様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　４ 年　 ６ 月 １１ 日現在

機関番号：１７１０２
研究種目：基盤研究(C)（一般）
研究期間：2018～2021
課題番号：１８Ｋ０３９８３

研究課題名（和文）A molecular approach to optimisation of liquid coolants

研究課題名（英文）A molecular approach to optimisation of liquid coolants

研究代表者
Ｃａｎｎｏｎ　Ｊａｍｅｓ（Cannon, James）

九州大学・工学研究院・准教授

研究者番号：８０６４８８６６

交付決定額（研究期間全体）：（直接経費）　　3,300,000円

研究成果の概要（和文）：現代社会における冷媒の用途は広範囲に及んでいます。しかし、冷却剤の性質は過去１００年間ほとんど変わっておりません。ほとんどの開発が実験的に行われています。近年の計算能力の向上と機械学習の発展に伴います。本研究では、シミュレーション、統計学、機械学習の観点から、熱物性と冷媒技術の検討に焦点をあてています。

研究成果の学術的意義や社会的意義
The results of this research have yielded new insights into the mechanisms of thermal transport in liquids relevant to coolant technology. Furthermore, the research has demonstrated a new technique to learn this information.

研究成果の概要（英文）：The application of coolants in modern society is far and wide. However the nature of coolants has been largely unchanged during the past 100 years, and most development is largely experimental in nature. With the recent increase in computational power and development of machine learning, this research has focused on the investigation of thermal properties and coolant technology from a simulation and statistical point of view.

研究分野：Thermal Engineering

キーワード：coolant

1．研究開始当初の背景

Coolants play an important role in modern society. They enable machinery and electronics to operate at high capacity. There has however not been significant development of coolants during the past 100 years, and part of this is due to the experimental basis of development which is relatively slow, and the multitude of potential alcohols and other molecules that could make up the coolant.

There are multiple critical properties of coolants, but a significant property is thermal conductivity. This can be important in terms of minimising heat loss from a system in order to conserve energy, or to ensure that heat is transported away from the machinery as quickly as possible in order to prevent overheating. This latter example can typically be found in computer CPU cooling and car engine cooling, for example.

In order to obtain such control of thermal conductivity, coolants are typically used. These are often in liquid form, and aim to transport the heat away from the critical mechanical or electronic parts of machinery. Although the effects of thermal properties can be most clearly observed on the macroscale, the mechanisms of thermal transport are very much defined on the nanoscale. Therefore, in order to understand the nanoscale thermal transport properties relevant to coolants, molecular simulation can play an important role.

2．研究の目的

There are primarily two different ways to study nanoscale thermal transport. One is to use a non-equilibrium thermal gradient. The other way, and the way utilised in this research, is to use a Green-Kubo analysis method. One advantage of using the Green-Kubo method is that it does not require the large temperature gradients found in the first method, and it furthermore permits deep analysis into the heat flux and thermal conductivity. The aim of this research has therefore been to understand if the deep analysis afforded by the Green-Kubo method can give new insight into the mechanisms of thermal transport.

3．研究の方法

The Green-Kubo method is used. In particular, the heat-flux vectors are calculated using

$$\lambda = \frac{V}{k_B T^2}\int_0^\infty \langle J(t).J(0)\rangle dt \qquad \text{(eqn 1)}$$

where J(t) is the heat flux vector at time t, V is the volume and T the temperature, while k_B is the Boltzmann constant. The heat flux vector can be split further into a convective and virial term respectively:

$$J(t) = J_c(t) + J_v(t) \qquad \text{(eqn 2)}$$

this then further results in auto-correlation and cross-correlation contributions to the net thermal conductivity:

$$\lambda = \frac{V}{k_B T^2}\int_0^\infty \langle (J_c(t)+J_v(t)).(J_c(0)+J_v(0))\rangle dt$$

$$= \lambda_{cc} + \lambda_{vv} + \lambda_{cv} + \lambda_{vc} \qquad \text{(eqn 3)}$$

Thus the thermal conductivity has been broken down into convective, virial, and two cross-correlation terms respectively. It is possible to further break down the heat flux vector into interactions based on interactions between molecules and within molecules, and even identify which atom types give rise to what fluxes. Further details can be found in *Manjunatha, L., Takamatsu, H. & Cannon, J.J. Atomic-level breakdown of Green–Kubo relations provides new insight into the mechanisms of thermal conduction. Sci Rep 11, 5597 (2021).*

Simulations are conducted using LAMMPS with some in-house customisation to realise the analysis.

A differential approach is taken: The thermal conductivity paths through five different, yet similar, molecules are observed, and the difference in thermal conductivity and flux are then related to the subtle differences in molecular structure.

4．研究成果

It is clear that the virial contribution to thermal conductivity is greatest (fig 1).

