

The effect of temperature rise on the thermal conductivity of composite (perovskite/polymer) solar cell

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Abstract

The efficiency of solar cell and the corresponding output power is mainly affected by the rise in the film temperature, and in such cases, the stability in thermal conductivity value is required to ensure adequate rate of heat loss. This study investigates the effect of temperature rise on the overall thermal conductivity of the perovskite cell including the response of polymeric additives that have been used recently to enhance the interaction between different grains in perovskite films. The polymers under study are: polyethylene oxide (PEO), polyacetylene (PA), polythiophene (PT), polystyrene (PS) and polymethyl methacrylate (PMMA). The work assumes mathematical procedure that links the variation of temperature to the degree of polymer's crystallinity and then converting this impact in terms of thermal conductivity. The calculations assumed three different concentrations of the polymer as: 1, 3 and 5%. Each polymer has served with two degrees of crystallinity. In general, the results show that the effect of added polymers on the final value of the thermal conductivity of the film may be positive or negative depending on three main factors: k-value of the perovskite-based material, k-value of the polymer and concentration of the polymer. The effect of the polymer appears more when it is used with the low k-value perovskite material (0.2 W/m·K) rather than with high k-value perovskite material (0.8 W/m·K). The addition of PEO to the perovskite-based materials makes the overall k-value of the film reaches to 3.7 W/m·K and even higher when the temperature rises up. Hence, this polymer is useful in high operation temperature. In the case of using polymers (PA and PT), the overall k-value of the film was less sensitive. The use of amorphous polymers (PS and PMMA) has not witnessed any significant contribution in the overall k-value of the film.

Keywords: temperature rise, thermal conductivity, polymers, perovskite, solar cells

Kulcsszavak: hőmérséklet-emelkedés, hővezető képesség, polimerek, perovszkit, napelemek

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1. Introduction

Recently, solar cells including many categories, such as: silicone-based cells, concentrated photovoltaics, organic photovoltaics, cadmium telluride cells, copper indium gallium selenide (CIGS) cells, multi-junction cells, tandem cells, quantum-dot cells and perovskite cells. These types have been receiving attention from both researchers and manufacturers [1]. A perovskite solar cell is a developed type of solar cells which includes perovskite-structured compounds, mostly semiconductors such as: calcium titanium oxides with hybrid organic-inorganic lead-based materials or tin halide-based materials. The materials used in the perovskite type have witnessed many improvements to become more economic as well as energy efficient, where maximum efficiency has reached up to 25% [2-6]. The characteristics of organic-inorganic perovskite regions are the interest for recent studies to develop the performance of these cells by enhancing many preferable parameters such as: absorption coefficient, bandgap, carrier mobility and carrier lifetime [7, 8]. The crystallization of the cells is another imperative feature for high performance perovskite cells, where it is related to the light absorption and carrier transportation. The presence of defects and crosslinking

patterns may affect the performance of the cells by accelerating the carriers' recombination. Therefore, more attention has been seeking to reduce the defects and pinholes in order to obtain perfect perovskite cells [9-11].

Polymeric materials have been involved in perovskite cells as additives to enhance the general performance of the solar film and the overall conversion efficiency. In this aspect, common polymers that are used include: polyethylene oxide (PEO), polyvinylpyrrolidone (PVP), polyacrylonitrile (PAN), polyetherimide (PEI), polymethyl methacrylate (PMMA), polyacetylene (PA), polythiophene (PT), polystyrene (PS) and some other polymers [12-15]. From previous studies, polymers can be used as additives to improve the crystallization morphology by enhancing the interaction between different grains in perovskite films [16, 17], and by facilitating the nucleation and regulating the crystal growth [18-20]. Polymers can be immersing easily in polar solvents, which reduces the contact angle [21] and increases the device stability [22, 23]. Furthermore, polymers demonstrated for the improving of electrical, optical and other physical properties of perovskite-structured materials [24-31].

Polymers usually have med-to-high degree of crystallinity, where the structure of the molecules is identical partially. The degree of crystallinity has a big influence on many thermo-physical properties as well as mechanical features. However, synthetic polymers are usually semi-crystalline and their degrees of crystallinity are typically range between 10-80% [32-35]. The degree of crystallinity is an important factor affecting the thermal conductivity of semi-crystalline polymers. Where, maximum thermal conductivity achieved at high degree of crystallinity, where packed structure accelerate the heat [34]. However, in semi-crystalline polymers, thermal conductivity above room temperature gradually decreases by the increasing of temperature. This can be attributed to the loss of crystalline regions due to the heat flow [36-39]. Since, the most of solar radiation incident on the film is not converted into electricity, but it is absorbed as a heat thus it rises up the film temperature which causes a reduction in the efficiency. Access heat can be removed from internal layers mainly by heat conduction, which is related to the thermal conductivity of the material. Therefore, higher heat conductivity means efficient performance, and thus higher output power [40-45]. Hence, when temperature rises, a variation in thermal conductivity of the polymer affects the cell performance.

