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Innovative artificial leather with high thermal conductivity as a new leather product

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Abstract

Artificial leather has been developed for decades as a key element in apparel design due to its thermal insulation function. However, with the changes of public preferences, the focus has shifted from functionality to a greater awareness of aesthetics. Therefore, an area for innovative artificial leather with high thermal conductivity as the cover of smart electronic devices is emerging. This leather will enable both special functions and aesthetical appearance to catch the attention of the public. It can also be utilized for spring–summer clothing in following fashion trends. Exploring new markets also means breaking away from the traditional seasonal cycle. However, the only current method for improving thermal conductivity (k) is to reduce its thickness, which sacrifices durability and appearance. Therefore, in this research, a novel solution is established, in which an additive with high k (TCA), aluminum oxide (Al_2O_3), is added into the resin layer of artificial leather at appropriate ratios to improve its thermal conductivity. Currently, the two most adopted production methods are used to develop a prototype without generating any extra cost. The results indicate that the modified leather has a high k , has an aesthetically pleasing appearance and a soft and cool hand feeling. The k of HTCAL is 48.3% higher than that of normal leather. In addition, in wear trials, a larger temperature difference is found between the skin and the HTCAL, indicating that HTCAL has a good heat transfer. Finally, a prototype for an iPhone case is developed and it is established that the method can be used for mass production to realize benefits.

Keywords

artificial leather, resin layer, thermal conductivity, additive

Artificial leather has been the subject of research for decades as a key element in apparel design systems, due to its thermal insulation function, which can resist extreme climates. However, during the past, its function has been limited to thermal insulation to create a warm internal microenvironment for wearers.^{1–6} This traditional way of using artificial leather means that the commercial value of artificial leather is very much affected by the climate and thus is limited to certain seasons. However,

times have changed and, due to the demands of modern lifestyles and the changes in public preferences from a focus on functionality to a greater awareness of aesthetics, the industry is also changing in sync with the market demands. As production technology has matured, the performance of leather has also improved and its many functions have been realized. Artificial leather is now slowly replacing real animal leather. However, the competition is becoming more intense than ever and the market now tends to be integrated and fully saturated at the same time. Artificial leather manufacturers continue to seek technological breakthroughs so that there are new markets, and consequently achieve new business growth. Meanwhile, the problems associated with environmental concerns, such as over-supply and environment sustainability, are increasingly becoming salient, thus revealing that the industry is in a precarious situation.⁷ Therefore, manufacturers are now seeking technical innovations, starting from the analysis of new market needs and social trends. Taking advantage of the upcoming market trends so that artificial leather will become a product of modern life will be the key to addressing the current dilemma.

One of the means is to market artificial leather as an innovative material with high thermal conductivity (HTCAL); it has never been marketed in this way before. The material can be used as the outside packaging or protective cover of smart electronics, especially smart phones. In considering the popularity of social connections made through social media and the pursuit of human-oriented design approaches, the continuing popularity of information technology in the next decade cannot be contested.^{8,9} Therefore, the proposed innovative HTCAL, as packaging or a protective cover for electronic devices, could be a very popular item in the market. Furthermore, with fashion trend shifts in that more and more often leather is being applied as part of spring–summer clothing collections, artificial leather will be even more popular, because the thermal insulation property of leather severely impacts its wear comfort. The traditional business model that considers leather use only during cold weather is in need of an update. Therefore, this research will provide a novel solution to improve the thermal conductivity of artificial leather to explore more possibilities for its use in product designs and realize both function and aesthetics. Therefore, a summer vest and an iPhone case are the prototypes made by using HTCAL to demonstrate the future potential of this innovative product.

Current production methods

Artificial leather is usually composed of two layers: (1) polymer resin and (2) fabric base layers (as shown in Figure 1(a)).¹⁰ The resin layer is laminated onto the base layer. In the current market, there are two methods for producing artificial leather: the (1) dry and (2) wet methods. The processes of these two production methods are shown in Figures 1(b) and (c), respectively. The dry method is an indirect method in which the resin layer is laminated onto the release paper by using a roller first and is later coated onto the fabric base layer with the backside of the resin layer. Normally, the resin layer, which is produced by using the dry method, is thicker than 0.2 mm. The wet method is a direct production method in which the resin layer is laminated onto the base layer and immersed into water to solidify afterwards. During the solidification, a foaming process will take place in the polymer of the resin layer when the resin layer is immersed in water. Therefore, resin layers produced by using the wet method are usually thicker than those produced by using the dry method. Normally, the thickness is more than 0.8 mm. Besides the difference in thickness, different methods mean that the resultant products will present different properties, such as leather that is produced by using the dry method showing a smooth surface or special embossed appearance with a firm and stiff inner polymer structure, while leathers that are produced by using the wet method shows a soft and blocky hand feel with a micro-cell foam structure in the polymer of the resin layer.^{11–13} However, there is no specific technique to create a special embossed appearance on leathers produced using the wet method. Therefore, different production methods are adopted to create different samples with different concentrations of additives with a high thermal conductivity (TCAs) added into the resin layer. Their thermal transfer performance is evaluated. Afterwards, in order to simultaneously achieve both function and aesthetics, and offer more opportunities for product designs to cater to mass production requirements, both the dry and wet methods are combined and adopted consecutively with their aforementioned strengths merged into the final HTCAL with no extra cost generated, which could facilitate the progress of transferring this research technology into real industry applications.

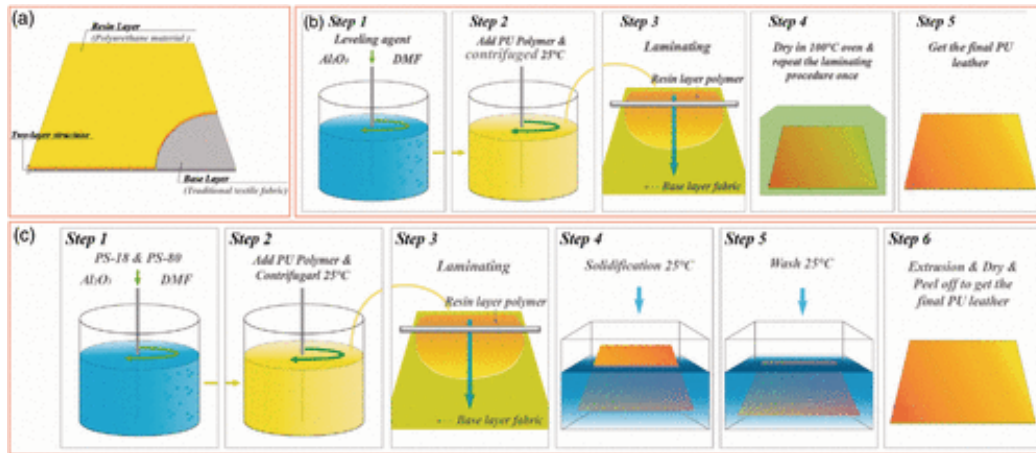


Figure 1. (a) Two-layer structure of artificial leather. (b) Production steps with the dry method, an indirect coating method. (c) Production steps with the wet method, a direct coating method.

Additives with high thermal conductivity

There are two main approaches to improve the thermal conductivity (k) of polymer composites: (1) modify and synthesize polymers to provide improved structure that has high thermal conductivity, such as full crystallization during the synthesis procedure to achieve heat transfer through phonons; or (2) add a large amount of TCA into the polymer. The most utilized TCAs are metals and inorganic fillers. Between these two approaches, the latter is more economic and efficient, easier to operate and enables the resultant product to have better corrosion resistance.^{14,15} Aluminum oxide (Al_2O_3), silicon carbide (SiC) and aluminum nitride are the three most utilized TCAs. They all can provide superior abrasion, electrical insulation and chemical resistance. However, during the wet method production processes, water will react with the AlN added into the resin layer, which generates air cells. Meanwhile, when SiC is added into polymer paste of the resin layer, it cannot be evenly dispersed. These two TCAs therefore affect product standards. Thus, in this research, Al_2O_3 is used to improve the thermal conductivity of artificial leather with its addition into the resin layer.

Meanwhile, in considering that TCA will be utilized in mass production, the material thermal conductivity and the cost should be both taken into consideration.

