | 239.9                                                                     |
| XIR7C  | 1.068 | 80.8            | 226.8                                                                     |
| XIR7   | 1.068 | 82.6            | 233.6                                                                     |
| XI1    | 1.270 | 54.0            | 137.1                                                                     |
| XIR1   | 1.270 | 57.3            | 147.3                                                                     |
| XI4    | 1.910 | 38.4            | 91.7                                                                      |
| XIR4   | 1.910 | 57.5            | 148.1                                                                     |
| XIR4C  | 1.910 | 63.1            | 166.0                                                                     |
| XI6=X2 | 2.670 | 63.8            | 168.2                                                                     |

Units: -  $R = \frac{PY_t}{NP_t}$  = thickness ratio ; No unit

- Perc = Percentile
- Perc  $\Delta p$  = Percentile PY coef enhancement after bonding ; %
- Perc  $\Delta P_{denMax}$  = Percentile maximum power density ( $P_{denMax}$ ) change after enhancement ; %
- Perc  $\Delta \eta_{Res}$  = Percentile efficiency ( $\eta_{Res}$ ) change after enhancement ; %
- Perc  $\Delta k_{Lam}^2$  = Percentile electrothermal coupling factor for laminate composites ( $k_{Lam}^2$ ) change after enhancement ; %

Figure 10 depicts the proportional correlation between Percentile  $\Delta p$  and Percentile  $\Delta P_{denMax}$ ,  $\Delta \eta_{Res}$ , and  $\Delta k_{Lam}^2$ . It represents a case where both PZT-5H and St were assumed present when considering both before and after bonding. Hence this represents the case where the overall volume of initial materials (as PY and NP are not yet bonded, they are not a composite) is equal to the volume of resultant laminate composite after bonding. As there is no additional thermal mass (volume) after the bonding (it was added beforehand), the only quantity that affects the energy harvesting parameters is the enhanced PY coef. This correlation between Percentile  $\Delta p$  and the improvements in all three energy harvesting parameters is also present in table 4, which formed the basis for Figure 10.

As one would expect, in the case of table 4 and figure 10, larger the PY coef enhancement larger the improvement in the energy harvesting parameters. It must be noted that all three parameters demonstrated the same amount of percentile improvement since the only reason Percentile  $\Delta P_{denMax}$  behaved differently from the other two parameters in table 3 was due to its dependence on the overall volume and independence of heat capacities. Sample XI7 ( $R = 1.068$ ) is expected to show up to 240% increase in  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$  from PY coef enhancement of only 84.4%, which suggests that thin PZT-5H with thick St attached could improve the overall energy harvesting performance quite drastically for the same volume of PY material and 2-2 connectivity laminate composite. This indicates that the use of thin-films, or at least the thinnest possible bulk material, in the PY energy harvesting applications will be a good idea despite the scaling behaviour of thin films, as this leads to reduction in the significantly negative role played by the additional thermal mass. In addition, use of other NP materials with lower  $c_{vol}$  values than that of PZT-5H, such as Al, Zn,

PTFE or CPVC, should lead to even larger improvements as this will lead to reduced thermal mass. This reduced thermal mass should also enable the use of higher  $f$  values, potentially resulting in very high  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$ .

Although we only focused our attention on the linear pyroelectricity based energy harvesting application as this employs pyroelectricity under the short circuit condition, under which we have experimental results of the enhancement as stated in our previous communications[4, 5], it should be noted that analysis on the phase transition based non-linear pyroelectric effect should also be possible. However, it requires the use of the pyroelectric coefficients under both short and open circuit electric boundary conditions while its practical realization also necessitates additional electronic circuitry such as synchronised electric charge extraction (SECE)[15] or synchronised switch harvesting on inductor(SSHI)[16]. Hence we have only dealt with simpler case of linear pyroelectric effect with resistive load in this communication although we have no doubt that our enhanced 2-2 connectivity laminate composites can also improve the performance of non-linear pyroelectric energy harvesting application. In addition, various hybrids of this and other methods of energy harvesting also exist, such as the hybrid between magnetostriction and piezoelectricity in Magnetoelectric[17, 18] devices, and it is hoped that our work can also find use in such applications as well.

