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Evaluation of Peppermint Based Menthol as a Cooling Agent for Development of Hydrogel Cooling Patch

Mohamad Syahir Hasnadi[1], Aliff Hisyam A. Razak[1]*

[1]Department of Chemical Engineering Technology, Faculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia, 84600 Pagoh, Johor, MALAYSIA

*Corresponding Author Designation

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Abstract: Peppermint as a cooling agent helps to give cooling and fresh sensation and specific sensory toward human's skin. A cooling agent also has its potential to helps tone down the heat from someone with fever. It is related with menthol that gives the cooling sensation. Menthol are derived from peppermint because it has a high menthol content. In this project, the combination of hydrogel which consist of polyvinyl alcohol, hydroxypropyl methylcellulose, polyethylene glycol and peppermint oil were used for the formulation of hydrogel cooling patch. The extraction of menthol from peppermint was using Soxhlet extraction process towards 1000 g of dried peppermint leaves to produce peppermint oil. The formulation of hydrogel was evaluated for its adhesiveness property via lap shear test and its cooling sensation property via sensory evaluation test. Sample 6 has a good adhesive property and it was very easy to apply the patch to apply on skin. The cooling sensation also was good and the sample scored a high mark during sensory evaluation test. As the conclusion, the hydrogel formulation of sample 6 gives the best result as a cooling patch.

Keywords: Peppermint, Fever, Menthol, Hydrogel, Polyvinyl Alcohol, Hydroxypropyl Methylcellulose, Polyethylene Glycol, Soxhlet Extraction, Peppermint Oil, Cooling Patch

1. Introduction

Fever is sometimes called pyrexia, because of changes to the body's thermal regulation set point, the temporary temperature rises over the typical range of 36.5 - 37.5 ºC [1]. It is a typical naturally occurring bodily reaction to physiologic stress, such as ovulation, vigorous exercise, or mental stress, microorganism infection, or the associated inflammation or production of pyrogenic materials such as leukaemia. It alerts individuals that they are unwell to see the doctor. One of the alternatives that can be used to treat fever other than taking antipyretic drug such as paracetamol, cooling patch can also be used as a way to treat fever.

*Corresponding author: aliff@uthm.edu.my

Over the years, hydrogels have been characterized in many ways by researchers. To describe more accurately the polymeric cross-linked network structures, scientist employ two interchangeable terms which are gels and hydrogels [2]. A hydrogel is a hydrophilic (3-D) hydro pylon network which, because to the chemical or physical interconnection of individual polymer chains, may expand in water to store enormous amounts of water. It is firstly reported in 1960 [3]. A second definition is the polymeric substance which is capable of swelling, but not dissolving in the water, and preserves a large portion of the water inside its structure[4].

Menthol is a $C_{10}H_{20}O$ terpenoid alcohol (mol wt. 156.27) with three chiral centres leading to eight possible stereoisomers (4 enantiomeric pairs) [5]. A wide selection of goods ranging from common cold to toothpastes, candies and insecticides include menthol and similar cooling substances [6]. Menthol was once seen as a flavouring ingredient or an odour that increased medicinal and confectionary, but a variety of biological action on sensory neurons and smooth muscles was found in pharmacological investigations.

A plant from the Labiatae family, Mentha piperita L. or peppermint with vernacular names is traditionally employed as an antiseptically, carminative stimulant or as a flavouring ingredient in the cosmetics and medicines industry across the world [7]. Peppermint is a widely used herb used in many ways (i.e., oil, leaf, leaf extract, and leaf water)

2. Materials and Methods

Main substances needed in producing formulation of hydrogel are, peppermint leaves, polyvinyl alcohol, hydroxypropyl methylcellulose, polyethylene glycol, silicone elastomer and heptane. The apparatus/equipment needed are chiller, drying oven, dry blender, weighing machine, film applicator, hot tack tester, magnetic stirrer, rotary evaporator, Soxhlet extractor and speed mixer.

2.1 Extraction of Menthol from Peppermint Leaves

Soxhlet extractor was used for the extraction of menthol from peppermint leaves. Fifty grams of sample was placed in the teabag in the Soxhlet extractor. The solvent used for the extraction is distillation, where 240 ml of distilled water was placed in the bottom flasks of the Soxhlet extractor. The extraction process was performed for around 6 hours, where the sample was heated at around 100.0 °C.