Al_2O_3 (thermal conductivity (k) ($35 \text{ W/m}\cdot\text{K}$)) has the most excellent cost-to-part life performance record among these three kinds of materials and could be widely used in the production process in the industry for polymer synthesis. It is a kind of white

amorphous powder, which is insoluble in water. Besides, compared to the two other kinds of TCAs, Al_2O_3 can be more evenly distributed in polymer composites and maintain its stable chemical property.^{16–18} Therefore, in this research, Al_2O_3 , which is added into the resin layer, is used to improve the thermal conductivity of artificial leather instead of reducing the leather thickness and sacrificing its mechanical properties. The spherical particles of Al_2O_3 , which have a diameter of around 1 μm , are used with consideration of the large difference between the thickness of the resin layer and diameter of the Al_2O_3 particles so the minor differences of other mechanical properties, such as elongation, tensile strength, crease recovery, etc., compared to the original will result. Therefore, starting from the desire to achieve state-of-the-art work in the industry, and the pursuits of both manufacturers and consumers, an innovative HTCAL is developed. In order to satisfy different design requirements, two existing production methods are adopted and later combined to provide possible modification methods for polymer materials. Furthermore, the appropriate weight ratio of Al_2O_3 is fully investigated to evaluate whether there is any significant difference and a means is established to develop the HTCAL prototype, by adding Al_2O_3 into the polymer resin layer and combining the strengths of the two different production methods without generating any extra cost in industry applications.

Experimental design

Materials

Base layer fabric materials

A woven fabric was selected as the base layer fabric for all of the artificial leather samples with variations of the base layer fabric controlled. The base layer was fabricated by using polyester yarn (as the weft yarn) and silver-coated conductive nylon yarn (as the warp yarn), as shown in Figure 2.

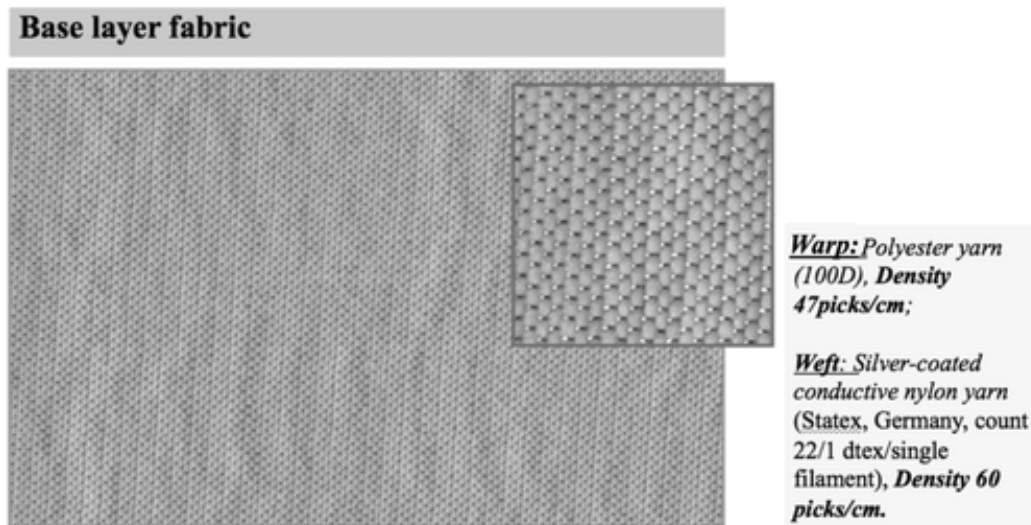


Figure 2. Woven fabric as the base layer of all samples.

Polymer materials of the resin layer

The TCA, which is Al_2O_3 , was filled into the resin layer of the artificial leather by using a dry method process to improve the thermal conductivity (k) of the artificial leather and, accordingly, slurry with Al_2O_3 added in different percentages was prepared and added to the resin layer (Table 1) and any improvements were then evaluated.

Parameter						
TCA	Al ₂ O ₃ particles			1 μm, α- phase (%) ≥ 95, 6.0 < pH < 9.0)		
Resin layer produced by using dry method						
Sample	Resin- Control	Resin- Al ₂ O ₃ 9%	Resin- Al ₂ O ₃ 12%	Resin- Al ₂ O ₃ 15%	Resin- Al ₂ O ₃ 18%	Resin- Al ₂ O ₃ 21%
XCS-2023S ^a	50 g	50 g	50 g	50 g	50 g	50 g
XCS-2045B ^b	50 g	50 g	50 g	50 g	50 g	50 g
DMF	40 g	40 g	40 g	40 g	40 g	40 g
Al ₂ O ₃	0 g	9 g	12 g	15 g	18 g	21 g
Leveling agent	0.3 g	0.3 g	0.3 g	0.3 g	0.3 g	0.3 g

Table 1. Materials of the resin layer (dry method)

Materials of artificial leather samples

After a trial was carried out to develop a resin layer by using the dry method, different artificial leather samples produced by using both the dry and wet methods were fabricated with Al_2O_3 added into the resin layer to evaluate the different effects and

improvements that are associated with different additional ratios of TCA (wt.%). The details of the materials used and composites for developing the artificial leather samples are listed in Table 2.

<i>GROUP 1: Artificial leather samples produced with dry method</i>					
Base layer	Base layer fabric with same thermal conductivity				
Resin layer	Materials of resin layer				
XCS-2023S	50 g	50 g	50 g	50 g	50 g
XCS-2045B	50 g	50 g	50 g	50 g	50 g
DMF	40 g	40 g	40 g	40 g	40 g
AL ₂ O ₃	0 g	12 g	15 g	18 g	21 g
LEVELING AGENT	0.3 g	0.3 g	0.3 g	0.3 g	0.3 g
LEATHER NO.	Dry-Control Sample	Dry-Al ₂ O ₃ 12%	Dry- Al ₂ O ₃ 15%	Dry- Al ₂ O ₃ 18%	Dry- Al ₂ O ₃ 21%
<i>GROUP 2: Artificial leather samples produced with wet method</i>					
Base layer	Base layer fabric with same thermal conductivity				
Resin layer	Materials of resin layer				
HDW-6020	100 g	100 g	100 g	100 g	100 g
DMF	40 g	40 g	40 g	40 g	40 g
AL ₂ O ₃	0 g	12 g	15 g	18 g	21 g
PS-80 ^a	1.5 g	1.5 g	1.5 g	1.5 g	1.5 g
PS-18 ^b	1.5 g	1.5 g	1.5 g	1.5 g	1.5 g
LEATHER NO.	Wet-Control Sample	Wet- Al ₂ O ₃ 12%	Wet- Al ₂ O ₃ 15%	Wet- Al ₂ O ₃ 18%	Wet- Al ₂ O ₃ 21%

Table 2. Materials of artificial leather (use of dry and wet methods)

Materials of the final HTCAL prototype

After the artificial leather samples were fabricated by using the dry and wet methods, and their thermal conductivity evaluated, a further advanced production method was implemented that would combine the strengths of both the dry and wet methods.

Therefore, the proposed HTCAL was developed by adopting both the wet and dry methods consecutively. Furthermore, a multilayered structure was developed as follows: (1) a base layer at the bottom; (2) Resin Layer 1 in the middle, produced by using the wet method; and (3) Resin Layer 2 on the top, produced by using dry method. Details on the material information are listed in Table 3.

Base layer fabric	Resin layer 1 (wet method)		Resin layer 2 (dry method)	
	Material	Mass (g)	Material	Mass (g)
Base layer fabric with same thermal conductivity	XCW-6020	100	XCS-2023S	50
	PS-80	1.5	XCS-2045B	50
	PS-18 (Penetrant)	1.5	DMF	40
	DMF	55	Al ₂ O ₃	21
	Al ₂ O ₃	21	Shimmer black pigment	2
	Shimmer black pigment	2	Leveling agent	0.3

Table 3. Ingredients for preparing resin composite

Artificial leather from the market: control

The majority of artificial leathers on the market are thermal insulating, due to the polyurethane resin used and their main use, which is for fall–winter clothing. In order to evaluate the improvements of the proposed HTCAL compared to products normally found on the market, four different types of artificial leathers (E1–E4, shown in Figure 3) were randomly selected for further investigation and their properties evaluated.