The object of the current study is to predict theoretically the effective thermal conductivity of the perovskite film due to the thermal behavior of polymeric additives that influence the overall performance of the solar cell. It is worth to mention that there is limited literature regarding the thermal conductivity of composite perovskite/polymer films. The novelty of the study is to present a mathematical model that links the effect of temperature on thermal conductivity of polymeric additives (with their specific concentration ratio and degree of crystallinity) to the overall thermal conductivity of the perovskite solar cell.

2. Materials and methods

In order to find the variation in the thermal conductivity of perovskite film including the thermal behavior of polymeric additives in the cell, some common polymeric materials are considered, such as: polyethylene oxide (PEO), polyacetylene (PA), polythiophene (PT), polystyrene (PS) and polymethyl methacrylate (PMMA). These materials show different features and wide range of thermo-physical properties, as shown in Table 1.

Polymer	Degree of crystallinity (%)	Density of crystalline structure (g/cm ³)	Density of amorphous structure (g/cm ³)	Thermal conductivity of crystalline structure (W/m·K)
PEO	60-80 [46]	1.24 [47]	1.12 [47]	60 [48]
PA	50-70 [49]	0.6 [50]	0.4 [50]	0.95 [51]
PT	40-60 [52]	1.18 [53]	1.12 [53]	0.62 [51]
PS	3-8 [54]	1.05 [55]	1.03 [55]	0.18 [56]
PMMA	~ 0 [57]	1.19 [57]	1.19 [57]	0.16 [58]

Table 1. Thermo-physical properties of polymers (pure solid additives)
1. táblázat A polimerek hőfizikai tulajdonságai (tisztá szilárd adalékok)

Since the intended goal is to determine the change in thermal conductivity of these polymeric materials due to the variation in the temperature of the film, hence the current mathematical procedure suggests finding the variation in the temperature with respect to an intermediate parameter which is the degree of the crystallinity, and then seeking to link this parameter to the thermal conductivity of the polymer and corresponding impact on the thermal conductivity of solar cell as overall.

The thermal conductivity of common polymeric materials is usually ranged between 0.2-1.0 W/m·K, with unique values for some others [35]. Polymers with high thermal conductivity values are required when the purpose is to release the heat. Many factors affect the thermal conductivity of a material such as: temperature, density, porosity, moisture, degree of crystallinity, chain structure, orientation of grains, size of molecules and impurities [59, 60]. The coefficient of thermal conductivity of polymers has different behaviors whether the polymer is thermoplastic or thermosetting, crystalline or amorphous and below or above glass-transition temperature [61, 62]. The increasing of the crystallinity leads to increase the thermal conductivity, thus thermal conductivity of crystalline region is higher than thermal conductivity of amorphous region [63-65]. However, the thermal conductivity of a semi-crystalline polymer is given by [34]:

$$k_{po} = X_c k_c + (1-X_c) k_a \quad (1)$$

Where;

k_{po} : thermal conductivity of the semi-crystalline polymer.

k_c : thermal conductivity of crystalline part.

k_a : thermal conductivity of amorphous part.

X_c : degree of crystallinity of the polymer.

The last equation can be re-written as;

$$k_{po} = [(1 - Z) X_c + Z] k_c \quad (2)$$

Where (Z) is the ration between (k_a) and (k_c).

Since, the thermal conductivity is influenced by the density, where the higher density material has the higher thermal conductivity and the increasing in the crystallinity of the polymer leads to increase both: density and thermal conductivity in the same manner [66]. Thus, the ratio (Z) can be presented in terms of density, as following;

$$Z = \rho_a / \rho_c \quad (3)$$

During temperature rise, the thermal conductivity of the semi-crystalline polymers (PEO, PA and PT) decreases with respect to the increasing in the temperature. This can be attributed to the phenomenon of Umklapp scattering process, which occurs in the boundary of lattice where it increases the momentum of phonon. By rising the temperature, this process becomes more frequent and the phonon free path decreases, thus less ability to conduct the heat [61, 62]. For PS and PMMA, since the structure of these polymers is amorphous, so they show different behavior with temperature. Where, the thermal conductivity increases with the increasing of the temperatures in association with the increasing of the specific heat, and this attributed to the increase of the thermal activation of

the phonons due to the molecular vibration within the lattice [58, 61].

