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Electrical Power Estimation of Thermoelectric Cement Composites with Inclusion of Nanostructured Materials

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ABSTRACT: *Measurement of electrical power output for a thermoelectric generator is of great importance for thermoelectric materials research and development. In this work, the calculation of output power based on the thermoelectric properties measurements of cement composites with graphene nanoplatelets and zinc oxide is reported. The maximum output power of 6.0 μW is estimated when the graphene and ZnO contents are 10 wt%, respectively, by cement mass. Approximately 1.5 W power could be achieved by a one square meter graphene nanoplatelets and ZnO added cement composite with a height of 10 mm under a temperature difference of about 50 °C. This paper also discusses the fabrication as well as the thermoelectric properties measurements of cement composite.*

Keywords: Cement composites; Graphene nanoplatelets; Output power; Thermoelectric generator; Zinc oxide

1. INTRODUCTION

Cement is one of the widely used materials for structural purposes. However, it has non-structural applications too. The non-structural functions emerged when other functional elements or additives are introduced into the cement. Some particular examples of non-structural characteristics are sensing, electromagnetic interference shielding, joule heating and energy harvesting [1]. Recently, the thermoelectric cement materials on the basis of the thermoelectric phenomenon are becoming appealing for energy harvesting applications. The system does not have to depend entirely on local electricity, which is beneficial to create grid-free zero-energy buildings (ZEB) [2] as well as smart energy [3]. It is also advantageous for the isolated structure, which is distant from the utility companies. Besides, since the building generates electricity, the storage facility requirement (e.g., battery) can be reduced, thus saving weight and cost. Because of the thermoelectric effect, cement composites can convert the heat generated at the building surfaces and pavements into electrical energy, particularly in urban areas [4]. In addition, solar thermal energy can be converted directly into electricity through the thermoelectric cement composites leaving minimal thermal power to be drained into the urban atmosphere. This process could be useful in mitigating the effect of the worldwide urban heat island (UHI) phenomenon [5]. Sun et al. first investigated the effect of thermoelectric in cement composites, with the addition of short carbon fibers and reported the Seebeck coefficient of +5.5 $\mu V^{\circ}C^{-1}$ [6]. Since then, a lot of researchers [7][8][9] have been working on increasing its thermoelectric effect. Three properties exhibited by a thermoelectric material: electrical conductivity, the Seebeck coefficient and thermal conductivity. A good thermoelectric material should possess a high Seebeck coefficient and electrical conductivity and low thermal conductivity. In order to improve the thermoelectric properties, additives such as expanded graphite [10], carbon nanotube [11][12], and graphene [13] have recently been introduced into cement.

Different types of nanoparticles were combined with cement, for instance ZnO [14][15][16], Fe₂O₃ [15] and MnO₂ [17][18].

Because of their broad range of properties in electronic, chemical, mechanical and thermoelectric fields, metallic oxides are attracting extensive investigation [19]. They also possess a lot of environmentally friendly attributes than traditional thermoelectric Bi₂Te₃ materials [20]. One of the extensively investigated metallic oxides, zinc oxide (ZnO) has a large variety of applications, which include in the thermoelectric sector [21]. They are also abundant in nature. The Seebeck coefficient for ZnO added cement composites were reported to be more than 1000 $\mu V^{\circ}C^{-1}$; however, a very limited electrical conductivity ($1.7 \times 10^{-8} \text{ Scm}^{-1}$) [15] hindered their applications in energy harvesting.

In order to increase the electrical conductivity of thermoelectric cement composites, additives such as graphene nanoplatelets is an excellent applicant. It exhibits high electrical conductivity. Also, its production cost is relatively cheaper than carbon nanofibers and nanotubes [22]. Besides, graphene nanoplatelets have superior thermoelectric properties than carbon nanotubes [23]. Moreover, the addition of graphene is beneficial because phonons mainly drive the heat conduction in graphene. Graphene added cement composite exhibited low thermal conductivity because the phonons scattered strongly in the cement matrix resulting in a high value of thermal resistance [24]. Here, ZnO nanoparticles are employed in increasing the Seebeck coefficient, whereas graphene is selected for the electrical conductivity enhancement. Materials with good thermoelectric properties are applied for energy harvesting.