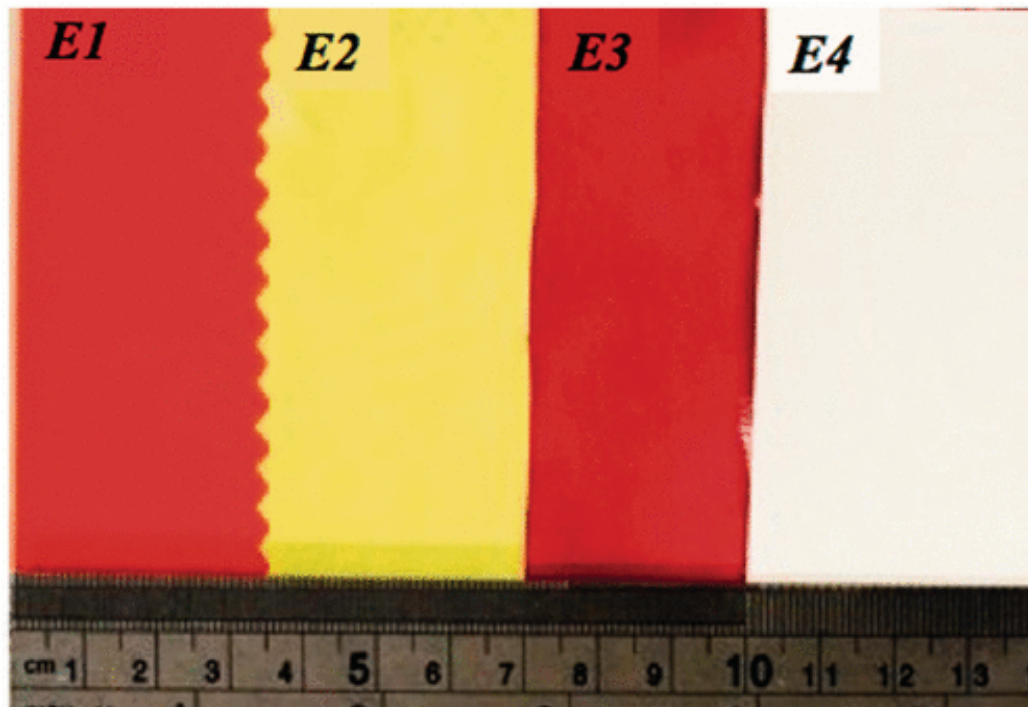


Figure 3. Randomly selected artificial leathers.

Equipment

Equipment for resin layer lamination and artificial leather production

The equipment listed in Table 4 is utilized for laminating the resin layer, modifying the artificial leather samples and developing the proposed HTCAL.

Equipment	Model	Manufacturer
Blender	SGJ110	Shanghai Shanggui Co., Ltd
Padder	LP-100	Foshan Hongxin Machinery Co., Ltd
Oven	00-101-03	Foshan Hongxin Machinery Co., Ltd
Rotating viscometer	NDJ-8S	Shanghai Intelligent Technology Co., Ltd
Electric-heated thermostatic water bath	SHB-III	Shanghai Heng Industrial Development Co., Ltd

Table 4. Equipment for fabricating samples

Equipment for evaluating sample performance

Three different evaluation methods were adopted to evaluate the artificial leather samples and the instruments shown in Table 5 were utilized.

Evaluation method		
1) Thermal conductivity (k) & Q-max (measurement of warm/cool feel) testing.	2) Use of infrared (IR) thermal imaging system	3) Temperature sensing (wear trials)
• KES-F7 Thermal Labo II; (Measuring thermal conductivity and Q-max – instant warm–cool feeling) • SDL thickness gauge.	• Direct current power supplier; • Constant electric heating fabric; • FLIR E60 Infrared Camera.	• FLIR E60 Infrared Camera; • Temperature sensors.

Table 5. Equipment for evaluating artificial leather performance

Production method

Resin layer development process

A batch of polymer resin layer samples was prepared with Al_2O_3 added in different concentrations (wt.%) to evaluate the resultant effects on the untreated resin layer. Different weigh percentages of Al_2O_3 particle fillers were added and evenly dispersed into a dimethylformamide (DMF) solution, and mixed with polymer (XCS-2023S and

XCS-2045B) afterwards. After being subjected to high-speed stirring for 15 min, and defoamed through high-speed centrifugation, the adhesive paste was coated onto release paper with at least a thickness of 0.10 mm. After they were dried at 120°C for 10 min, the resin layers with different concentrations of Al_2O_3 were peeled off from the release paper.

Procedures for the development of artificial leather

Based on the evaluation results of the resin layer samples, and in order to assess the contribution of the proposed approach to the final thermal transfer performance of the artificial leather, different whole samples of artificial leather were prepared, with the base layer attached, and Al_2O_3 added in different concentrations onto the resin layer. There were two groups of artificial leather samples, which were produced by using the dry or wet method, respectively, to compare the different effects caused by the two different production methods.

Group 1: artificial leather samples produced through the dry method

The Group 1 samples, which comprise artificial leather samples produced by using the dry method, were prepared with Al_2O_3 added in different concentrations in the resin layer (from 12% to 21%). The coating thickness onto the resin layer was changed to 0.3 mm, and after drying at 120°C for 10 min, an adhesive layer (the same polymer composite, 0.1 mm) was coated on, dried for 1 min at 120°C and coated onto the woven base layer fabric afterwards. After they were dried at 100°C for 10 min, the artificial leather samples produced by using the dry method were obtained.

Group 2: artificial leather samples produced through the wet method

The Group 2 samples, which were produced by using the wet method, were prepared with Al_2O_3 added in different concentrations in the resin layer (from 12% to 21%). Al_2O_3 was added into the resin layer polymer (HDW-6020, 100 g) and DMF solution (40 g) in different concentrations (with 1.5 g SP-80 and 1.5 g PS-18 penetrant added). After high-speed stirring for 30 min to ensure that the additives were evenly dispersed and centrifugally defoamed, the mixture composite was laminated onto the base layer fabric at a thickness of 0.8 mm, and then immersed in water to solidify for 5 min.

Afterwards, the samples were thoroughly washed (temperature: 50°C, duration: 10 min), rolling washed twice (pressure: 2 kg) and finally dried at 100°C for 60 min.

Advancing manual production of HTCAL

Based on the evaluation results of the artificial leather samples, and in order to include the advantages of (1) a glossy surface morphology, aesthetically pleasing appearance and instant cooler feeling (higher Q-max value) of the resin layers produced by using the dry method, and (2) a soft hand feel and higher thermal conductivity (k) of the resin layers produced by using the wet method, into one HTCAL product, a multilayer structure was designed (Figure 4), and with the addition of Al_2O_3 as the TCA, an internal network with high thermal conductivity was established.

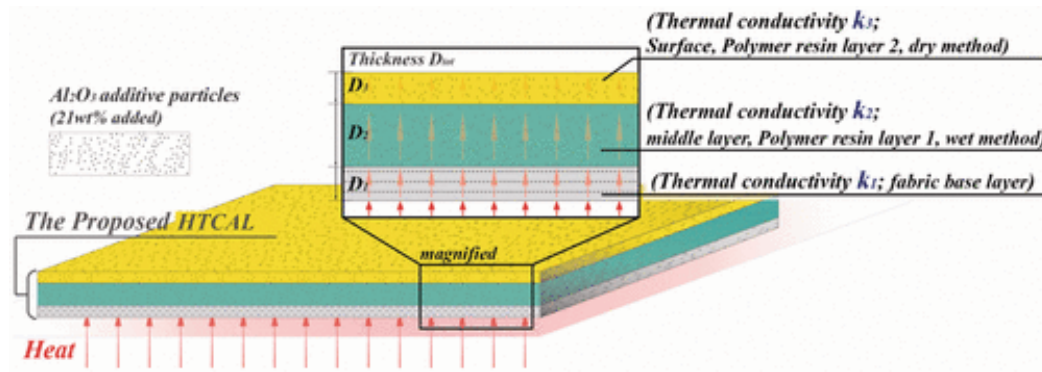


Figure 4. Proposed three-layer structure of HTCAL.

In all of these processes, no extra investment in terms of equipment replacement is required, which could facilitate the transfer of laboratory results to the production of industry prototypes, with more market opportunities created and new benefits generated.

Multilayer structure

The proposed HTCAL has a three-layer structure that consists of the following: (1) the base layer; (2) Resin Layer 1 in the middle, made by using the wet method with the addition of 21 wt.% Al_2O_3 ; and (3) Resin Layer 2 on the top, made by using the dry method also with the addition of 21 wt.% Al_2O_3 (Figure 4). The wet method was

first applied to laminate Resin Layer 1 onto the base layer, so as to form the core body with high thermal conductivity (k) and to obtain a soft hand feel. Afterwards, the dry method was adopted to laminate Resin Layer 2 onto the base layer, thus providing a glossy surface appearance with an instantly cooler feeling.

Production steps

Both dry and wet polymer resin composites were prepared with the addition of 21 wt.% Al_2O_3 as the additive, and then centrifugally defoamed (viscosity of Resin Layer 1 with the use of the wet method: 4000–5000 cPs; viscosity of Resin Layer 1 with the use of dry method: 3500–5000 cPs). The base layer fabric was ironed and coated with a 0.8 mm thick Resin Layer 1 by using the wet method. After the product semi-coagulated, it was washed and dried, and Resin Layer 1 was coated onto release paper with a thickness of 0.2 mm and dried (dry method). A thin film (the same dry method composite) was laminated onto the base layer with a thickness of 0.1 mm and dried at 120°C for 30 s. Afterwards, the formerly semi-coagulated product that was produced with the wet method was pasted onto the release paper, dried at 140°C for 20 min and then peeled off the release paper. The proposed HTCAL prototype was thus obtained. Meanwhile, in order to evaluate the overall effects, a control sample that was produced in the same way and composed of the same material ingredients, except that no Al_2O_3 fillers were added, was developed for comparison purposes.