Hence, the general form of thermal conductivity-temperature relationship has the following linear formula:

$$k_p = k_{p0} + \mu T \quad (4)$$

Where:

k_p : thermal conductivity of the polymer as a function of temperature.

k_{p0} : thermal conductivity of the polymer at room temperature.

μ : index of variation.

T : temperature.

The index of the variation with the temperature within the range 20-100 °C can be extracted from specific studies that investigated the temperature effect on the thermal conductivity of the selected polymers, as shown in Table 2.

Polymer	Index of variation
PEO	- 0.3 [48]
PA	- 0.004 [51]
PT	- 0.002 [51]
PS	+ 0.0006 [56]
PMMA	+ 0.0008 [56, 58]

Table 2 The index of variation of thermal conductivity of polymers with the temperature

2. táblázat A polimerek hővezető képességének változási indexe a hőmérséklet függvényében

Perovskite-based materials usually have thermal conductivity values between 0.2-0.8 W/m·K at room temperature, and it may be even more for some compositions [67, 68]. During the operating and exposing to sunlight, high film temperature can lead to the degradation of perovskite solar cells due to burn-in, defects formation, films cracking, and delamination at the layers' interfaces. Keeping high thermal conductivity is essential to reduce the film temperature and facilitating the dissipation of accumulated heat. However, the interested thing about these materials is that as temperature increases, many parameters increases as well, such as: number of free electrons, lattice vibrations, thermal activation of the phonons and specific heat [69]. Thus, the thermal conductivity of the perovskite-based material is expected to increase. It is noticed that the variation of thermal conductivity with the temperature (within 20-100 °C) for low k -value of perovskite material is generally has the form [70, 71]:

$$k_v = k_{v0} + 0.001 T \quad (5)$$

Where, k_{v0} is the k -value of the perovskite-based material at room temperature. While, the variation of k -value with the temperature for high thermal conductivity perovskite material is generally appeared as [72, 73]:

$$k_v = k_{v0} + 0.02 T \quad (6)$$

Finally, the overall k -value of the whole perovskite film is the summation of both k -value of perovskite-based materials (k_v) and k -value of polymeric additives (k_p) according to the mixing rule of composite materials and depending on the content of

perovskite-based materials (φ_v) and the content of polymeric materials (φ_p). Thus:

$$k = k_v \varphi_v + k_p \varphi_p \quad (7)$$

Take into account that the concentration of polymeric materials involved within the whole structure of the film in the perovskite cell is between 1-5 wt% [12-31].

3. Results and discussion

This study includes theoretical analysis to predict the overall thermal conductivity of the perovskite film including the variation happens in the thermal conductivity of polymers that usually involved within the film to enhance the performance of the solar cell. As mentioned previously, five types of polymeric materials have been included in the calculations with their properties. The polymers are: polyethylene oxide (PEO), polyacetylene (PA), polythiophene (PT), polystyrene (PS) and polymethyl methacrylate (PMMA). The calculations assumed three different concentrations of the polymeric additives as: 1, 3 and 5%. Each polymer has served with two degrees of crystallinity; i.e. the maximum value and the minimum value. The thermal conductivity of the perovskite-based material has two values as; 0.2 and 0.8 W/m·K (designed values). The range of temperatures considered was between 20-100 °C.

In general, the low thermal conductivity of perovskite composites is attributed to the shorter lifetimes of the optical phonons and the smaller group velocity of acoustic phonons [73]. Take into account the effects of the values and distribution of band gaps [74].

The results show that the effect of added polymers on the final value of the thermal conductivity of the film may be positive or negative depending on three main factors: k -value of the perovskite-based material, k -value of the polymer and concentration of the polymer. As for the degree of crystallinity, it had no significant effect because the variance in the k -value was very small. Also, the effect of the polymer appears more when it is used with the low k -value perovskite material (0.2 W/m·K) because of the closeness of this k -value with the k -value of the polymer. But when it is used with the high k -value perovskite material (0.8 W/m·K), the perovskite will dominate for two reasons; firstly, its high k -value compared to the polymer, and secondly because of the accelerated increase of this value when the temperature rises comparing to the slow change that occurring on the k -value of the polymer.