## 5. Conclusion

Mathematical expressions/parameters that are important in judging the energy harvesting credentials of any PY material or PY-NP 2-2 connectivity laminate composites, namely Maximum power density  $P_{denMax}$ , Efficiency  $\eta_{Res}$ , and Electrothermal coupling factor  $k^2$  have been derived. In doing so, it is discovered that while electrothermal coupling factor ( $k^2$ ) quoted in the literature[2] is adequate for stand alone PY materials, for PY coef enhanced laminate composites its more general counterpart in electrothermal coupling factor specifically designed for laminate composites ( $k_{Lam}^2$  from 4.1) should be used. It seems possible that for other connectivity configurations of composites, various electrothermal coupling factor expressions may be needed. The main application parameters that affect  $P_{denMax}$  are identified as  $\Theta_H - \Theta_L$  and  $f$ , while  $\eta_{Res}$  was independent of  $f$ , and  $k_{Lam}^2$  of both. The independence of  $k_{Lam}^2$  from  $(\Theta_H - \Theta_L)$  and  $f$  made it an ideal parameter for judging the energy harvesting credentials of PY materials or composites, provided  $\Theta_H$  is sufficiently large enough. Comparisons between PY material without and with added NP layers (different total volume), and between PY material with NP (not bonded) and its laminate composite counterpart of the same volume have been made. It was discerned that percentile improvement in  $k_{Lam}^2$  (Percentile  $\Delta k_{Lam}^2$ ) was able to demonstrate a proportional correlation with the percentile improvement in  $P_{denMax}$  (Percentile  $\Delta P_{denMax}$ ) in both cases, which percentile PY coef enhancement (Percentile  $\Delta p$ ) failed to do in the case of differing volumes due to the introduction of additional thermal mass in the form of NP layer leading to dissimilar volume expression before and after the bonding of the laminate composite.

From considering the resistive cycle energy harvesting credentials of the thirty-six PY-NP 2-2 connectivity laminate composites, LTO single crystal was determined to show immense promise as a prime PY material candidate for energy harvesting application. PZT5H-Zn ( $R = 1.005$ ) and PZT5H-CPVC ( $R = 0.045$ ) 2-2 connectivity laminate composites also display potential in PY energy harvesting application. In particular, PZT5H-Zn ( $R = 1.005$ ) composite's  $P_{denMax}$  of  $12.35 \text{ Wm}^{-3}$  at

$\Theta_L = 300K$ ,  $\Theta_H = 310K$ , and  $f = 0.01Hz$ , is very respectable, which can easily be further improved by increasing  $\Theta_H - \Theta_L$  and  $f$ , when compared to that of a typical thermoelectric module, i.e.  $30 Wm^{-3}$ [2]. The possibility of further improvements were noted by facilitating potentially huge spatial temperature gradient when PY element is in contact with the surfaces of hot and cold reservoirs, which can improve the thermal conductivity even further leading to higher available frequency ( $f$ ), and hence ultimately greater  $P_{denMax}$ .

When bonded laminate composites of PZT-5H and St were considered, the composites with  $R > 1$  outperformed the typical PZT5H's energy harvesting parameters. Although smaller  $R$  leads to higher PY coef enhancement, it also means larger additional thermal mass from NP layer, resulting in larger effective volumetric heat capacity ( $\bar{c}_E$ ) which in turn has a negative effect on all the energy harvesting parameters,  $P_{denMax}$ ,  $\eta_{Res}$ , and  $k_{Lam}^2$ . This suggests maximising the PY coef alone is not enough to optimise the energy harvesting system. Other issues such as the additional thermal mass needs to be considered.