Assessment methods

Testing of thermal conductivity (k) and Q -max (measurement of warm/cool feeling)

All of the samples were conditioned under standard conditions (temperature (T) $21.1^\circ\text{C} \pm 2^\circ\text{C}$, relative humidity (RH) $65\% \pm 5\%$) for 24 h prior to each test. All of the sample thicknesses were measured by using an SDL thickness gauge. The thermal conductivity (k) and Q -max of all of the samples were tested by using a KES-F7 Thermo Labo II thermal property measurement instrument, Kato Tech. Co., Ltd, Japan (Figure 5). The T_0 of the hot plate (BT-box, Figure 5(b)) and the cold plate (water box, Figure 5(b)) were controlled at $(30 \pm 0.3)^\circ\text{C}$ and $(20 \pm 0.3)^\circ\text{C}$, respectively. The tested samples were placed between the water and BT-boxes with a heat flow loss W (watt) of the BT-box measured when a constant value was reached

(Figure 5(c)). The thermal diffusion along the horizontal direction was not considered. Based on Fourier heat conduction equations (equation 1), the rate of the heat flow W is proportional to the cross-section area A and temperature gradient dT/dx

$$W = -kA \frac{dT}{dx} \quad (1)$$

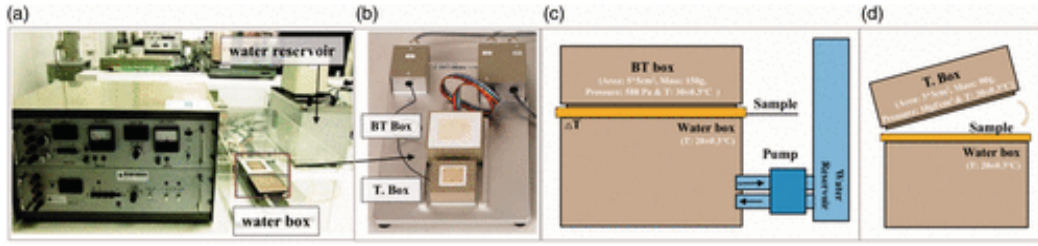


Figure 5. (a) KES-F7 Thermo Labo II instrument. (b) Enlarged view of BT-box, T.box and water box. (c) Schematic diagram of the thermal conductivity (k) test. (d) The schematic diagram of the Q -max test.

The thermal conduction model is assumed to apply to the proposed HTCAL shown in Figure 6. Heat flow passes through three layers, composed of different materials (base layer fabric and Resin Layers 1 and 2) with different thermal conductivities. Since every layer is closely bonded to each other at both sides, the temperature gradient can be further assumed to be constant through the combined plane. For each layer, the heat flow passed through will be

$$W = -k_i A \frac{(T_i - T_{i-1})}{\Delta x} \quad (2)$$

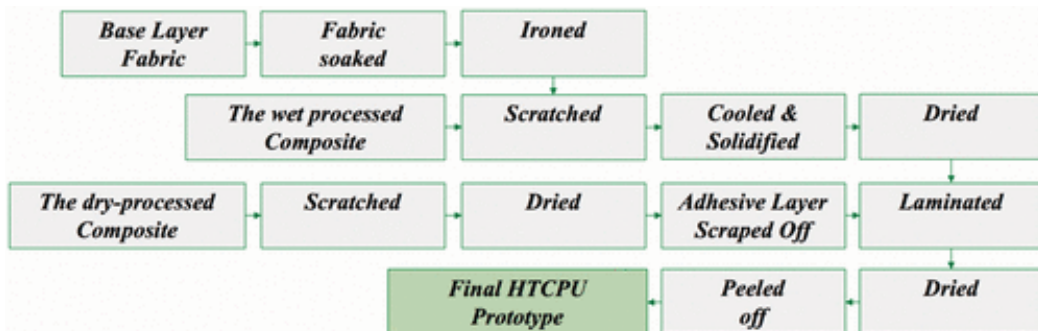


Figure 6. Flow chart of producing the HTCAL prototype.

For all three layers

$$T_3 - T_2 = -q \frac{\Delta x_3}{k_3} T_2 - T_1 = -q \frac{\Delta x_3}{k_2} T_1 - T_0 = -q \frac{\Delta x_1}{k_1}$$

$$\Delta T = -q \left(\frac{D_1}{K_1} + \frac{D_2}{K_2} + \frac{D_3}{K_3} \right), q = \frac{W}{A} \quad (3)$$

$$q = -\Delta T \left(\frac{D_1}{K_1} + \frac{D_2}{K_2} + \frac{D_3}{K_3} \right) \quad (4)$$

where D_1 , D_2 , D_3 and D_{tot} are the thicknesses of the base layer fabric, Resin Layers 1 and 2 and all of the HTCAL samples; T_1 , T_2 and T_3 are the surface temperature of the base layer fabric and Resin Layers 1 and 2, and k_1 , k_2 , k_3 and k' are the thermal conductivity of the base layer fabric, and Resin Layers 1 and 2 and all of the HTCAL samples. Moreover, q is the heat flow rate per unit area. When the thermal conductivity is measured as the integration layer

$$q = -\Delta T \frac{D_{\text{tot}}}{K}, D_{\text{tot}} = D_1 + D_2 + D_3$$

From equation (4)

$$k' = D_{\text{tot}} \left(\frac{D_1}{K_1} + \frac{D_2}{K_2} + \frac{D_3}{K_3} \right) \quad (5)$$

Q -max (w/cm²) is an indicator that is used to quantify the instant warm/cool feeling when the skin comes into contact with objects. A higher value means a cooler feeling instantly. The Q -max was measured by building up heat in a solid copper plate (0.41855 J/°C) that was 9 cm² in size and 9.79 g in mass, to measure the stored energy passed to the lower temperature end once in contact with a hotter surface (Figure 5(d)). The peak value (peak reached 0.2 s after contact with the filter) of the shifted

heat current is the Q -max, which can directly differentiate between a warm–cool feeling with an instant touch.^{19–21}

Use of the infrared thermal imaging system

An intelligent electric heating machine for fabric (Figure 7) with heat insulating ability that could reach a constant surface temperature with a voltage of 7 V and current of 0.8 A after 30 min was used as the heat supply. The modified artificial leather samples, the proposed HTCAL and artificial leather from the market were laid onto the electric heating machine in sequence for 10 s and their surface temperature was recorded by using an FLIR E60 infrared (IR) camera. This heat supply is like a battery inside an intelligent electronic device (giving off heat) so that the proposed HTCAL acting as an outside packaging or protective cover could be mimicked. Meanwhile, in considering that the heating machine provides a constant heat supply, an experimental time of 10 s was adequate to evaluate the surface temperature differences, because over time, heat balance will be achieved and there will be slight temperature differences among the various samples.

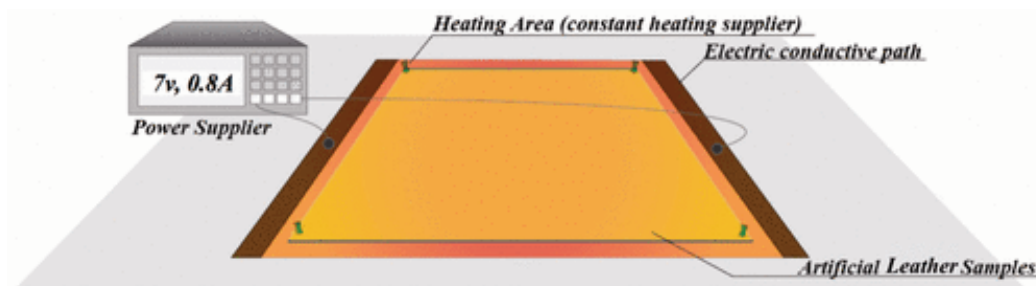


Figure 7. Components of the heating machine that provide a constant heat supply.

Wear trial (temperature sensing)

Besides applications as packaging or protection of intelligent electronic devices, this innovative HTCAL can also be utilized as clothing, especially in terms of the new trend of spring–summer leather wear. Therefore, to simulate real wear circumstances, two wear trials were conducted to compare the differences between the HTCAL and the control samples.