The deep analysis of the results has shown that the thermal conductivity of semi-crystalline polymers (PEO, PA and PT) has decreased by the increasing in the temperature, due to less mean free path of phonons, thus it may have a negative impact on the overall thermal conductivity of the perovskite film. On the other hand, the thermal conductivity of amorphous polymers (PS and PMMA) has increased by the increasing in the temperature, due to thermal activation of the phonons, thus it may have a positive impact on the thermal conductivity of overall perovskite cell. However, each polymer has shown some interesting behaviors and intervention effects of polymeric concentration and thermal conductivity of perovskite-based materials.

The addition of PEO to the film has introduced a unique contribution as a semi-crystalline polymer to the perovskite cell due to its high thermal conductivity value (60 W/m·K). This makes the overall k -value of the film reaches until (3.7 W/m·K) at room temperature which is higher than the k -value of the perovskite-based material and it remains high at any range of temperatures or polymeric concentrations, as shown in Fig. 1. The overall k -value was between 0.7-3.0 W/m·K for the film of perovskite-based thermal conductivity of (0.2 W/m·K), and between 1.3-4.0 W/m·K for the film of perovskite-based thermal conductivity of (0.8 W/m·K). Hence, this polymer can be useful in application requires efficient heat conduction and thus to obtain higher power efficiency. However, for low perovskite thermal conductivity (0.2 W/m·K), the rise in the temperature up to 100 °C has shown a decreasing in the k -value by 25-45% in comparing to the reference value at room temperature (20 °C). While, for high perovskite thermal conductivity (0.8 W/m·K), the rise in the temperature up to 100 °C has shown an increasing in the k -value by 10-90% in comparing to the reference value at room temperature. The decreasing in the first case is mainly due to the polymer thermal behavior which was the dominant, where at low value of perovskite k -value (0.2 W/m·K) and its corresponding thermal response which was low comparing to the highly response of the polymer that made the overall film follows the polymer behavior. While, in the second case ($k_v=0.8$ W/m·K), the thermal response of the perovskite was higher, thus the overall film follows the perovskite behavior not the polymer. Furthermore, the increasing in the degree of crystallinity of the polymer from 60 to 80% enhanced the thermal conductivity of the film by 2-3% only, which is disappointed. Note that the increasing in the PEO concentration results in even higher k -values in overall.

In the case of using polymers (PA and PT) that have relatively closest thermal conductivity values to the perovskite-based materials, the overall k -value of the film was less sensitive to the thermal behavior of the polymeric materials, especially at the high design k -value (0.8 W/m·K). The results of adding PA to the film, shown in Fig. 2, have revealed an effective thermal conductivity usually more than the designed value at any range of temperatures or polymeric concentrations. However, at low design k -value (0.2 W/m·K), the behavior of increasing in overall k -value is attributed to the fact that the k -value of the polymer was higher in comparing to the perovskite-based material, even though the decreasing ratio in the polymer k -value was higher than the increasing ratio in the perovskite k -value. At high design k -value (0.8 W/m·K), the sensitivity of polymer effect was relatively less due to higher k -value of the perovskite-based material compared to the polymer, and secondly because of the faster increase in the k -value of the perovskite-based material with the temperature comparing to the slow decreasing in the k -value of the polymer. A similar behavior can be seen by the adding of PT to the film, as shown in Fig. 3, with even less influence due to the lower k -value of PT than that of PA, and corresponding thermal response of PT which was lower than that of PA. However, the rise in the temperature up to 100 °C increased the referenced k -value of the film for both PA and PT up to 50% for ($k_v=0.2$ W/m·K) and up to 150% for ($k_v=0.8$ W/m·K), where this is actually the same increasing in the k -value of the perovskite-based materials. Also,

the enhancement in k -value due to increasing in the degree of crystallinity of the polymer was negligible.

In case of adding amorphous polymers (PS and PMMA), that already have low values of thermal conductivity and slow thermal response, the overall k -value of the film is rather a function of perovskite-based materials only, even at high temperatures, as shown in Fig. 4 and 5, respectively. However, the involving of more than 1% of these polymers within the film was not sufficient to rise the effective thermal conductivity higher, thus it may be a useless choice in the design.