The yield of extraction of essential achieved by using Soxhlet extraction was determined by using the equation 1 below [8], where the results were indicated as a percentage (%):

$$Y = \frac{mass\ of\ essential\ oil\ obtained}{mass\ of\ peppermint\ sample} x100 \qquad Eq.1$$

2.2 Preparation of hydrogel

Before preparing hydrogel, Design-Expert software was used to determine the number of samples needed to be ready for this experiment. Box-Behnken Design is chosen as the design. There are three factors selected for this experiment: the amount of PVA, the amount of HPMC, and the speed level of the mixing. Table 1 below shows number of samples needed to be prepared.

Table 1: Design of Experiment (DOE) of preparing hydrogel samples from PVA and HPMC

Sample	Amount of Polyvinyl Alcohol, PVA (g)	Amount of Hydroxypropyl Methylcellulose, HPMC (g)	Stirrer speed level
1	0.8	2.4	10
2	4	4	6.5
3	2.4	2.4	6.5
4	2.4	2.4	6.5
5	2.4	2.4	6.5

6	0.8	4	6.5
7	2.4	2.4	6.5
8	2.4	4	10
9	2.4	0.8	10
10	4	2.4	3
11	2.4	2.4	6.5
12	4	2.4	10
13	2.4	4	3
14	2.4	0.8	3
15	0.8	0.8	6.5
16	0.8	2.4	3
17	4	0.8	6.5

Polyvinyl Alcohol (PVA) (Mw=89000 g/mol, Sigma-Aldrich) and Hydroxypropyl methylcellulose (HPMC) (Mw=1261.4 g/mol, DChemie) solution was prepared first. First, at room temperature, 0.8 g of PVA was dissolved in 25 ml of distilled water. Next, at room temperature, 0.8 g HPMC was dissolved in 25 in distilled water. And then, both PVA and HPMC solutions were mixed. Meanwhile, 1 ml of peppermint oil was mixed with 0.5 g of Polyethylene Glycol (MW=380-420 g/mol, EvaChem). The mixtures were then stirred together using a magnetic stirrer until the solution was homogenized. The mixture was then poured into a round container with a diameter of 145 mm, and the solution was then allowed to cure at room temperature for 48 h. After 48 h, the hydrogel was appropriately stored in a chiller before being characterized.

2.3 Preparation of Silicone Film as the Outer Film

The ratio used for the silicone film preparation is 1:1:3, where one part of premix A, one part of premix B and three parts of heptane were mixed in a speed mixer at a speed of 1350 rpm. The silicone mixture was let to mix continuously for around 1 hour. Once the silicone mixture is homogenized, it is then poured onto a film applicator, where the mixture is let to run through the film applicator. The mixture is then let to cure for 2 hours at 40.0 °C, then the temperature was raised to 110.0 °C for 15 minutes.

2.4 Lap Shear Test

By following the method modified from ASTM F2255 [9], the hydrogel samples' adhesive strength was measured by using a hot tack tester machine. First, three hydrogel samples were chosen, which are sample 6, sample 8, and sample 11. Next, sections of hard plastic used as the layering to hold the hydrogel were cut into a long rectangular shape. The hydrogel samples were 2.5 cm x 1cm in size, then placed in between the plastic. Once the samples have been set up at the testing area where the plastic is clamped on a fixed grip, the plastic component is pulled apart by the movement of the grip in which the pull speed was 100 mm/s.

2.5 Sensory Evaluation Test

Twenty respondents aged twenty to forty years old were evaluated using the question form provided via Google Form. A cooling patch with different compositions of hydrogels containing menthol from peppermint oil will be evaluated. Around 4 g of each sample were placed on the respondents' hands to assess. The questions were divided into four sections.

3. Results and Discussion

3.1 Lap Shear Test

There were three samples tested for the characterization of the hydrogel in respect to its adhesiveness property, which are sample 6, sample 8 and sample 11. By using the data obtained from the test, a force-time graph can be plotted as shown in Figure 1(a), 1(b) and 1(c).