Thermoelectric energy harvesting is the process of generating electricity, particularly in the micro-scale from external energy sources such as solar thermal energy, waste heat, ambient heat and human body heat. The main inspirations for small scale thermoelectric energy harvesting devices are to add simplicity and comfort in daily life, lower cost and longer lifetimes,

reliability and respect the nature of ecosystems. Moreover, harvesting ambient thermal energies can be a great solution as they can help to reduce the urban heat island (UHI) effect suggesting ecologically friendly and renewable. Small scale energy harvesting thermoelectric generators can be installed in a wristwatch [25], wood stove [26] and biomass cook stove [27]. Therefore, investigations of thermoelectric energy harvesting systems for easy powering of miniature wireless and mobile electronics are very much welcome. Such energy harvesters are also used in self-powered devices and wireless sensor networks [28][29] in smart structures, as they can sustain operation and operate independently without requiring an external power supply.

In a thermoelectric system, a bunch of equations is employed to classify a thermoelectric material. Among them, the figure of merit (ZT) and maximum output power (P_{max}) are crucial parameters and expressed in the following equations.

$$ZT = \frac{S^2 \sigma}{\kappa} T \quad (1)$$

$$P_{max} = \frac{(S \Delta T)^2}{4(L/\sigma A)} = \frac{S^2 (T_h - T_c)^2}{4(L/\sigma A)} \quad (2)$$

Where S, σ , κ , L, A, T_c and T_h are the Seebeck coefficient (μVK^{-1}), electrical conductivity (Scm^{-1}), thermal conductivity ($\text{Wm}^{-1}\text{K}^{-1}$), sample's length (cm), sample's area (cm^2), cold side temperature (K) and hot side temperature (K), respectively.

In this work, the nanostructured ZnO and graphene are included in the cement composites. Based on the measured thermoelectric properties, maximum output power and efficiency are calculated for the thermoelectric cement composites with nanostructured additives. The highest value of P_{max} equals to 6 μW is obtained at 70 °C with both 10 weight percent graphene nanoplatelets and ZnO additions.

2. METHODOLOGY

2.1 Materials

Commercially available graphene nanoplatelets (H-grade) from XG Sciences, USA and Portland cement from Toyo Matelan Company Limited, Japan, are used for this research. Zinc oxide (ZnO) is purchased from Sigma Aldrich. Details of the properties of graphene nanoplatelets (GnP), ZnO and cement are presented in Table 1.

Table 1. Properties of the materials used for this work.

Material	Average particle size (μm)	Molecular weight (g/mol)	Specific density (g/cm^3)	Surface area (m^2/g)
GnP	~ 25	-	2.2	50-80
ZnO	≤ 0.05	81.39	-	> 10.8
Cement	-	-	3.12	0.35

The GnP and ZnO both used 5.0, 10.0 and 15.0 weight (wt) percent by weight of cement in the composites, and then labelled by 5_CC (5% GnP, 5% ZnO), 10_CC (10% GnP, 10% ZnO) and 15_CC (15% GnP, 15% ZnO), respectively. The composites are listed in Table 2. The additive concentration is limited to 15.0 wt percent since the GnP loading of 15.0 wt percent yielded maximum

thermoelectric properties [13]. All concentration ratios were kept strictly during the processes of sample preparation.

Table 2. A list of prepared cement composites included GnP and ZnO.