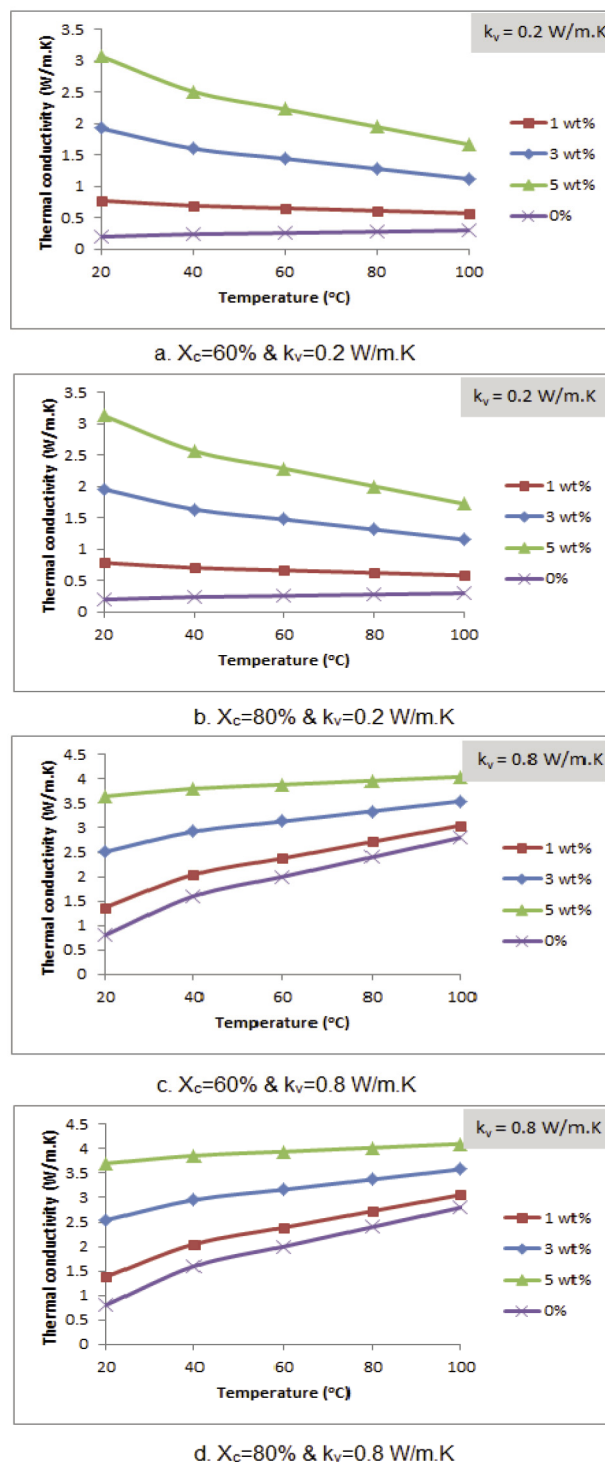
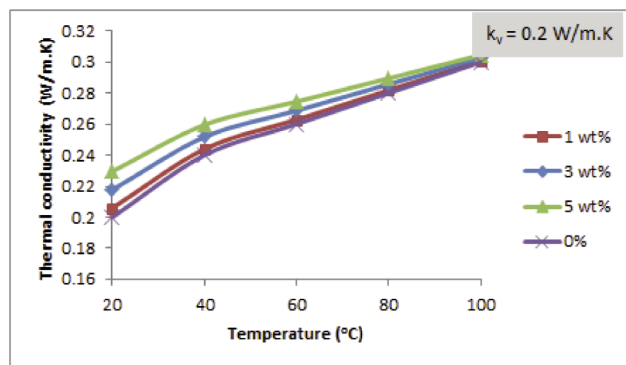
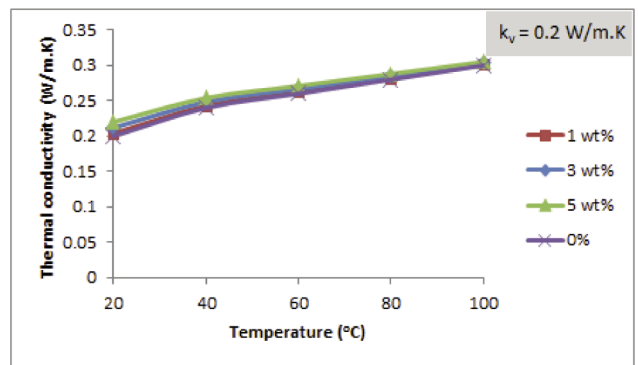


