様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科研費 KAKENHI

科学研究費助成事業　　研究成果報告書

令和　４年　６月２９日現在

機関番号：１１３０１
研究種目：基盤研究(C)（一般）
研究期間：2019～2021
課題番号：１９Ｋ０４２３０

研究課題名（和文）Making cold from natural rubber: a low-cost and environment friendly refrigeration alternative

研究課題名（英文）Making cold from natural rubber: a low-cost and environment friendly refrigeration alternative

研究代表者
ＳＥＢＡＬＤ　ＧＡＥＬ（SEBALD, Gael）

東北大学・高等研究機構等・客員教授

研究者番号：１０７９２１６１

交付決定額（研究期間全体）：（直接経費）　3,200,000円

研究成果の概要（和文）：本研究では，弾性熱量効果を用いた冷凍機構の開発に向けて天然ゴムに焦点をあて，材料としての利用の可能性についてその性能評価を行った．その結果，天然ゴムは他の弾性体物質に比して拡張および収縮度合いから大きな温度差を実現でき，かつ再利用が十分に可能であることを明らかにした．また，発熱体としての天然ゴムから作動流体への伝熱機構のモデル化，および実験の評価手法の確立も図り，天然ゴムの拡張/伸縮と流動の位相差と流量が伝熱機構に大きな影響を及ぼすことを明らかにした．天然ゴムの弾性熱量効果を用いることで，温度差を最大7℃まで上げることに成功し，2Wの冷却能力を証明した．

研究成果の学術的意義や社会的意義
Current refrigeration systems uses refrigerant gases based on a tradeoff between environmental impact, toxicity, and performance. Alternatively, refrigeration using caloric materials may change this paradigm. Particularly, natural rubber is abundant, cheap, and environmentally friendly.

研究成果の概要（英文）：In order to make a refrigeration device from a so-called caloric material like natural rubber, it is necessary to convert time variations of temperature into a spatial gradient. To do so, the project focused on regenerative systems, where a fluid moves cyclically in contact to the rubber when it is cyclically stretched and unstretched. The complex heat and mass transport was solved analytical, leading to figures of merit from materials properties and geometry. The nature of the flow plays an important role, and the impact of the frequency and velocity was assessed. The importance of the heat transfer between the fluid and the rubber was quantified, and the low thermal conductivity impact on the performance was discussed. Finally, a functional heat pump prototype was developed which proved that natural rubber is a valuable candidate for next generation refrigeration systems. A prototype of 10g of natural rubber showed a temperature span of 7°C and a cooling power of 2W, with a COP>5.

研究分野：Solid-test cooling

キーワード：refrigeration　rubber　elastomer　regenerative

様式Ｃ－１９、Ｆ－１９－１、Ｚ－１９（共通）

1　研究開始当初の背景

Alternatives to conventional refrigeration technologies is an important issue for (i) increased efficiency and lower primary energy consumption, and (ii) for minimizing environmental impact for fabricating the device and of its components (*1*). The current generation of refrigerant gases is based on a trade-off between performance, environmental impact (ozone depletion and CO2 equivalent impact), and safety (flammability). In parallel to the search to the 4th generation of refrigerant gases, alternative technologies using so-called caloric materials might address some of the applications of refrigeration. As an example, several large scale magnetocaloric devices have been developed in recent years, with high power prototypes (kW range and more)(*2*)(*3*).

In the framework of our project, we focused on the use of natural rubber as a refrigerant, and checked the specificities of the polymer material as an active material. Natural rubber offers several key advantages compared to other metallic or ceramic compounds used in magnetocaloric, electrocaloric and elastocaloric devices. It is highly abundant, low cost, and recycling routes were recently proposed(*4*).

2　研究の目的

The objective of this project is was develop a proof of concept of refrigerating device based on natural rubber (using its phase transition under stress).

Key scientific question: How to convert time variations of temperature into spatial gradients in regenerative systems in an efficient manner?