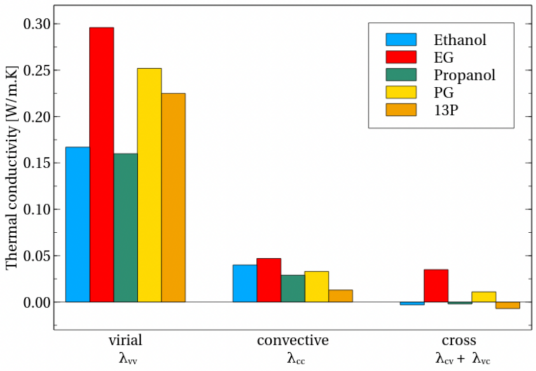

Figure 1: The virial contribution to thermal conductivity is clearly greater than other contributions. Source: Manjunatha, L., Takamatsu, H. & Cannon, J.J. Atomic-level breakdown of Green–Kubo relations provides new insight into the mechanisms of thermal conduction. Sci Rep 11, 5597 (2021).

A significant contribution of this work is to the understanding of the mechanisms of thermal transport on the atomic level. As shown in figure 2, the contribution to thermal conductivity can be broken down into different terms on the atomic level.

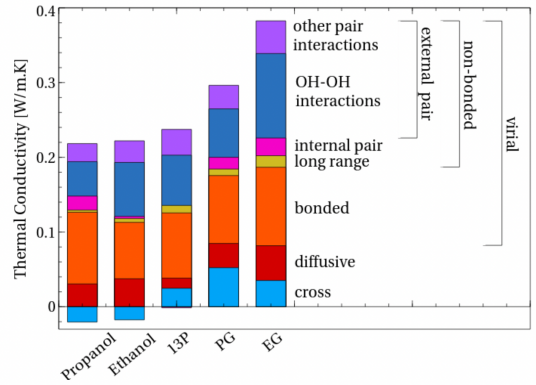

Figure 2: An atomic level breakdown of thermal transport for the 5 alcohols studied here. External pair interactions refer to interactions between molecules. These can be understood in terms of Van der Waals interactions between hydroxyl groups (OH-OH) and others. There are also internal pair interactions between atoms within the molecule, and non-Van der Waals long-range pair interactions. Contributions from bonded interactions, diffusive and cross correlations as mentioned in equation 3 can also be isolated using this technique. Source: Manjunatha, L., Takamatsu, H. & Cannon, J.J. Atomic-level breakdown of Green–Kubo relations provides new insight into the mechanisms of thermal conduction. Sci Rep 11, 5597 (2021).

Overall, this work has demonstrated a new technique to understand the atomic-level contributions to thermal conductivity. This makes a contribution to the understanding of thermal transport which is critical for the successful development of coolants. Furthermore, this demonstrates how simulation can play a useful role to help develop future coolant liquids.

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　1件）

１．著者名 Manjunatha Likhith、Takamatsu Hiroshi、Cannon James J.	４．巻 11
２．論文標題 Atomic-level breakdown of Green-Kubo relations provides new insight into the mechanisms of thermal conduction	５．発行年 2021年
３．雑誌名 Scientific Reports	６．最初と最後の頁 5597
掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41598-021-84446-9	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計7件（うち招待講演　0件／うち国際学会　2件）

１．発表者名 Likhith Manjunatha
２．発表標題 The influence of molecular structure on alcohol thermal conductivity - Understanding conduction mechanisms using break down of the Green-Kubo correlations
３．学会等名 第57回 日本伝熱シンポジウム
４．発表年 2020年

１．発表者名 Likhith Manjunatha, Hiroshi Takamatsu and James J. Cannon
２．発表標題 Ethylene glycol and propanol: Understanding the influence of an extra hydroxyl group on the mechanisms of thermal conductivity
３．学会等名 The 16th UK Heat Transfer Conference, UKHTC2019（国際学会）
４．発表年 2019年

１．発表者名 Makoto Enokimaru, James Cannon, Kazuki Sawayama, Keigo Kitamura and Yasuhiro Fujimitsu
２．発表標題 Dependence of electrical conductivity of geothermal fluid on temperature, pressure and NaCl concentration using molecular dynamics
３．学会等名 International Symposium on Earth Science and Technology 2018（国際学会）
４．発表年 2018年

１．発表者名
榎丸 眞，J．Cannon，澤山 和貴，藤光 康宏

２．発表標題
Dependence of electrical conductivity of subcritical fluid on temperature, pressure and NaCl concentration

３．学会等名
The Geothermal Research Society of Japan

４．発表年
2018年

１．発表者名
L. Manjunatha, H. Takamatsu, J. J. Cannon

２．発表標題
Green-Kubo法を用いた分子動力学シミュレーションによるアルコールの熱伝導率の検討

３．学会等名
日本機械学会 熱工学コンファレンス

４．発表年
2018年

１．発表者名
榎丸 眞、J．Cannon 、澤山 和貴、北村 圭吾、藤光 康宏

２．発表標題
分子動力学を用いた地熱流体の電気伝導度に対する温度・圧力・NaCl濃度の影響

３．学会等名
Japan Geoscience Union Meeting 2018

４．発表年
2018年

１．発表者名
L. Manjunatha, H. Takamatsu, J. J. Cannon

２．発表標題
Investigation into influence of hydroxyl group placement on the thermal conductivity of propane-base alcohols using molecular dynamics simulation

３．学会等名
第8回マイクロ・ナノ工学シンポジウム

４．発表年
2018年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

―

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

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