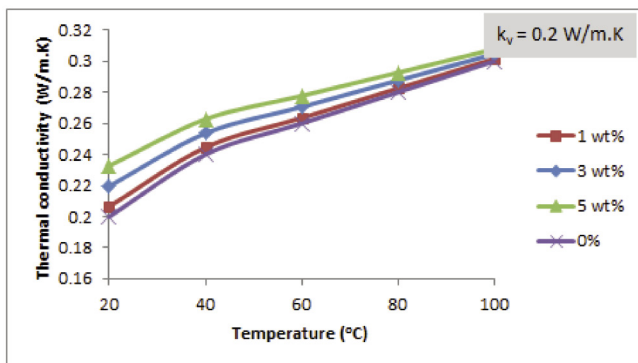
Fig. 1 Overall thermal conductivity of perovskite cell at different PEO concentrations
1. ábra Perovskit cella teljes hővezető képessége különböző PEO koncentrációk mellett



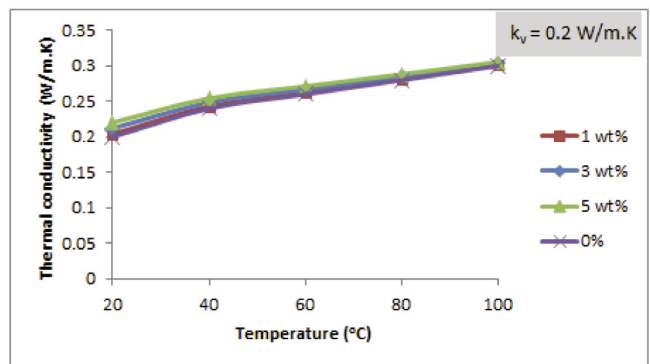
a. $X_c=50\%$ & $k_v=0.2$ W/m.K



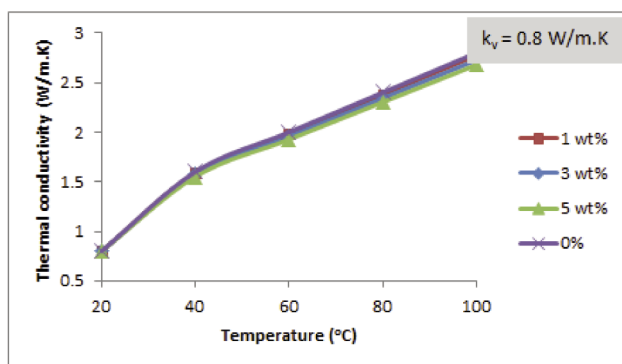
a. $X_c=40\%$ & $k_v=0.2$ W/m.K



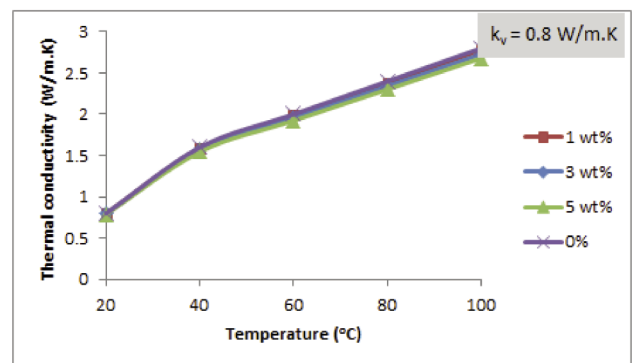
b. $X_c=70\%$ & $k_v=0.2$ W/m.K



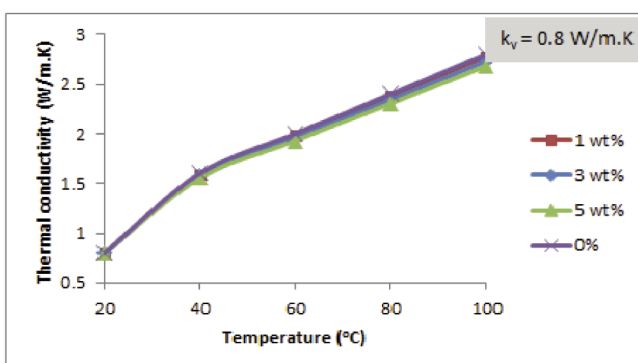
b. $X_c=60\%$ & $k_v=0.2$ W/m.K



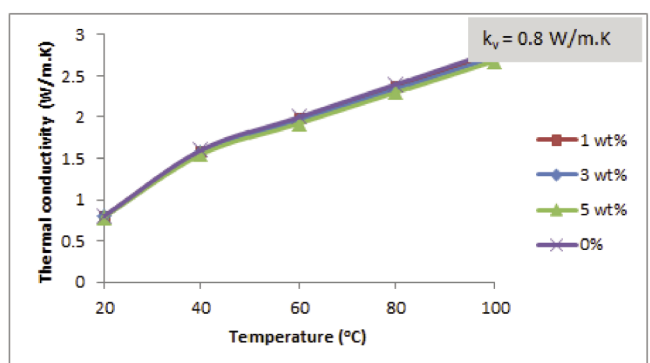
