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Explaining the "anomalous" transient hot wire-based thermal conductivity measurements near solid-liquid phase change in terms of solid-solid transition

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ABSTRACT

Performance of the commonly-used transient hot wire method for measuring the thermal conductivity of fusible materials near solid-liquid phase change is reported with the focus placed on explaining the observed "anomalous" increase of thermal conductivity in relation to the solid-solid transition. Utilizing a 1-D transient formulation and considering a one-step thermal conductivity model, the improved computational methodology captured the monotonic dependence of the predicted thermal conductivity value on the initial temperature of the solid medium more effectively. Hypothesizing that the reported measurements of increase of the thermal conductivity near the solid-liquid phase change temperature are linked to the solid-solid transition, a two-step thermal conductivity model was adopted. This model featured a higher thermal conductivity over a narrow temperature range before a sharp drop upon melting. The predicted values of the thermal conductivity in relation to the initial solid-state temperature were discussed. A rising trend for the predicted thermal conductivity values was observed followed by a smooth decline once the initial solid-state temperature was increased. This predicted trend based on the piecewise thermal conductivity vs. temperature model closely resembled the reported "anomalous" thermal conductivity measurements observed in experimental studies utilizing transient techniques.

1. Introduction

Transport properties of materials, such as thermal conductivity and viscosity, are widely determined using well-established experimental techniques that are based on the fundamental relations between the pertinent flux and the gradient of the relevant quantity. Specifically, determination of the thermal conductivity based on the Fourier's Law has led to a number of techniques categorized as steady-state and transient methods. Among these, determination of the thermal conductivity of gases, liquids, pastes, etc. is generally obtained by the transient hot wire (THW) technique that is based on a time-dependent heat conduction problem (Carslaw and Jaeger [1]). In this idealized limiting case problem, an infinitely long thin wire with a high thermal conductivity placed within an unbounded homogeneous material is considered. The transient signature of the wire's temperature in response to heating of the wire for a short time period is then used to obtain the thermal conductivity of the surrounding material (the relation among the various parameters will be discussed below). Given the slight heating above the initial temperature of the specimen, the THW method leads to an effective thermal conductivity over a temperature range. Smaller heating rate and/or its shorter duration are other considerations that will affect the error associated with the extracted thermal conductivity. An overview of the evolution of the transient hot wire technique from 1780 to 2010 was discussed by Assael et al. [2]. Many corrections to the THW method including deviations from the idealized formulation have been reported. Among these, nine (9) modifications that consider effects of finite inner cylinder (heated wire), composite cylinders, the Knudsen effects, radiation, required condition at the outer cell circumference, the role of compressibility and natural convection, finite cell dimensions, variable fluid properties and the correction for finite length of wire were highlighted by Healy et al. [3].

In recent years, operational deviations from the idealized case [1] associated with performing actual experiments have been treated more conveniently using computational tools. In effect, the above-mentioned and/or other effects are modeled by solving the

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Nomenclature

a	radius of the wire, mm
b	radius of the cylindrical block, mm
C_p	specific heat, J/kg K
G	heat generation term in Eq. (1), W/m^3
k	thermal conductivity, W/m K
L	latent heat of fusion, J/kg
q	strength of the line heat source, W/m
r	radial coordinate, mm
s	instantaneous position of the liquid-solid interface, mm
t	time, s
T	temperature, K

Greek symbols

α	thermal diffusivity, m^2/s
ε_T	temperature difference $(T_m - T_i)$, °C
ε'_T	temperature difference $(T_R - T_i)$, °C
γ	Euler's constant, dimensionless
ρ	density, kg/m^3
τ	time duration s