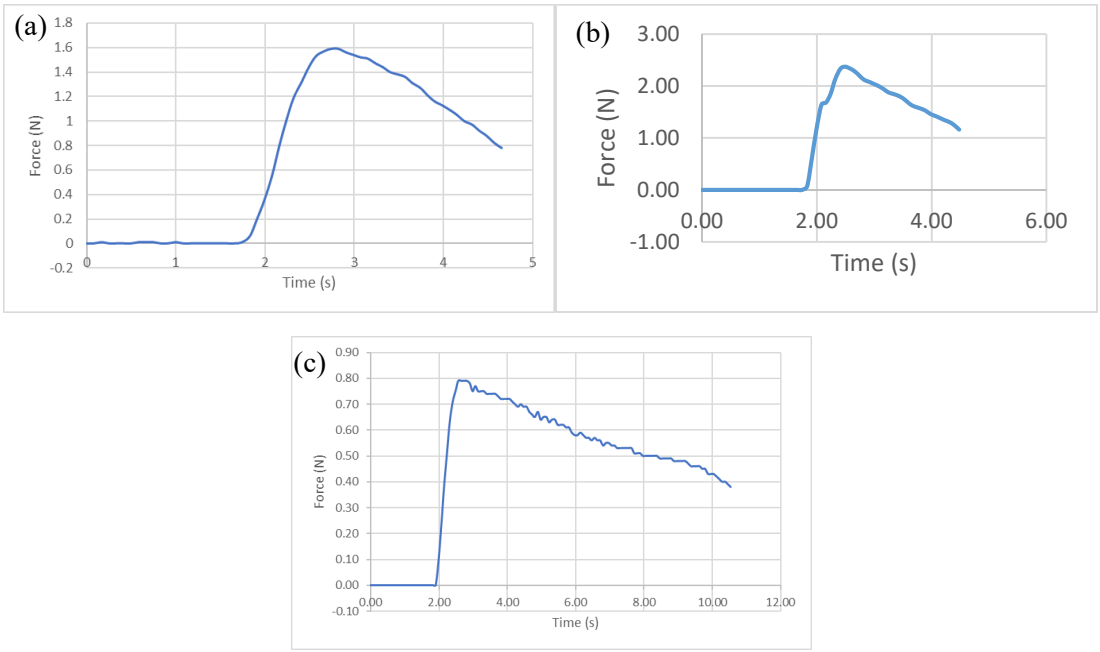

Figure 1: (a) Graph of Force (N) vs Time (s) for sample 6; (b) Graph of Force (N) vs Time (s) for sample 8; (c) Graph of Force (N) vs Time (s) for sample 11

Using Eq. 2, the lap shear strength of the sample can be determined. The lap shear strength data of every sample is then tabulated into Table 2.

$$\tau = \frac{F_{max}}{l_o \cdot w} \qquad Eq.2$$

Table 2: Lap shear strength for every sample tested

Sample	Lap shear strength (kPa)
Sample 6	6.36
Sample 8	9.48
Sample 11	3.16

A high lap shear strength value indicates that a higher force is required to break the bond between the hydrogel sample and the surface it sticks to, while a low shear strength value that a lower force is required to break the bond [10]. Therefore, a high value of lap shear strength is favourable for developing the hydrogel so that the sample does not peel off from the skin easily.

3.2 Sensory Evaluation Test

17 samples were given to all 20 respondents for them to evaluate. For section B of the sensory test which is the hydrogel evaluation, the average score of cooling effect of the hydrogel and the patch adhesion of every sample are as shown in Table 3 below:

Table 3: Average score of hydrogel cooling effect and hydrogel adhesiveness on skin

Sample	Cooling effect of hydrogel	Patch adhesion of hydrogel
1	3.65	2.8
2	3.55	2.55
3	3.65	2.7
4	3.55	2.5
5	3.8	2.65
6	4.2	4.6
7	3.6	3.2
8	3.25	3.45
9	3.35	3
10	3.4	2.85
11	3.5	3.45
12	3.55	3
13	3.35	3.1
14	3.2	3.15
15	3.05	3.4
16	3.25	3.3
17	2.9	3.2
Average	3.46	3.11

From the average score shown, it can be said the hydrogel sample gives cooling effect when applied onto human skin. The average score of cooling effect for all sample is 3.46, which indicates that the cooling effect from the sample is in between moderately cool and cool. It also can be said that the adhesiveness of the hydrogel is also quite good with an average score of 3.11 for all samples, thus sitting in between moderately adhere and adhere.

Based on Table 3, it is shown that sample 6 has the highest average score among all samples where it scores 4.2 and 4.6 on the cooling effect and the adhesiveness, respectively. Meanwhile, sample 17 score the lowest average among all samples where it score scores 2.9 and 3.2 on the cooling effect and the adhesiveness, respectively. Thus, it can be said that sample 6 is the best sample among of all other samples.

The next section of the sensory evaluation test, mainly focused on the properties of the silicone outer film for the cooling patch product. Based on Figure 2, it can be said that all respondents agree that the character of the silicone outer film is non-sticky, the thickness of the silicone outer film is acceptable and also colour of the silicone film is transparent.