Wear trial I

A symmetrical vest was fabricated in which artificial leather found on the market (same color and similar thickness) and the control leather were used as the upper and bottom left of the vest, respectively, and the modified HTCAL for both the upper and bottom right sides of the vest (Figure 8(a)). The differences among the three different types of leathers could be observed. The vest pattern is a slim fit, based on standard size 160/85A (model size 10). In standard conditions (T : $21 \pm 2^\circ\text{C}$, RH : $65 \pm 5\%$), an anonymous subject wore the vest without any undergarments, so that the vest could come into direct contact with the skin and heat exchange between the vest and subject reached stability after 10 min. IR images were taken to obtain the surface temperature of the three types of leathers.

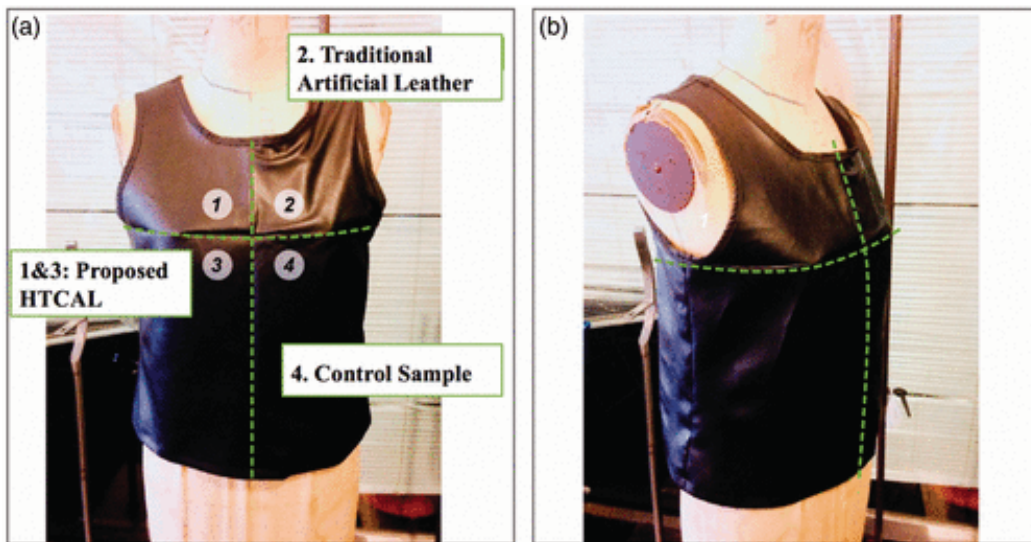


Figure 8. Vest prototype: (a) front view; (b) side view.

Wear trial II

In order to further quantitatively evaluate the differences between warm/cool feelings when wearing a garment produced from HTCAL, artificial leather from the market and the control leather, Wear Trial II was conducted to detect the differences in temperature between the inside of the leather vest and the skin surface. The differences in temperature directly determine the level of heat exchange between the human body and microenvironment; thus, a larger temperature difference will mean a cooler feeling. Eight points, allocated as shown in Figure 9, were selected to detect

the temperature differences during a 10-min wear trial under standard conditions ($T: 21 \pm 2^\circ\text{C}$, $\text{RH}: 65 \pm 5\%$). The wear trials were repeated three times. The temperature and RH at the eight points were recorded every 5 s and average value calculated.

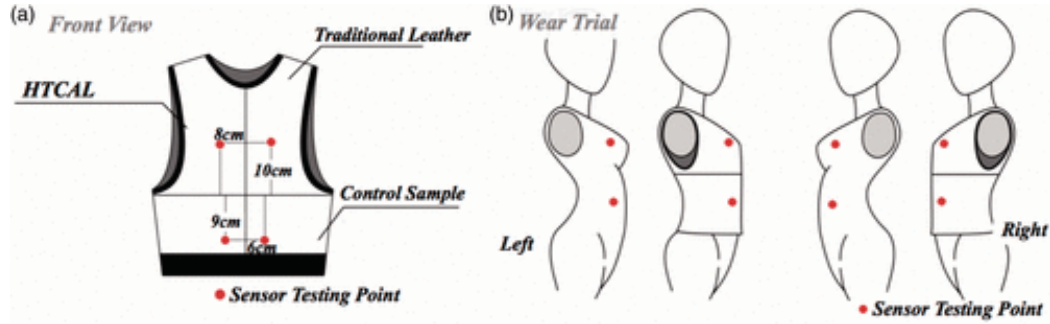


Figure 9. Wear trial II tested points: (a) points on the inside of the vest; (b) points on the subject.

Results and discussion

Resin layer group

The thermal conductivity (k_2) of the polymer resin layer, produced by using the dry method, with the addition of different concentrations of Al_2O_3 (0–21 wt.%), was evaluated. As shown in Figure 10(a), the thermal conductivity value is positively correlated to the concentration of Al_2O_3 , which means that the thermal conductivity of the polymer resin layer could be improved by filling the layer with TCA. With a larger concentration of TCA, more improvement in the thermal conductivity was observed. Meanwhile, compared to the control sample, the k_2 value of the sample with 21% wt.% of Al_2O_3 increased 127.3%. In order to obtain a 50% improvement in the thermal conductivity, more than 12 wt.% of Al_2O_3 should be added.

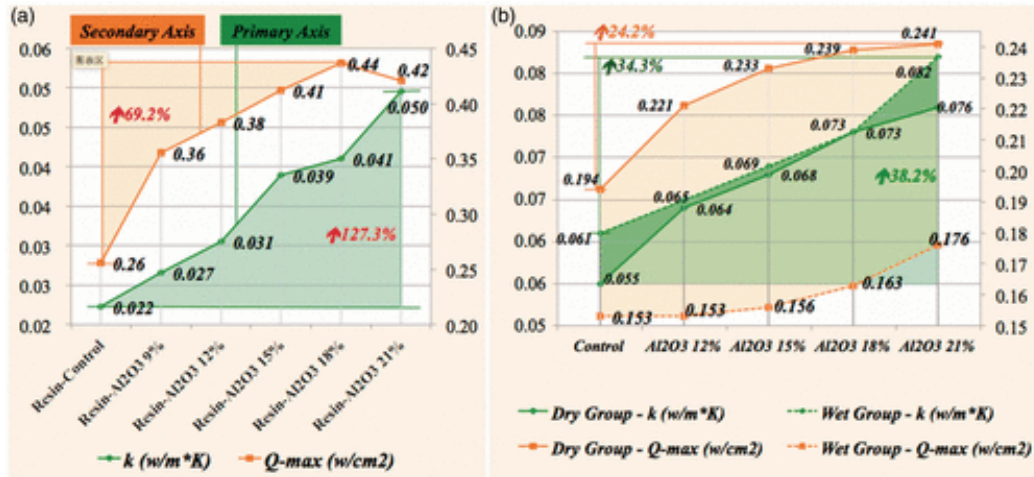


Figure 10. Thermal conductivity (k) and Q -max testing result of (a) resin layers with Al_2O_3 (wt.%) added and (b) artificial leathers with Al_2O_3 (wt.%) added onto their resin layers, produced through dry or wet methods.

In addition, the Q -max values of the samples were also recorded. The relationship between the Q -max value and Al_2O_3 concentration showed a similar trend as the thermal conductivity (k), which means that the resin layers with a higher concentration of Al_2O_3 have a cooler hand feel. The addition of Al_2O_3 forms an intensive network of thermal transfer in the resin polymer and, based on the evaluation result, the feasibility of modifying the approach is successful.

Considering that artificial leathers are composed of similar polymer composites and produced by following similar procedures, this approach can be applied to other developments of thermal conductivity in artificial leather. Consequently, more business opportunities could be derived from the approach.