Subscripts

i	initial
j	generic index
l	liquid
m	melting
p	heat pulse
R	rotator phase
s	solid
w	wire

governing differential equations, thus replacing elaborate analytical relations, e.g. Assael et al. [4], Duluc et al. [5] and Rusconi et al. [6]. Assael et al. [4] used the finite-element method (FEM) to solve a set of energy conservation equations pertinent to a THW experiment that led to accurate predictions of thermal conductivity when compared with experimental values for Argon at high pressure conditions (accuracy of ±0.35%). Duluc et al. [5] studied 2-D transient buoyancy-driven convection in liquid nitrogen around a pulse-heating thin bronze wire by adopting the velocity–pressure formulation, spectral methods and domain decomposition techniques. Considering the wire's temperature rise, the theoretical heat conduction solution and computational results were found to be in close agreement during early time steps signifying a dominant conductive heat transfer regime. Beyond an onset time duration, natural convection effects became important and the experimental data diverged from results of the numerical simulation. Rusconi et al. [6] reported on the same phenomenon using a FlexPDE FEM analysis of a 2-D cylindrical model that entailed simultaneous solution of the Navier-Stokes and heat transport equations in the fluid region. Upon comparing measurements of thermal conductivity obtained from a custom-built THW setup to predictions of the numerical simulations, onset of natural convection effects beyond a specific time duration was clearly elucidated. To remove convection effects from the thermal conductivity measurements, an operational time scale was quantified that depended on the dissipated power from the wire and the physical properties of the test fluid. Recently Bran-Anleu et al. [7] utilized a numerical model to test an algorithm that was developed to select the optimum data range from a transient hot wire experiment automatically, leading to accurate values of thermal conductivity of fluids. Other measurement techniques have also benefited from adoption of computational techniques. For instance, Shen and Khodadadi [8,9] utilized computational fluid dynamics for assessing the roles of thermocapillary and combined thermocapillary-buoyancy convection in determination of the thermal diffusivity of levitated liquid droplets under low-gravity containerless processing conditions.

Greater utilization of computational techniques to improve/extend the range of applicability and account for the actual operating conditions of thermal conductivity measurements were outlined above. In light of the nonexistence of studies that address issues related to the performance of the commonly-used transient methods of measuring the thermal conductivity near the solid-liquid phase transition, Nabil and Khodadadi [10] recently adopted a 1-D unsteady formulation for the transient hot wire method. The model medium of interest exhibited constant values of thermal conductivity in both liquid and solid phases having a sharp transition temperature. Predictions of the wire temperature obtained from the computational methodology for cases without and with solid-liquid phase change were compared to the limiting analytical solutions. The results exhibited a monotonic dependence of the predicted thermal conductivity value of the sample on the initial temperature of the solid medium. The predicted thermal conductivity moved toward the value of the liquid sample as the initial temperature of the solid-state sample approached the melting point. Recommendations were also provided for performing measurements of thermal conductivity using the transient hot wire technique involving solid-liquid phase change.

Utilizing and modifying the approach of Nabil and Khodadadi [10] to explain the "anomalous" THW-based thermal conductivity measurements near solid-liquid phase transition in terms of increased thermal transport linked to a model solid-solid transition is reported in this paper.

2. Problem statement

In order to determine the temperature-dependent thermal conductivity of phase change materials (PCM) in their solid-state using the transient methods, i.e. transient plane source (TPS) and transient hot wire methods, researchers have investigated thermal conductivity values as the reported measurement temperature nears the solid-liquid phase transition temperature of the PCM. An idealized one-step model of variation of the thermal conductivity with temperature that assumes constant values of thermal conductivity of the solid (k_s) and liquid (k_l) phases on both sides of the distinct melting temperature (T_m) is shown in Fig. 1. As the measurement temperature of the sample nears the melting temperature, an idealized sudden drop in the thermal conductivity values is noted. The high value of thermal conductivity is associated with orderly structure of matter in the solid phase, whereas random movements of molecules in the liquid phase lead to a lower thermal conductivity. Operation of the THW (or TPS) technique under such non-equilibrium condition is not compatible with the applicable theory [1]. Moreover, the transient techniques are ideally-suited to determine the thermal conductivity of a homogenous medium. Upon violation of these conditions, the solid PCM starts transforming into two separate phases and the measured thermal conductivity values are not accurate. Nabil and Khodadadi [10] addressed issues related to the single-step thermal conductivity model of Fig. 1 by adopting both analytical and computational approaches. Since the current extension of [10] utilizes the same approach, a brief overview is provided. In the geometry of the