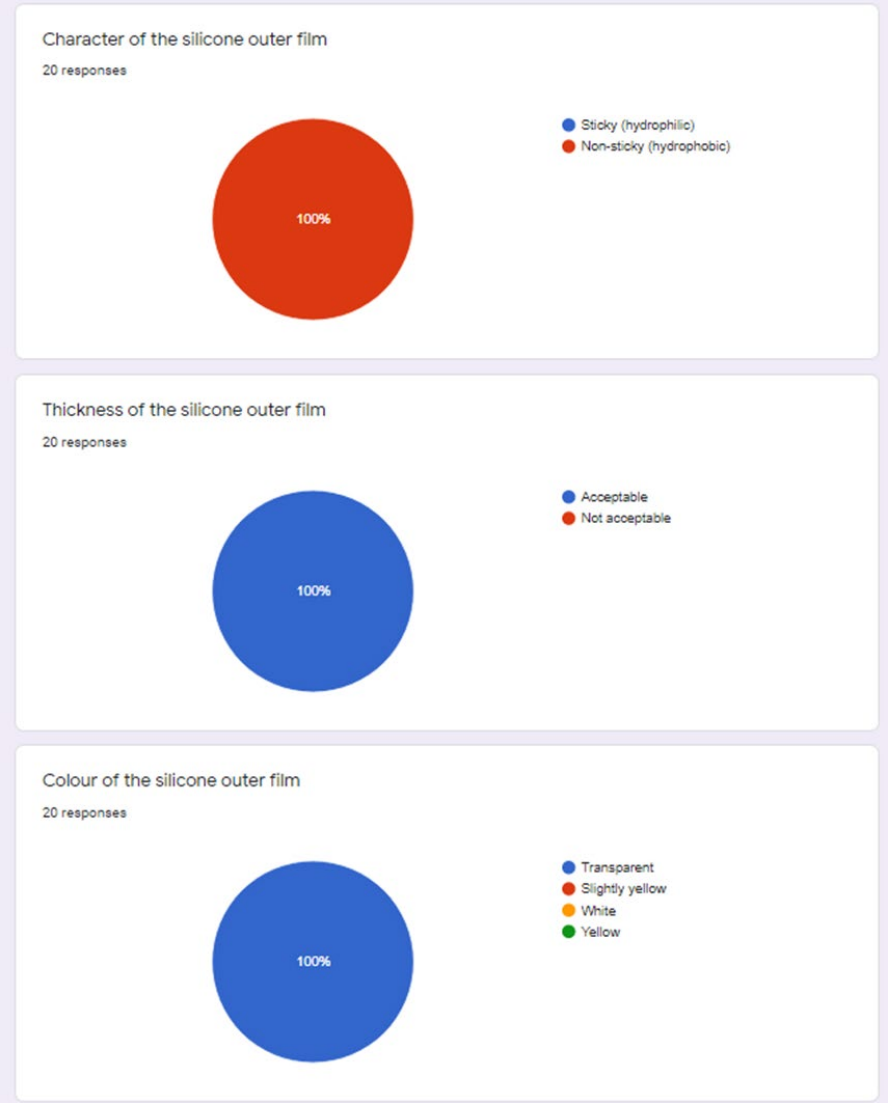

Figure 2: Responses on the properties of the silicone outer film

For the last section of the sensory evaluation, it mainly focused on the overall product in term of its appearance, stickiness and also the comforts while wearing the patch. Based on Figure 3, it can be said that the most respondents' comfort while wearing the patch on the skin are comfortable, most of the respondents also said that the product can stick well on their skin and most of the respondents agree that the appearance of the product is appealing.

Figure 3: Responses on the evaluation of the overall product

4. Conclusion

The findings of this study have shown the effectiveness of peppermint as the cooling agent for the development of hydrogel cooling patch. According to the findings of this study, it was proven to completely achieve the objectives stated. Peppermint oil acting as a cooling agent in hydrogel cooling patch was able exhibit cooling sensation on the human skin. Also, this product can also be labelled as natural product since the cooling agent used is from nature compared to typical cooling patch on market which usually used chemically-synthesised menthol as the cooling agent. It can be said that this study's objectives were met with success, where menthol was successfully extracted from peppermint leaves in the form of peppermint oil via Soxhlet extraction with the help of distilled water as the solvent, in which the yield of extraction was 3.1 %, hydrogel cooling patch containing menthol from peppermint leaves was successfully made and the hydrogel cooling patch containing menthol from peppermint leaves was successfully characterised, where the hydrogel successfully exhibit its adhesiveness property via lap shear test, and the hydrogel also successfully exhibit its cooling sensation when tested via sensory evaluation test.

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