Groups 1 and 2

Testing of thermal conductivity and Q -max (measurement of cool/warm feeling)

The polymer resin layers with the addition of different concentrations of Al_2O_3 were bonded with the base layer fabric to form artificial leather samples produced with the use of either the dry or wet method, respectively. The thermal conductivity (k) and Q -max value of the modified artificial leather samples (Groups 1 and 2) were tested to evaluate the effects of Al_2O_3 as an additive. From equation (6), the thickness of each

layer is assumed to be constant and modification of any of the layers is expected to improve the performance of the overall leather product

$$k' = D_{\text{tot}} / \left(\frac{D_1}{K_1} + \frac{D_2}{K_2} + \frac{D_3}{K_3} \right) \quad (5)$$

As shown in Figure 10(b), both indexes of Groups 1 and 2 produced with increases in concentrations of Al_2O_3 show a similar tendency as the results of the resin layers. Compared to the blank control sample, the k value of the Group 1 and 2 samples with 21 wt.% of Al_2O_3 increases by 38.3% and 34.3%, respectively. The Q -max value of the same groups of samples with 21 wt.% of Al_2O_3 also increases by 24.2% and 15.0%, respectively. Furthermore, the relationship between the thermal conductivity (k) and the Q -max value with the addition of Al_2O_3 in (a) the single resin layer produced by using the dry method, (b) Group 1 artificial leathers and (c) Group 2 artificial leathers were analyzed (see Figures 11 and 12). Figure 11 shows the excellent linear relationship between thermal conductivity and the added wt% of Al_2O_3 , with an R value approximately equal to 1. This prediction model can be utilized before the production process to ensure that the prototype meets the product requirement based on thermal conductivity and predict its thermal transfer performance at the same time. On the other hand, another forecasting model was used on the Q -max values of the single resin layer and artificial leather made by using different methods (Figure 12). Here, when adding the TCA or Al_2O_3 into the resin layers made by using the dry method, an increase in the Q -max still demonstrated linear growth, with R approximately equal to 0.90. While adding Al_2O_3 into the resin layer of artificial leather made by using the wet method, the Q -max value grew exponentially, with an R approximately equal to 0.92. This means that the addition of a higher wt.% of Al_2O_3 into the resin layer of artificial leather produced by using the wet method will result in faster growth of the Q -max value. Due to these two prediction models, the co-relationship between the heat transfer capability of artificial leather and the wt.% of Al_2O_3 as a TCA has been verified. Both models can be applied towards HTCAL prototype development, when utilization in different fields and with different levels of thermal transfer capabilities are required.

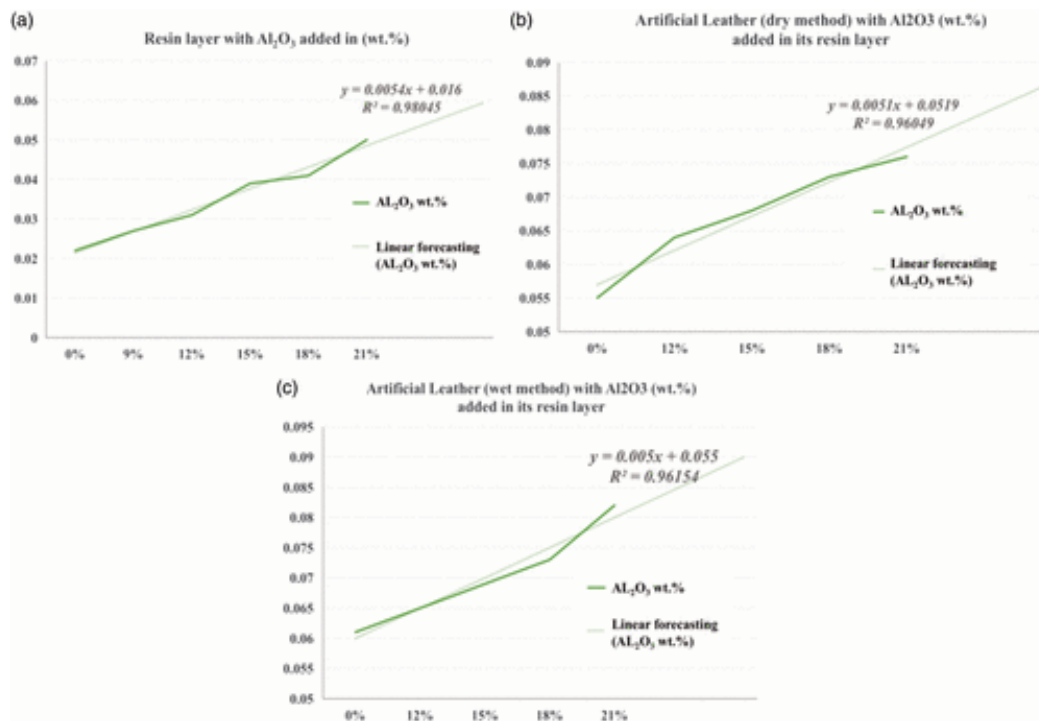


Figure 11. Linear forecasting of the co-relationship between the thermal conductivity (k) and the wt.% of Al_2O_3 added into (a) the resin layer produced by using the dry method, (b) artificial leather produced by using the dry method and (c) artificial leather produced by using wet the method.

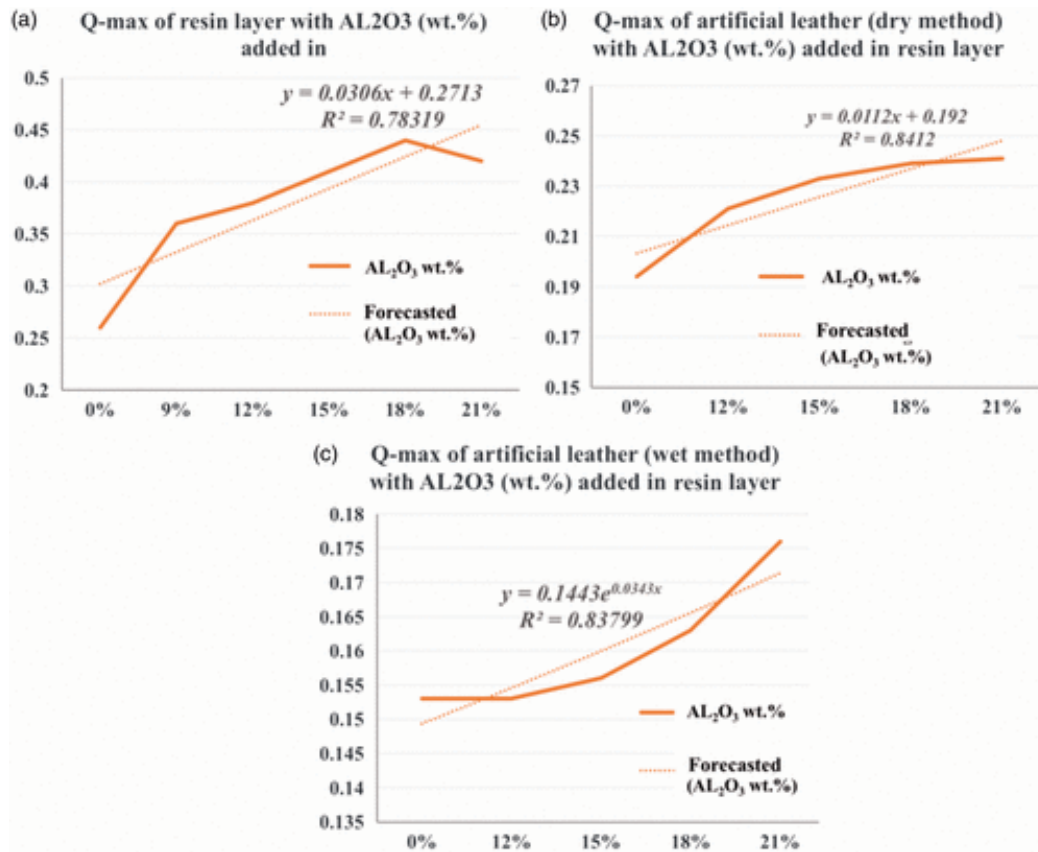


Figure 12. Forecasted trend of the relationship between the Q -max value and wt.% of Al₂O₃ added into (a) the resin layer produced with the dry method, (b) artificial leather produced by using the dry method and (c) artificial leather produced by using the wet method.

In addition, these two different bonding methods produce different thermal performances of the final artificial leather products. Samples prepared by using the wet method have a greater thermal conductivity than those produced by using the dry method, while Q -max testing showed the opposite result. Thus, in order to combine the strengths of these two methods and provide an optimal and innovative product for the market, a soft hand feel and higher thermal conductivity produced by using the wet method and glossy surface appearance and instantly cooler feeling produced by using the dry method are considered to be the key factors of the proposed HTCAL without any trade-offs among them. An industrial prototype with a multilayer structure was developed afterwards by consecutively adopting the wet and dry methods.