c. $X_c=50\%$ & $k_v=0.8$ W/m.K



c. $X_c=40\%$ & $k_v=0.8$ W/m.K



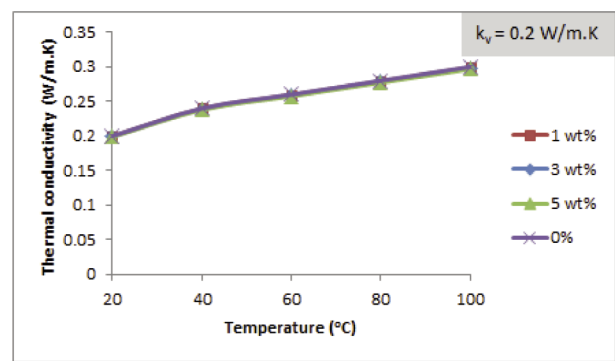
d. $X_c=70\%$ & $k_v=0.8$ W/m.K



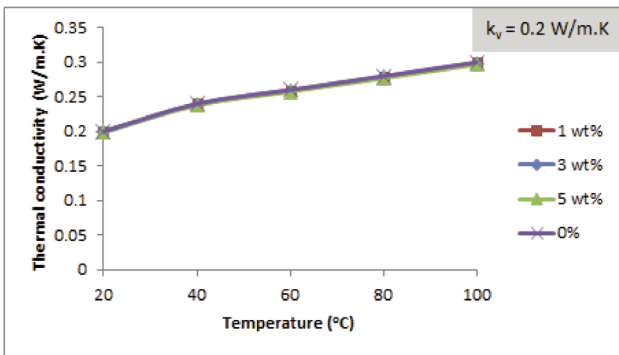
d. $X_c=60\%$ & $k_v=0.8$ W/m.K

Fig. 2 Overall thermal conductivity of perovskite cell at different PA concentrations
2. ábra Perovszkit cella teljes hővezető képessége különböző PA koncentrációk mellett

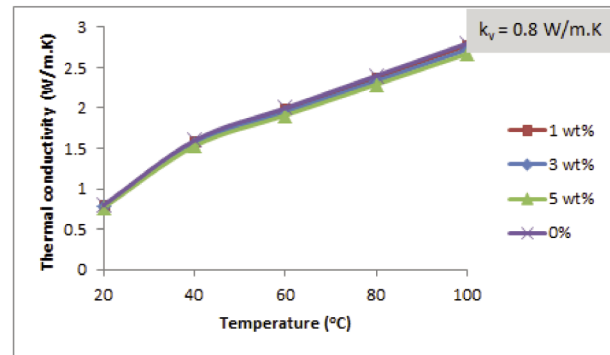
Fig. 3 Overall thermal conductivity of perovskite cell contains at PT concentrations
3. ábra A perovszkit cella teljes hővezető képessége PT koncentrációban