Composites	Graphene nanoplatelets (GnP)	Zinc oxide (ZnO)	Cement
5_CC	5 wt%	5 wt%	90 wt%
10_CC	10 wt%	10 wt%	80 wt%
15_CC	15 wt%	15 wt%	70 wt%

2.2 Sample Preparation

Appropriate amounts of graphene nanoplatelets (GnP), zinc oxide (ZnO) and cement poured into a zirconia container. The appropriate quantity (30 g) of zirconia balls (diameter = 1 mm) was applied to the jar and later loaded into a Planetary Micro Mill (Pulverisette 7 Premium Line by Fritsch Japan Company Limited). Then all the components were mixed at a speed of 600 rpm. The mixture was running for 12 cycles, where one cycle involves a run time of 60 min and a pause time of 5 min. This technique is a dry dispersive operation, allowing for the homogeneous placement of GnP and ZnO in the cement composite. The added balls were removed from the blended mixture using a vibrating sieve mechanism (Nitto, ANF-30). By these procedures, uniform particle size can be ensured throughout the sample as well as helps to generate reproducible data. The sieve machine was operated for 20 min with a filter aperture size of 106 μm (140 mesh). Regardless of the GnP and ZnO loadings, the water to cement ratio was preserved at 0.1:1. Water is carefully combined with the mixture and then transferred to a die of steel ($\phi 20 \times 30 \text{ mm}^2$). The mix was pressed at 40 MPa for 1 min by a compression system (NT-200H, NPa system) to form bulk samples with a size of approximately $\phi 20 \times 4 \text{ mm}^2$. The specimens are left for curing at room temperature under ambient conditions, followed by heating for 5 hours at 200 °C to avoid the effect of moisture on the experimental results. A Micro Cutter made by Maruto Company Limited has occasionally been used to meet the sample size required for a given measurement. The general procedures for sample preparation are shown in Fig. 1.

2.3 Measurement

In order to measure electrical conductivity and the Seebeck coefficient, the samples were cut from the pellets to the bars ($2 \times 2 \times 10 \text{ mm}^3$) by the micro cutter, as shown in Fig. 1. Composite surfaces were polished by sandpaper and then platinum wires mounted around the two edges of the composite. The Seebeck coefficient and electrical conductivity were then simultaneously measured using a tool called RZ2001i based on four-probe methods provided by Ozawa science. One side was kept at room temperature and the other side was heated automatically from room temperature. The hot side temperature was increased up to 75 °C with a heating rate of 0.01 °C per second. At each increment, the temperature remained constant for 5 minutes in order to achieve voltage stability.

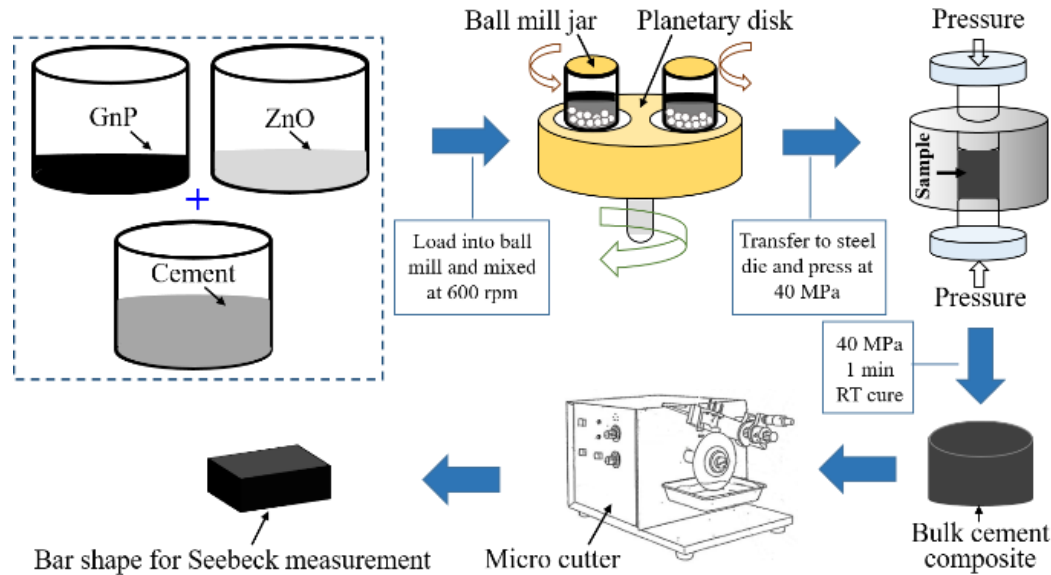


Fig. 1. Processes for the preparation of hybrid cement composites.