Infrared imaging

Figure 13 shows the IR images of artificial leather samples heated on the intelligent electric heating machine after 10 s. According to equation (7) and considering that the heat source (electric heating machine) and the environment temperature were relatively stable, the final surface temperature of the samples that were subjected to the electric heating machine would be proportional to the thermal transfer capability of the samples

$$\Delta T = -q \frac{D_{\text{tot}}}{k'} \quad (6)$$

$$\Delta T \propto \frac{1}{k'}$$

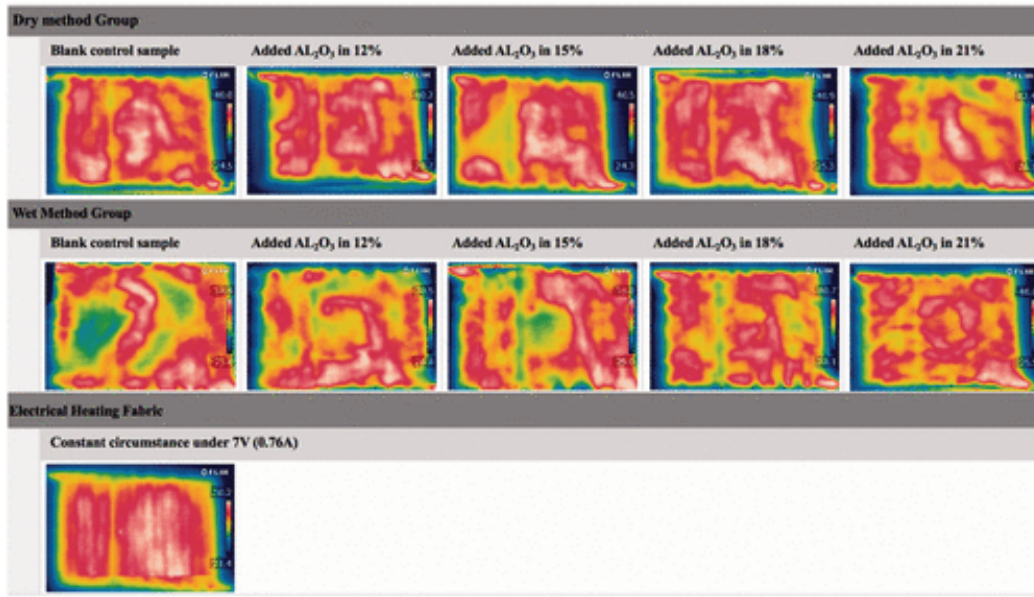


Figure 13. Infrared images recorded as surface thermal images of artificial leather samples on the electric heating machine.

When the thermal conductivity of the artificial leather was increased, the temperature difference between the surface temperature of the electric heating machine and artificial leather would simultaneously decrease. In this paper, the highest surface temperatures T_{max} of the thermal source, the two groups of artificial leather samples, were acquired and are shown in Figure 14. The difference between the highest T_{max} of

the artificial leather sample with the highest concentration of Al_2O_3 in each group and the $T_{\max}$ of the paired control sample reached 2.4°C , which clearly indicates that adding Al_2O_3 as an additive onto the resin layer has an obvious improvement on the thermal transfer of artificial leather. The surface temperature of the artificial leather sample is positively correlated to the concentration of Al_2O_3 , which strongly indicates that the thermal transfer ability of artificial leather could be improved by increasing the thermal conductivity of the polymer resin layer by filling the layer with Al_2O_3 .

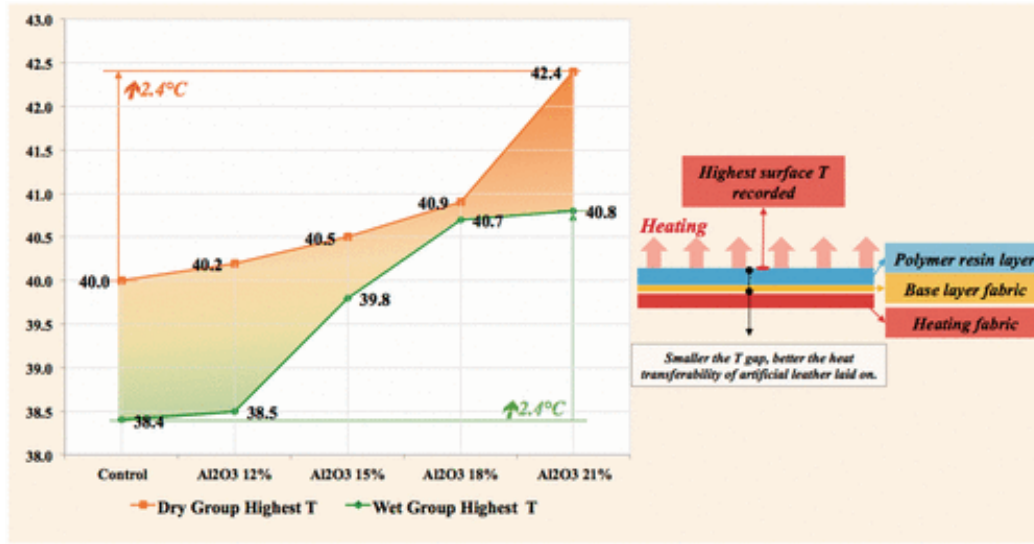


Figure 14. Surface $T_{\max}$ of the two sample groups.

HTCAL prototype versus artificial leather on the market

Testing of thermal conductivity (k) and Q -max (measurement of cool/warm feelings)

The thermal conductivity (k) and Q -max values of the HTCAL and four randomly selected samples were tested and their average value calculated and compared in plots (Figures 15(a) and (b)). In Figure 15(a), the plotted imaginary lines denote the average thermal conductivity level of the existing artificial leathers found on the market, and the benchmark for comparing the performance of the proposed HTCAL to state-of-the-art processes. The thermal transfer of the HTCAL was evaluated and is plotted in Figure 15(b). The performance of the HTCAL was compared to the average slope of the artificial leathers on the market. The thermal conductivity (k) of the

HTCAL is observed to be 48.3% higher, while its Q -max is 29.2% higher than those of the artificial leathers on the market. Here, a significant improvement is already evident, and in order to mimic the real circumstances of applying this prototype into product design, such as phone cases and summer wear, evaluation through IR imaging and wear trials were conducted as well.

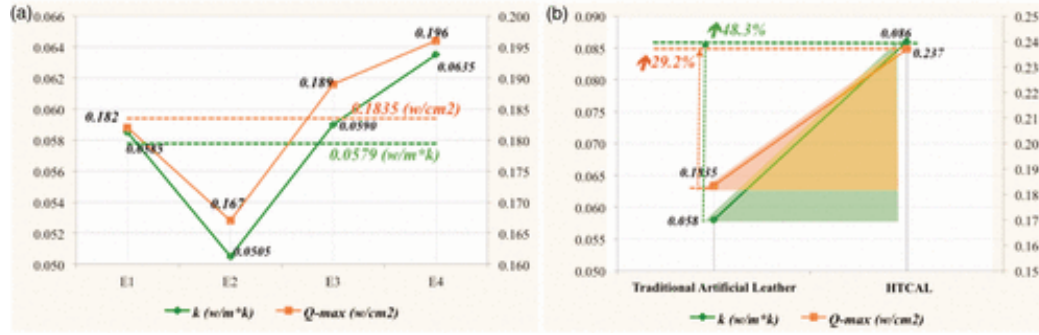


Figure 15. Thermal conductivity (k) and Q -max test results of (a) existing artificial leather on the market and (b) the proposed HTCAL.

Infrared imaging

By using the IR imaging testing system as mentioned earlier, a comparison between the proposed HTCAL and control sample was conducted. The thermal transfer of the control sample is maintained around the average slope and it is in the same color as the HTCAL to eliminate the heat-absorbing influence from various colors. This time, the electric heating machine was set to reach a constant surface temperature under a voltage of 10 V or a current of 1.09 A after 30 min to provide a higher heating temperature at the bottom so that the difference between these two samples could be increased and made more obvious. The stabilizing of the surface temperature of the electric heating machine after 30 min is shown in Figure 16(a). Figures 16(b) and (c) show the surface temperatures of HTCAL and the artificial leather from the market, respectively, at different points on their surface. The highest surface temperature of the HTCAL sample is 46.8°C, while that of the control sample is 43°C. In comparing the different heated parts of these two leather samples, the temperature difference in the same place ranges from 3°C to 5°C. In calculating the temperature differences of the five points illustrated in Figures 16(b) and (c), it was found that the average surface temperature of the HTCAL sample at any selected point is 4.6°C

higher than that of the control sample. This demonstrates that in practical applications, the proposed HTCAL has a better capability of transferring heat flow from one side to the other and the process is faster; thus, the improvement of its heat transfer performance is significant. This positive test result indicates that the proposed prototype could be applied as outside packaging for intelligent electronic devices. Furthermore, the proposed modification is based on current production technology, which could be quickly and directly applied in real mass production without any extra costs for the newly added benefits.