## Appendices

### Appendix A.1. Time constant derivation

*Appendix A.1.1. Thermal conductivity* The intensive property of a material that indicates its ability to conduct heat, defined as the quantity of heat  $Q$ , transmitted in time  $t$ , through a thickness  $L$ , in a direction normal to a surface area ( $A$ ), due to a temperature difference  $\Delta\Theta$ , under steady state conditions and when the heat transfer is dependent only on the temperature gradient.

$$\begin{aligned}
 k &= \text{Thermal Conductivity} \\
 &= \text{Heat flow rate} \times \frac{\text{Distance}}{\text{Area} \times \text{Temperature gradient}} \\
 &= \frac{Q}{t} \times \frac{L}{A \times \Delta\Theta}
 \end{aligned} \tag{A.1}$$

*Appendix A.1.2. Specific heat capacity and heat capacity* Amount of heat energy required to achieve temperature difference of  $1 K$  in  $1 m^3$  of a material. Heat capacity  $= c_P \times \rho = \text{Specific heat capacity} \times \text{Density} = [Jm^{-3}K^{-1}]$

### Appendix A.1.3. Thermal diffusivity

$$\begin{aligned}
 \frac{k}{\rho \times c_P} &= \text{Thermal diffusivity} \\
 &= \frac{\text{Thermal conductivity}}{\text{Density} \times \text{Specific heat capacity}}
 \end{aligned} \tag{A.2}$$

*Appendix A.1.4. Time constant ( $T$ )* This quantity will be defined as the minimum time required for the whole sample to change a single degree, or equivalently a single Kelvin. The derivation and mathematical expression of this entity will be displayed

in the following. To derive the "Time constant", from the definition of thermal conductivity (cf. A.1 and diffusivity (cf. A.2):

$$\begin{aligned} T &= \text{Time constant} = \text{Time taken for 1 K change} \\ &= \frac{QL}{A\Delta\Theta} \times \frac{1}{k} \end{aligned}$$

However, from the definition of heat capacity:

$$\begin{aligned} Q &= \text{Heat capacity} \times \text{Volume} \times \text{Temperature difference} \\ &= c_P \rho A L \Delta\Theta \end{aligned}$$

$$\begin{aligned} \therefore T &= \frac{(c_P \rho A t \Delta\Theta) t}{A \Delta\Theta} \times \frac{1}{k} = \frac{c_P \rho t^2}{k} \\ &= t^2 \times \frac{1}{\text{Thermal diffusivity}} \end{aligned} \tag{A.3}$$

*Appendix A.1.5. Evaluation of time constants for our composites* For a laminate structure with three layers (namely layers 1, 2, and 3):  $\text{Max}(T_1, T_2, T_3) < \tau < T_1 + T_2 + T_3$  where  $T_i$  is the time constant for layer i

This means if the time constant of our heating rate is larger than  $T_1 + T_2 + T_3$ , it should be larger than  $\tau$ , and hence PY coef measurement of the sample should not be affected by the heating rate. Typical values used for our sample time constant calculation:

- Thermal diffusivity of Stainless steel =  $4.05 \times 10^{-6} m^2 s^{-1}$
- $^{PT}c_p$  = Specific heat capacity of PZT =  $420 J kg^{-1} K^{-1}$
- $^{PT}\rho$  = Density of PZT =  $7.8 \times 10^3 kg m^{-3}$
- $^{PT}k$  = Thermal conductivity of PZT =  $1.25 Js^{-1} m^{-1} K^{-1}$

For 2 degrees a minute heating rate, the time needed for 1 K temperature change is 30 seconds. Meanwhile, our largest sample has dimensions:

- Layer 1 : Stainless steel of thickness  $^{St}t = 250 \mu m$
- Layer 2 : PZT of thickness  $^{PT}t = 267 \mu m$
- Layer 3 : Stainless steel of thickness  $^{St}t = 250 \mu m$

$$\begin{aligned} T_1 = T_3 &= \frac{^{St}t^2}{\text{Thermal diffusivity of St}} \\ &= \frac{(250 \times 10^{-6})^2}{4.05 \times 10^{-6}} \approx 1.54 \times 10^{-4} s \\ T_2 &= \frac{^{PT}c_p^{PT}\rho^{PT}t^2}{^{PT}k} \\ &= \frac{420 \times 7.8 \times 10^3 \times (267 \times 10^{-6})^2}{1.25} \approx 1.87 \times 10^{-1} s \\ \therefore \text{Time constant for our largest sample} \end{aligned}$$

$$= T_1 + T_2 + T_3 \approx 1.87 \times 10^{-1} s \ll 30s$$

Therefore, the use of heating rates up to 2 degrees a minute should definitely be okay while even higher heating rates should still be possible.