Objectives list:

- develop **theoretical and experimental approaches** of elastocaloric cooling devices,
- determine **the key properties** of elastocaloric polymers
- assess the **cooling device performances**

3　研究の方法

Natural rubber exhibits intrinsically a very low thermal conductivity, inducing open questions regarding its applicability into a refrigeration device where quick heat exchange is required between the active material and surrounding solids and fluids. Moreover, the fluid flow control is a key point for maximizing the heat transfer between rubber and fluid.

To address this question, in the case of coaxial flow, several actions were achieved:

1. Model of the refrigeration device, and assessment of the importance of the material's properties for the device performances
2. Numerical model were also considered for assessing the fluid flow impact on the refrigeration performance
3. Experimental proof of concept was designed, fabricated and tested.

4　研究成果

Prior to the design of a refrigeration device using natural rubber, it was chosen to develop an analytical model for the regenerative cooling type system. It consisted in an assembly of rubber sheets cyclically deformed, with a fluid moving cyclically between rubber sheets. The model allowed the test of several working conditions such as frequency, fluid displacement amplitude, and phase delay between rubber actuation and fluid displacement. It was obtained guidelines for the device development, notably in terms of natural rubber wall thickness for a proper heat transfer between the working fluid (water) and rubber sheets.

In harmonic regime, a thermal boundary layer δ was defined as

$$\delta = \sqrt{\frac{2K}{\omega\rho c}} \qquad \text{Eq. (1)}$$

where K, ω, ρ and c are the thermal conductivity of rubber, the angular frequency, the density and the heat capacity of rubber respectively.

The best performances of the system are obtained when the rubber wall thickness is lower than the thermal boundary layer thickness. In fact, for a frequency of 0.1Hz and the properties of rubber, it was found that δ~600µm, which was larger than the wall thickness of rubber of 500µm.
The model was compared with a preliminary experiment consisting of a single rubber tube. The experimental setup is shown in Figure 1, and the comparison in Figure 2.
The model is now fully functional, and may find applications not only for rubber based elastocaloric devices, but also for any other caloric coupling, such as magnetocaloric or electrocaloric coupling. As a perspective, it will be attempted to set collaborations with experts in these domains, in order to test the model limits on more geometries and materials properties. In addition, it can be used to assess the ultimate performances of a given system. The model constitutes a rare attempt of full analytical model but able to take into account temperature gradient on the refrigeration device as well as a full solving of the heat transfer equation across the fluid and active material thickness (*5*). Moreover, such modelling allowing the derivation of figures of merit based on geometry and materials' properties and play a key role in the devices design (*6*).

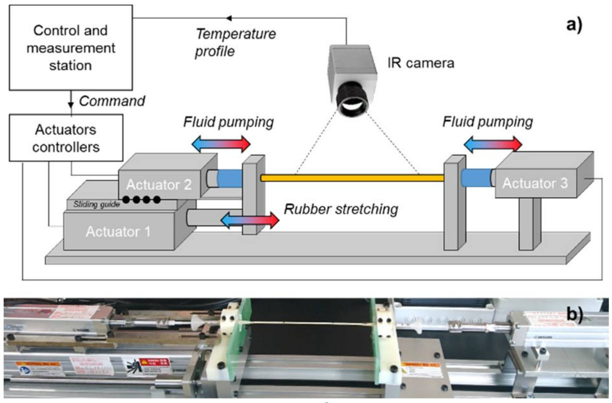

Figure 1 : Experimental setup for testing a single natural rubber tube heat pump. After (7).