The thermal conductivity (κ) was determined from the formulation given as follows: $\kappa = \alpha \rho C_p$. Where α , ρ and C_p are the thermal diffusivity in mm^2s^{-1} , density in gcm^{-3} and specific heat capacity in $\text{Jg}^{-1}\text{K}^{-1}$, respectively. Netzsch LFA 457 Micro Flash [30] conducted thermal diffusivity measurements at ambient conditions with a temperature rise of 10 °C from room temperature to 100 °C. Diffusivity is measured five times at each temperature point to deliver an accurate result. Specific heat capacities were studied by Shimadzu Corporation Limited with a Differential Scanning Calorimeter (DSC-60A) [31]. The density was obtained from the sample mass and geometry.

3. RESULTS AND DISCUSSION

3.1 Thermoelectric Properties

Table 3 presents the thermoelectric properties of the hybrid nanostructured cement composites. It is shown that the composite's electrical conductivity rises as the GnP and ZnO contents are increasing. The maximum electrical conductivity of 5_CC, 10_CC and 15_CC is measured as 5.5, 13.9 and 17.9 Scm^{-1} , respectively. These conductivity values are not much increased than the graphene based cement composites with the same GnP loading, as reported in [13]. Because water is used during the fabrication of GnP-ZnO based cement composite and when ZnO comes into contact with water, it reacts and produced zinc hydroxide, $\text{Zn}(\text{OH})_2$. This $\text{Zn}(\text{OH})_2$ is an amorphous layer, which may cover the cement molecules and can cause retardation between the GnP-GnP contact [32]. However, the obtained highest value of electrical conductivity (17.9 Scm^{-1}) was superior to the reported cement composite with carbon nanotubes [11][12] and graphene nanoplatelets [13]. The measured Seebeck coefficient values are all positive, indicating that the hybrid cement composites are a thermoelectric element of the p-type. Noteworthy, for the hybrid cement composite, the absolute values of the Seebeck coefficient are far superior to those without the nanostructured content of GnP and ZnO, since it was

coefficient improvement [15] than GnP [13]. In addition, it has been reported that the main factor in increasing the Seebeck coefficient in cementitious materials is the conduction of the hole [7][33]. This means that the increase in the cement composites Seebeck coefficient with GnP and ZnO could be related to the increase in the contribution of holes. The maximum Seebeck coefficient increases by increasing the content of nanoparticles until it reaches the value of 10.0 wt% GnP and 10.0 wt% ZnO of cement. For 5_CC and 15_CC, the maximum Seebeck coefficient is 91.9 μVK^{-1} and 87.73 μVK^{-1} , respectively. The greatest Seebeck coefficient of 141.5 μVK^{-1} is achieved at 71 °C for 10_CC. Although the most substantial electrical conductivity was shown by the composites at 15.0 wt% GnP and 15.0 wt% ZnO, their Seebeck coefficient is lower than 10_CC (10.0 wt% GnP and 10.0 wt% ZnO). It may be attributed to the relatively higher concentration of the carrier in the composites because the coupling effect for thermoelectric materials exists between the conductivity and the Seebeck coefficient [34].