#### Appendix A.2. Material properties

All the data quoted in this section are evaluated at the room temperature unless stated otherwise.

**Table A1.** Pyroelectric and thermal coefficients of various pyroelectric materials.

|             | PZT-5H <sup>a</sup> | PZT-5A <sup>a</sup> | BTO <sup>b</sup> | LTO <sup>c</sup> | LNO <sup>c</sup> | PVDF <sup>d</sup> |
|-------------|---------------------|---------------------|------------------|------------------|------------------|-------------------|
| $p_1^{T,E}$ | 0                   | 0                   | 0                | 0                | 0                | 0                 |
| $p_2^{T,E}$ | 0                   | 0                   | 0                | 0                | 0                | 0                 |
| $p_3^{T,E}$ | -5.0                | -3.0                | -2.0             | -2.3             | -0.83            | -0.274            |
| $c_{vol}$   | 3.15                | 3.15                | 3.19             | 1.87             | 2.92             | 2.3               |

Units: -  $p$ ;  $\times 10^{-4} \text{ Cm}^{-2}\text{K}^{-1}$  -  $c_{vol}$ ;  $\times 10^6 \text{ Jm}^{-3}\text{K}^{-1}$

<sup>a</sup> References [19, 20, 21]

<sup>b</sup> References [22, 23]

<sup>c</sup> Reference [24]

<sup>d</sup> References [25, 26]

**Table A2.** Dielectric constants of various pyroelectric materials.

|                   | PZT-5H <sup>a</sup> | PZT-5A <sup>a</sup> | BTO <sup>b</sup> | LTO <sup>c</sup> | LNO <sup>c</sup> | PVDF <sup>d</sup> |
|-------------------|---------------------|---------------------|------------------|------------------|------------------|-------------------|
| $\epsilon_{11}^T$ | 2438                | 1796                | 2920             | 51               | 84               | 7.35              |
| $\epsilon_{22}^T$ | 2438                | 1796                | 2920             | 51               | 84               | 9.27              |
| $\epsilon_{33}^T$ | 2874                | 1803                | 168              | 45               | 30               | 7.75              |

<sup>a</sup> Reference [21]

<sup>b</sup> References [27, 28]

<sup>c</sup> Reference [29]

<sup>d</sup> Reference [30]

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**Table A3.** Material properties of non-pyroelectric materials.

|           | St <sup>a</sup> | PTFE <sup>b</sup> | CPVC <sup>c</sup> | Al <sup>d</sup> | Zn <sup>e</sup> | Invar36 <sup>f</sup> |
|-----------|-----------------|-------------------|-------------------|-----------------|-----------------|----------------------|
| $\alpha$  | 14.4            | 79.0              | 80.0              | 24.3            | 30.2            | 1.0                  |
| $c_{vol}$ | 3.91            | 0.72              | 1.40              | 2.40            | 2.77            | 5.15                 |
| $Y$       | 193             | 0.5               | 3.15              | 73.1            | 108             | 141                  |

Units: -  $\alpha$ ;  $\times 10^{-6} \text{ mm}^{-1} \text{ K}^{-1}$  -  $c_{vol}$ ;  $\times 10^6 \text{ Jm}^{-3} \text{ K}^{-1}$   
-  $Y$  (Young's modulus);  $\times 10^9 \text{ Nm}^{-2}$  -  $\nu$  (Poisson's ratio); No unit

<sup>a</sup> Reference [31]

<sup>b</sup> Reference [32]

<sup>c</sup> References [33, 34, 35]

<sup>d</sup> References [33, 36, 28]

<sup>e</sup> References [33, 31]

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## **Laminate composites with enhanced pyroelectric effects for energy harvesting**

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