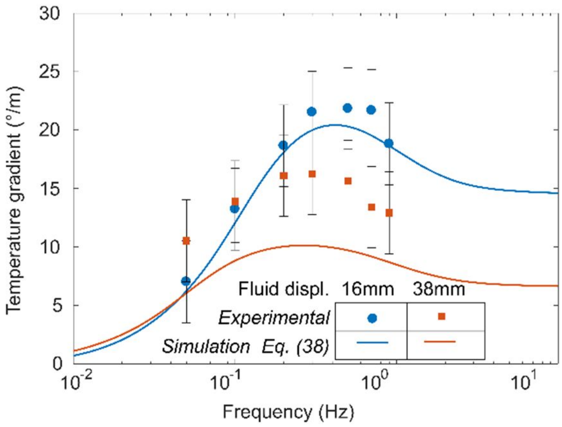

Figure 2: Experiment and simulation of the effect of frequency on the temperature gradient of the elastocaloric cooling system, for two fluid displacement amplitudes.

In addition to the analytical model, a 1D model was also developed for coaxial flows. It was revealed that the motion of the fluid combine with the elastocaloric activity of the natural rubber led not only to the heat pump function, but also to an active thermal conductivity that hinders the establishment of thee temperature span over the device. 1D model showed however strong limits in the determination of thermal active conductivity, as it required to consider the thermal gradients within the fluid channel, which would necessitate 2D or 3D models, which was out of the scope of the project.
As shown with the preliminary experiments and the analytical model, the impact of fluid displacement, frequency and phase delay was determined. As shown in Figure 2, a larger fluid displacement leads to an increase of the temperature gradient and cooling power until a limit, where the active thermal conductivity becomes predominant.
The use of flow diverters requires however further work. At the modelling level, the CFD models were yet not able to consider non coaxial flow. In terms of experiments, the practical mounting of the flow diverters – as additional solid objects – requires also further work to find technical solutions of realization and solve the proper collection of the fluid into reservoirs.
In order to maximize the heat exchange, it was foreseen alternatively that the part of the tube in connection with the pumping system allowed a diameter change with the position. Therefore, the fluid entry effect is accompanied with radial flow velocity which was thought to be beneficial. Moreover, this rubber tube part is subjected to higher stress levels, and the elastocaloric activity was investigated near the edges of the tube.

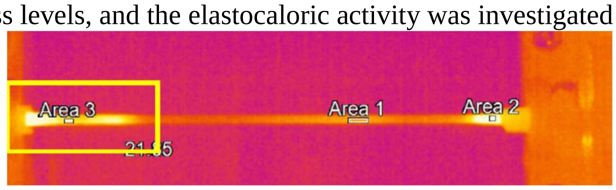

Figure 3 : Edge effect within an elastocaloric rubber tube: temperature rise when stretching a rubber tube, with an increased effect near the ends of the tube

The elastocaloric activity is much higher near the edges of the rubber tube. It was then attempted to add hard tubes inside the rubber in order to have a succession of edges effects. It was successful, but led to an accelerated degradation of the rubber tube after a few hundreds of cycles, the implementation into a prototype was not possible.

Finally, in the prototype development, the fluid flow was laminar over almost all the rubber tube, radial fluid flow existed only near the edges, as a tradeoff between fatigue life and higher heat flux flow.

In the light of the preliminary single tube tests, the very first prototype of a refrigeration device using natural rubber tubes was successfully fabricated and tested. It consisted in 55 parallel tubes connected to fluid reservoirs. The geometry was optimized for ensuring a homogenous distribution of the fluid flow in each tube, as it is known to be a critical parameter (*8*).