It is observed that the specific heat capacity of the hybrid cement composites increases with an increase in temperature while the thermal diffusivity shows an opposite characteristic. Multiplication of these two contrary parameters nullifies its thermal conductivity effects. As a consequence, the thermal conductivity is almost a constant value in the measured temperature range. The average value of κ for 5_CC, 10_CC and 15_CC is found to be 0.65, 0.99 and 0.92 $\text{Wm}^{-1}\text{K}^{-1}$, respectively.

The thermoelectric figure of merit is calculated by using equation (1). The highest figure of merit for the cement composite is 0.0101 at 71 °C with 10.0 wt% GnP and 10.0 wt% ZnO loadings. Due to the improved Seebeck coefficient, the figure of merit in cement composites with GnP and ZnO loadings is mostly enhanced.

Table 3. Measured thermoelectric properties of cement composites with GnP and ZnO.

Composites	Thermoelectric properties (maximum obtained values)			
	S ($\mu\text{V/K}$)	σ (S/cm)	κ (W/mK)	ZT
5_CC	91.9	5.5	0.65	0.0028

considered cement has no Seebeck coefficient. This high value reveals that the GnP and ZnO added in the cement composites have an active thermoelectric component. However, ZnO has a superior contribution to the Seebeck

10_CC	141.5	13.9	0.99	0.0101
15_CC	87.73	17.9	0.92	0.0048

3.2 Maximum Output Power

The maximum output power (P_{\max}) is calculated by using equation (2) for the hybrid cement composites. The obtained values of Seebeck coefficients, electrical conductivity and sample's dimensions ($2 \times 2 \times 10 \text{ mm}^3$) with GnP and ZnO inclusions are employed during power calculation. The maximum power output of the prepared composites with different loading of GnP and ZnO as a function of hot side temperature is shown in Fig. 2. It is seen that the calculated highest electrical power can be reached to $6 \mu\text{W}$ when the hot side is at 71°C while the cold side is kept at 25°C by using 10.0 wt% GnP and 10.0 wt% ZnO into the cement. The calculated maximum output power for the composite with graphene and ZnO is higher than the carbon fiber based cement composites [35]. Since the measured Seebeck coefficients for 10_CC (10% GnP, 10% ZnO) sample was higher than the other two composites (5_CC and 15_CC), the maximum power for 10_CC exhibited the most.

To realize the power output generated from the real-sized area and compare it with other energy harvesting devices, P_{\max} is again calculated for the cement composites considering the sample's surface of one square meter, as shown in Fig. 3. This figure indicates that by utilizing composites in the pavements, roof or building surfaces would possess $\sim 1.5 \text{ W}$ power per square meter under a temperature difference of about 50°C using only a p-type

cement composite with both 10 wt% GnP and ZnO having a height of 10 mm. This estimated value is higher than the reported value for a thermoelectric generator by utilizing a human body source where the obtained value was 0.285 Wm^{-2} [36]. It is reported that when a PN junction is made by p- and n-type cement composite, the Seebeck coefficient at the intersection exhibited higher values than their individuals [37]. So, a further increase in total output power might be obtained to join this GnP-ZnO composite and n-type cement composites, which could remarkably improve the Seebeck coefficient.

To estimate the output power from a cement composite based thermoelectric generator (TEG), a pair of p-type and n-type composite having a junction electrically connected in series and thermally in parallel is required. In this work, only p-type cement composite was obtained. So, consideration of an n-type is necessary. Similar to this work, i.e., in terms of loading concentration, Wei et al. [10] reported an n-type cement composite with a maximum Seebeck coefficient of about $-52 \mu\text{VK}^{-1}$ at $\sim 70^\circ\text{C}$ using expanded graphite as an additive. Considering the Seebeck coefficient of that n-type composite and p-type composite (GnP and ZnO), the maximum electrical power for a structural thermoelectric generator is calculated [12]. As it is shown in Fig. 4, when a cement composite device is fabricated with expanded graphite thermoelectric element (n-type), and graphene and ZnO thermoelectric element (p-type), maximum power output as 0.55 mW under a temperature difference of 50°C can be estimated.