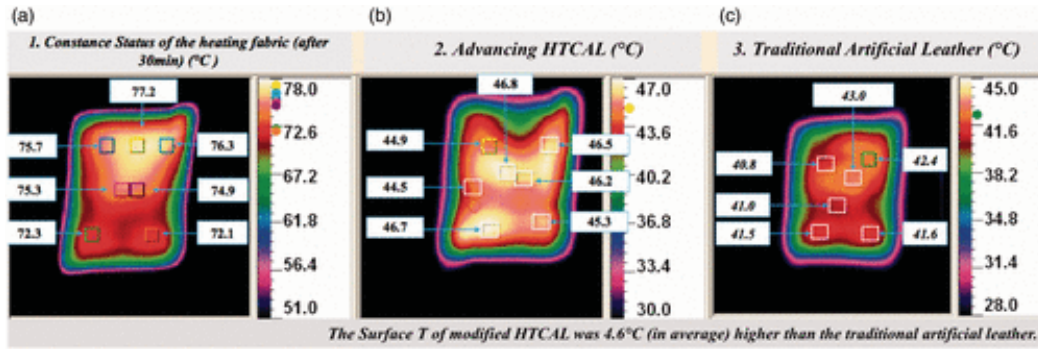


Figure 16. Recorded surface temperature record of images: (a) electric heating machine after 30 min (10 V, 1.09A); (b) HTCAL placed onto the electric heating machine for 10 s; (c) artificial leather from the market placed onto the electric heating machine for 10 s.

Wear trial

Aside from the IR image testing to simulate real applications, two wear trials were also conducted to quantify the warm-cool feeling and evaluate the performance of the HTCAL sample when worn on subjects (Figure 17(a)).

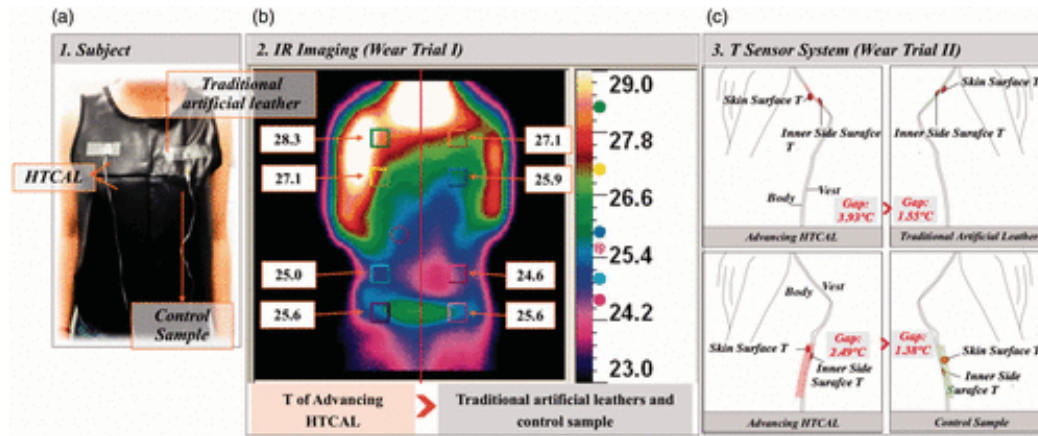


Figure 17. Wear trial test results: (a) vest worn by subject; (b) infrared recording; (c) temperature difference between the skin and inside of the vest.

Wear trial I

The testing procedures of the wear trials mentioned earlier were followed. Figure 17(b) shows the results of wear trial I. From the HTCAL sample (left of the figure) and artificial leather from the market (upper right of the figure, from which it can be observed that the thermal conductivity is similar to the previously established benchmark) and control sample (bottom right), it can be easily observed that the average surface temperature of the HTCAL sample is greater than that of the artificial leather and control samples, thus indicating that the HTCAL sample, with the addition of 21 wt.% of Al_2O_3 , has a significantly better thermal transfer performance. Thus, the HTCAL in this study is suitable for use in spring–summer leatherwear because it provides better thermal comfort.

Wear trial II

Another wear trial was designed to quantitatively evaluate the warm/cool feeling and wear comfort of HTCAL. The sensor allocations and their average recorded temperatures are shown in Table 6 and Figure 17(c). The temperature difference between the skin and the inside of the HTCAL sample is always greater than that of the artificial leather sample from the market and the control sample, which indicates that when the proposed HTCAL is used in a garment, the wearer will feel cooler. The proposed modification to derive the HTCAL is therefore considered effective.

Sensor allocation	Skin surface T (left)	T inside vest (left)	Skin surface T (right)	T inside vest (right)
Top	Sensor 1 (left)	Sensor 3 (left)	Sensor 2 (right)	Sensor 4 (right)
Bottom	Sensor 5 (left)	Sensor 7 (left)	Sensor 6 (right)	Sensor 8 (right)
	Upper part		Bottom part	
Average T (°C)	HTCAL (T difference = Sensor 1 – Sensor 3)	Normal artificial leather (T difference = Sensor 2 – Sensor 4)	HTCAL (T difference = Sensor 5 – Sensor 7)	Control Sample (T difference = Sensor 6 – Sensor 8)
Skin	30.33	29.54	29.21	28.99
Inside vest	26.40	27.99	26.72	27.61
Difference	3.93	1.55	6.12	1.38

Table 6. Average values of skin surface temperature and temperature inside vest

Conclusions

The demands of the market and leather manufacturers in terms of current social trends have been addressed in this study, in which an innovative type of artificial leather that has a high thermal conductivity has been developed. The use of an additive that has a high thermal conductivity, that is, Al_2O_3 , as part of the production of this new artificial leather along with both the wet and dry methods consecutively to form the proposed multilayer structure meant that the strengths of different production methods are placed into the final prototype. Resin Layer 1, which is the middle layer, is laminated onto the fabric base layer by using the wet method with a high concentration of Al_2O_3 (21 wt.%) as the main component. Resin Layer 2 is placed on the top, which is produced by using the dry method and also with the addition of 21 wt.% of Al_2O_3 , and coated on as the surface. Thus, the final HTCAL sample has a high thermal conductivity (k), a smooth surface appearance, soft hand feel and instant cool feeling.

Through various testing for evaluation purposes, the contribution of the addition of Al_2O_3 to the resin layer on the improvement of the performance of the overall product has been confirmed. With reference to the testing result, the improvement is found to be significant. The thermal conductivity (k) of the proposed HTCAL is 48.3% greater than that of the artificial leather found on the market. When applied in wear trials, the higher surface temperature of the artificial leather samples with Al_2O_3 and larger temperature gap between the HTCAL and the skin indicate the better heat transfer capacity of HTCAL compared to the artificial leather found on the market.

Furthermore, in order to facilitate the transfer of this technology from the laboratory to mass production in the industry, a production process that utilizes the wet and dry methods consecutively is adopted to produce an optimal prototype. No extra equipment or technology transfer costs are required, thus demonstrating that the proposed technology can be directly and immediately adopted. This innovative HTCAL can be applied not only in the development of spring–summer clothing and footwear, but also for accessories for electronic devices, in the interior of cars, for furniture, etc. There is a large potential market with substantial profit to be made. At the end of this research work, an iPhone case was fabricated by using the HTCAL as an example of the innovativeness of this material (Figure 18).

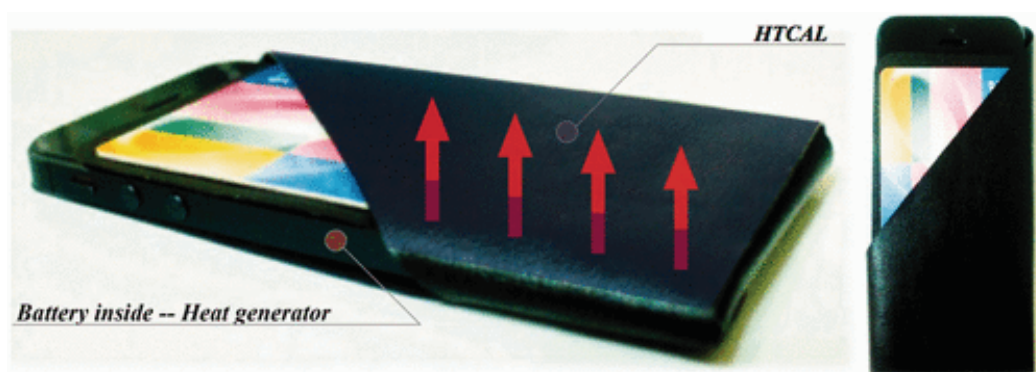


Figure 18. iPhone case prototype made of HTCAL.

Future work

There is still room for further progress, such as utilizing a coupling agent to enhance the bonding between each layer to further improve the heat transfer. Due to the addition of additives, the adhesive force between the base and resin layers might be affected. Thus, further research could be conducted to investigate the amount of influence, and a possible solution to address the trade-off between adding a TCA and the bonding force between the layers and developing an optimal final product. Meanwhile, according to the different purposes of the product design, different concentrations of TCA added can be amended to achieve better performance. Besides, other consumer products, such as clothing and footwear, can be produced by using the HTCAL for additional market opportunities.

Declaration of conflicting interests

The authors declared no potential conflicts of interest with respect to the research, authorship and/or publication of this article.

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