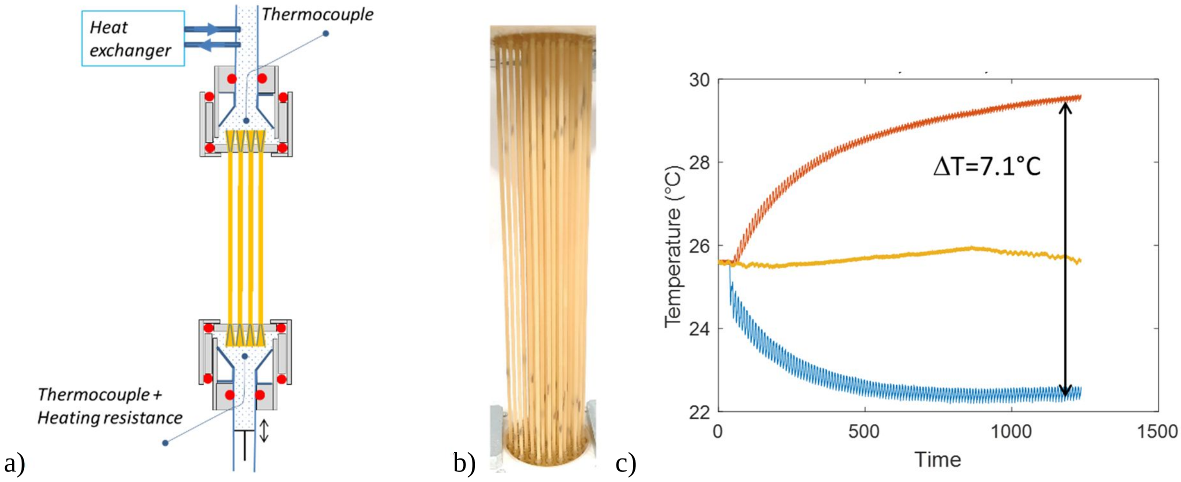

Figure 4 : a) schematic of the prototype, b) picture of the assembly of natural rubber tubes, and c) temperature span generation.

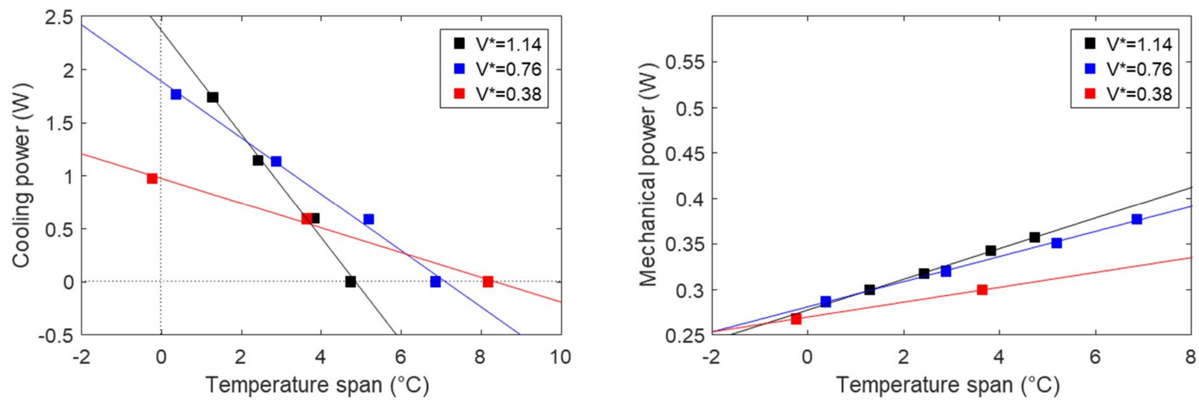

Figure 5 : Natural rubber heat pump performance (left) cooling power vs. temperature span and (right) mechanical work required as a function of the temperature span.

The schematic of the device is shown in Figure 4a, the picture of the rubber tubes assembly in Figure 4b, and the no-load temperature span establishment in Figure 4c. It is experimentally proved that a temperature span of more than 7°C was obtained reaching the performances of the best devices from the literature. The cooling power of the heat pump was determined by adding a heating source in the cold reservoir. The resulting cooling power vs. temperature span is given in Figure 5. The overall performance was assessed for several fluid displacement, represented as a relative volume of displaced fluid to the initial volume of fluid in the rubber tubes. For intermediate fluid displacement (V*=0.76), it is obtained a cooling power as high as 2W, and a maximum temperature span of 7°C, which fulfils entirely the initial target fixed in the project proposal. Moreover, in terms of cooling power density, our prototype ranges as high as other elastocaloric or magnetocaloric devices (*9*, *10*) and much higher than electrocaloric devices (*11*, *12*). In terms of temperature span, the results are above the general tendency proposed in (*13*), probably thanks to the low thermal conductivity of natural rubber that is an advance for lowering the passive heat loss across the device.