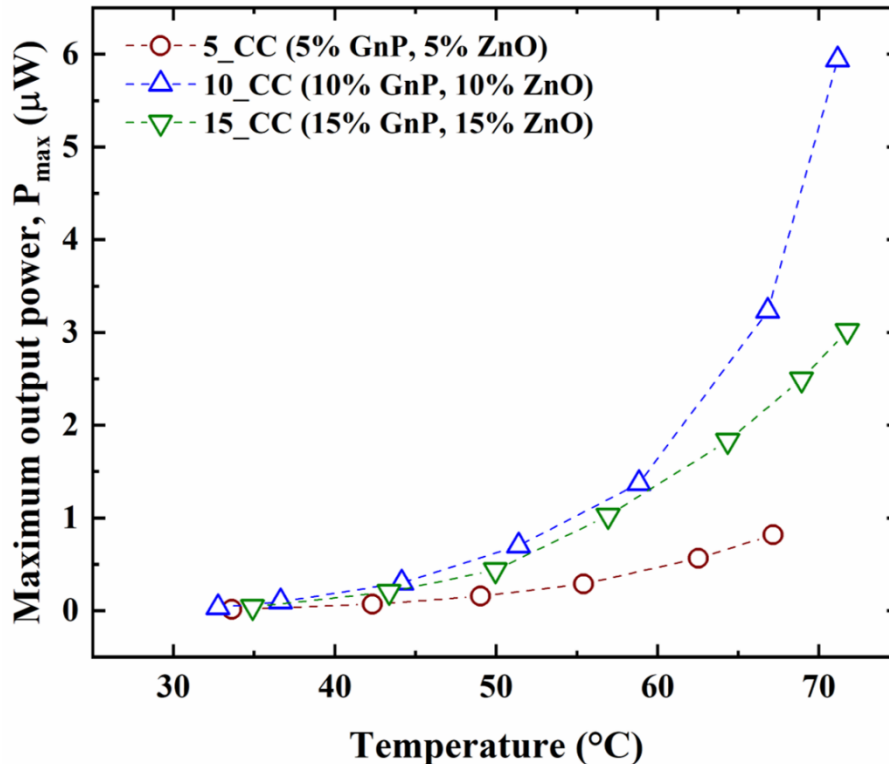


Fig. 2. Maximum power output with GnP and ZnO loadings as a function of hot side temperature.