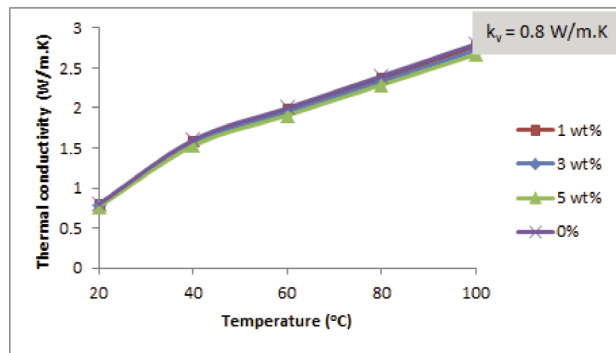
a. $X_c=3\%$ & $k_v=0.2$ W/m.K



b. $X_c=8\%$ & $k_v=0.2$ W/m.K



c. $X_c=3\%$ & $k_v=0.8$ W/m.K



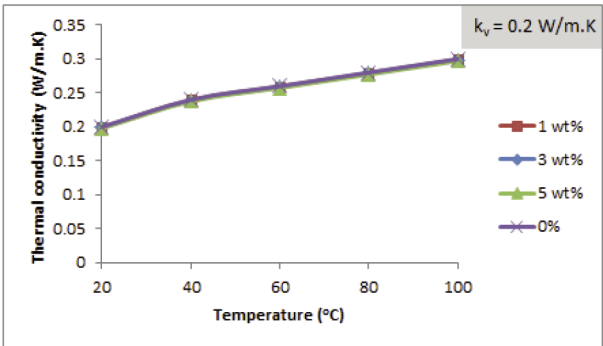
d. $X_c=8\%$ & $k_v=0.8$ W/m.K

Fig. 4 Overall thermal conductivity of perovskite cell at different PS concentrations
4. ábra Perovszkit cella teljes hővezető képessége különböző PS koncentrációk mellett

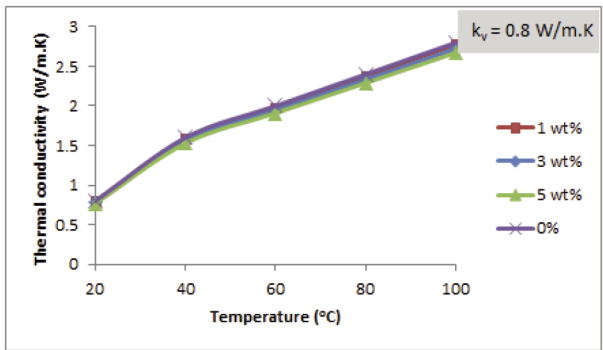
For validation purpose, the current results can be compared with that obtained from certain studies that may help to understand the differences, as shown in *Table 3*.

Study	Added polymers and loadings	Results
Current	PEO, PA, PT, PS and PMMA 1-5% wt.	The addition of PEO to the perovskite makes the overall k-value higher by 2-3 times, and it is reliable for any temperature rise. In the case of using polymers PA and PT, the overall k-value has fair effects. The use of PS and PMMA has no significant effects.
[16]	PFN, PMMA and PS 1-3% wt.	The addition of polymers enhances the crystalline growth. This leads to improve the thermal conduction. Results shown that increasing the amount of PMMA and PS is not required due to very large size aggregates and non-uniform structure, which leads to less ordered crystalline chains.
[17]	PEO 1% wt.	The addition of PEO enhances the power efficiency by 25%. This satisfies better thermal conduction.
[18]	PEI 1% wt.	The addition of PEI improves the crystallinity of the film, as well as enhances the power efficiency by 26%. This leads to improve the thermal conduction.
[23]	PVP 3% wt.	The addition of PVP improves the crystallinity of the film, as well as enhances the power efficiency by 30%. This leads to improve the thermal stability and thermal conduction.
[70]	N/A	The k-value kept between 0.3-0.4 W/m.K up to 150 °C.
[71]	N/A	The k-was kept between 0.25-1.0 W/m.K up to 100 °C with a drop in the inclination.
[75]	Theoretical approach	At elevated temperatures up to 54 °C, the k-value was 0.14 W/m.K for the iodine-based perovskite film, and 0.08 W/m.K for the chlorine-based perovskite film.

Table 3 Comparison with certain studies
3. táblázat Összehasonlítás egyes tanulmányokkal



a. $X_c=0\%$ & $k_v=0.2$ W/m.K



b. $X_c=0\%$ & $k_v=0.8$ W/m.K

Fig. 5 Overall thermal conductivity of perovskite cell at different PMMA concentrations
5. ábra Perovszkit cella teljes hővezető képessége különböző PMMA koncentrációk mellett

4. Conclusions

The study linked the impact of changing the thermal conductivity of some common polymeric additives due to temperature rise on the effective thermal conductivity of the overall perovskite film. The obtained results by the calculations yield the following conclusions:

- In general, polymeric additives may have positive or negative contributions on the final value of the thermal conductivity of the film based on the factors: k -value of the perovskite-based material, k -value of the polymer and concentration of the polymer.
- Polymers affect thermally when the perovskite-based material is low (0.2 W/m·K) rather than with high k -value perovskite material (0.8 W/m·K).
- The existence of PEO within the perovskite-based materials makes the overall k -value of the film high at any range of temperatures or polymeric concentrations.
- The overall k -value of the film involved PA and PT was less sensitive to the thermal behavior of the polymeric materials, especially at the high design k -value (0.8 W/m·K).
- The overall k -value of the film has not affected essentially by the adding of amorphous polymers (PS and PMMA).
- There is no significant influence of the degree of crystallinity of the polymer on the overall thermal conductivity of film.

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