It should be noted that the device fabrication required to find innovative technical solutions, and the project Principal Investigator and Co-Investigator applied for an international patent (EP22160602) containing the details of the device fabrication.

At the end of the project, several routes were not explored due to the lack of time and resources. However, the results are beyond the expectations, especially on a prototype performance. Further work should deal with (i) a further upscaling of the natural rubber system with a target of cooling power above 10W or even 100W, (ii) a better control of the fluid flow, to increase the effective heat exchange between natural rubber and fluid and (iii) determine and improve the long-term stability and fatigue life of the natural rubber tubes.

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５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　1件／うち国際共著　1件／うちオープンアクセス　0件）

１．著者名 Gael Sebald, Atsuki Komiya, Jacques Jay, Gildas Coativy, Laurent Lebrun	４．巻 127
２．論文標題 Regenerative cooling using elastocaloric rubber: Analytical model and experiments	５．発行年 2020年
３．雑誌名 Journal of Applied Physics	６．最初と最後の頁 94903
掲載論文のDOI（デジタルオブジェクト識別子） 10.1063/1.5132361	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計7件（うち招待講演　1件／うち国際学会　1件）

１．発表者名 Gael Sebald
２．発表標題 Elastocaloric Effect in Polymers
３．学会等名 Sixteenth International Conference on Flow Dynamics（国際学会）
４．発表年 2019年

１．発表者名 Gael Sebald
２．発表標題 Main key points for developing environmental friendly solid state cooling system based on the elastocaloric effect in rubber
３．学会等名 2020 European Materials Research Society (E-MRS) Fall Meeting
４．発表年 2019年

１．発表者名 Gael Sebald
２．発表標題 Modelling of regenerative cooling using elastocaloric elastomers
３．学会等名 Eurotherm Seminar #115 CALORIC HEATING AND COOLING
４．発表年 2021年

１．発表者名
Way Sze Xuen

２．発表標題
Effect of Fluid Flow on Heat Transfer in Elastocaloric Cooling Systems

３．学会等名
32nd International Symposium on Transport Phenomena

４．発表年
2021年

１．発表者名
Gael Sebald

２．発表標題
Elastocaloric Cooling Using Natural Rubber: Material Properties, Heat Transfer and Heat Losses Effects on Proof of Concept Performances

３．学会等名
18th International Conference on Flow Dynamics（招待講演）（招待講演）

４．発表年
2021年

１．発表者名
Way Sze Xuen

２．発表標題
Evaluation of the heat transfer performance of working fluid in elastocaloric cooling devices

３．学会等名
59th Heat Transfer Symposium Japan

４．発表年
2022年

１．発表者名
Way Sze Xuen

２．発表標題
Evaluation of the cooling performance and heat losses of elastocaloric cooling device

３．学会等名
21st Heat Transfer Conference

４．発表年
2020年

〔図書〕　計0件

〔出願〕　計1件

産業財産権の名称	発明者	権利者
CNRS, INSA-Lyon, Tohoku University	Gael Sebald, Atsuki Komiya	同左
産業財産権の種類、番号	**出願年**	**国内・外国の別**
特許、EP22160602	2022年	外国

〔取得〕　計0件

〔その他〕

The research topic includes an international collaboration with research groups in France (INSA Lyon), with complementary developments on the material and other types of heat pumps.

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
研究分担者	小宮　敦樹 (Komiya Atsuki) (60371142)	東北大学・流体科学研究所・教授 (11301)	

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関