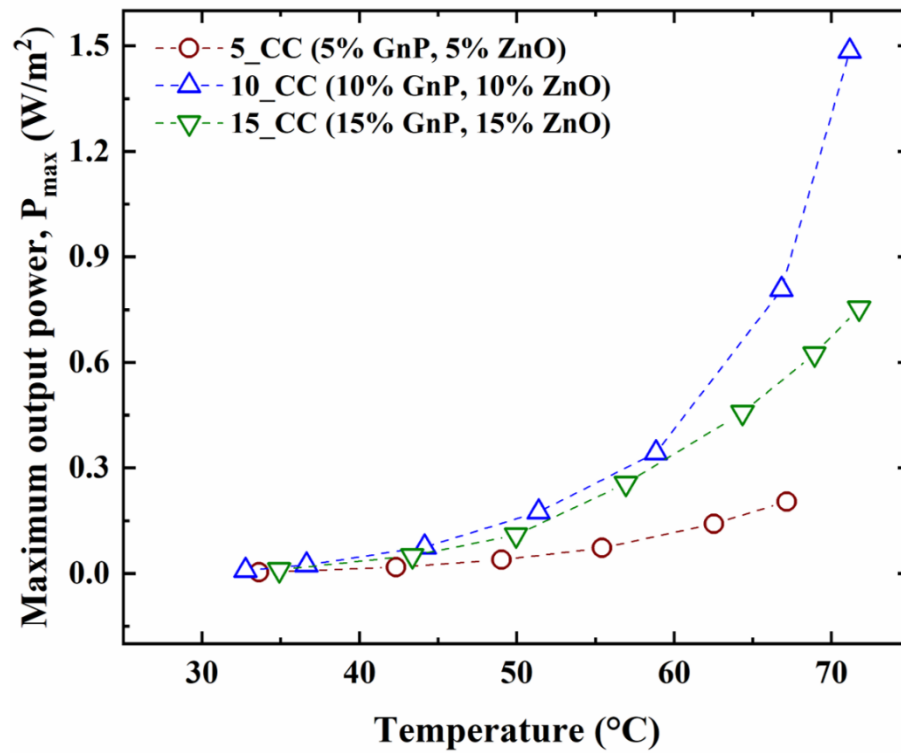


Fig. 3. Normalized maximum power output for the cement composites concerning hot side temperature with an area of one square meter.

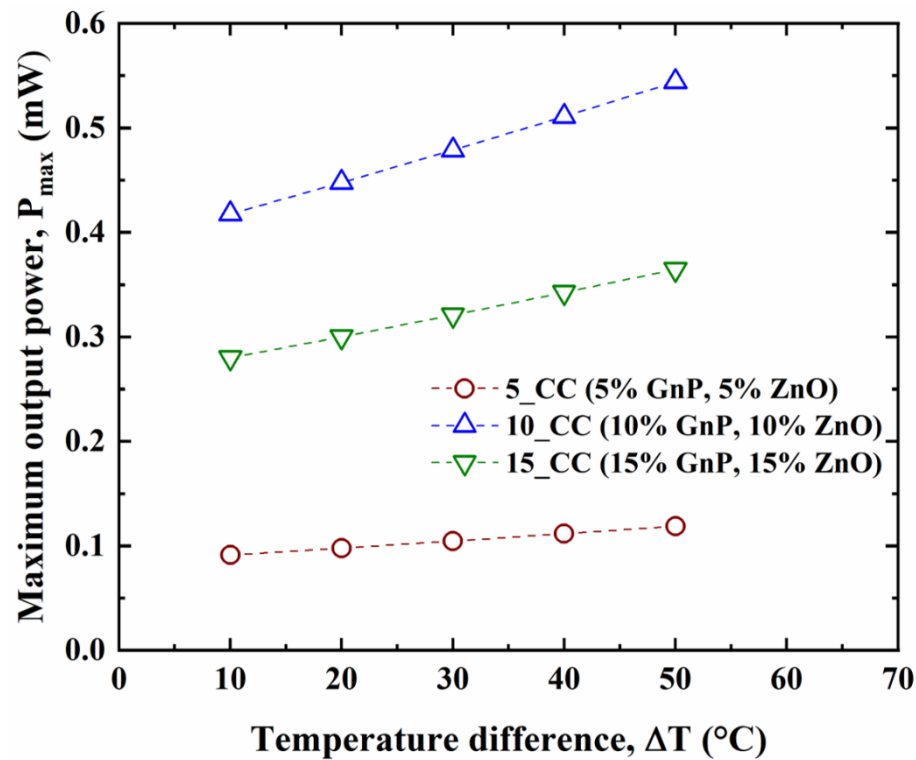


Fig. 4. Maximum power output for a cement composite based thermoelectric generator to the temperature difference.

4. CONCLUSIONS

A thermoelectric generator based on structural cement composites could potentially use in ambient energy harvesting by capturing the unused ambient heat energies in buildings, pavements, industrial environments, etc. and converting it into usable electrical energy. Approximately 1.5 W power could be achieved by a one square meter graphene nanoplatelets and ZnO added cement composite with a height of 10 mm under a temperature difference of about 50 °C. The estimated energy can be stored in a capacitor and utilized in a wired

or wireless communication network for the buildings. Integrated energy saving and harvesting technology in facilities will enable future energy-saving buildings and ‘greener’ construction towards multifunctional smart structures.

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