Postprint: Measurement of Typical Thermophysical Properties of Materials Under Electrostatic Levitation

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Abstract

As materials research advances, methods for materials preparation and analysis become increasingly important; however, certain fundamental physical properties constitute the basis for conducting relevant investigations. Due to the high melting points and refractory nature of some materials, traditional methods cannot circumvent contamination from container walls, cannot perform experiments under vacuum conditions to avoid gaseous contamination, or are limited to measuring specific materials due to experimental constraints; these methods encounter significant difficulties in measuring the thermophysical properties of materials in the superheated and supercooled regimes at elevated temperatures. This article systematically introduces electrostatic levitation technology as a novel method for achieving deep undercooling to enable measurement of material thermophysical properties. Electrostatic levitation technology suspends a sample between two electrode plates, utilizes laser heating of the sample while levitated to achieve high-temperature melting, and simultaneously performs thermophysical property measurements. This paper compares several methods for measuring typical thermophysical properties, details the advantages of electrostatic levitation, and provides a comprehensive introduction to the measurement of melt density, thermal expansion coefficient, surface tension, viscosity coefficient, and specific heat of materials using electrostatic levitation technology.

Full Text

Preamble

Thermophysical Property Measurements by Electrostatic Levitation in Material Science

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Abstract

As materials research advances, methods for material preparation and analysis have become increasingly important, yet these methods fundamentally rely on accurate knowledge of key physical properties. Many materials present significant challenges due to their high melting points and refractory nature, while conventional techniques suffer from container wall contamination, inability to operate under vacuum conditions to avoid gas contamination, or limitations to specific materials. These approaches struggle to measure thermophysical properties during superheating and undercooling stages at elevated temperatures. This paper systematically introduces electrostatic levitation as a novel technique for achieving deep undercooling and measuring material thermophysical properties. Electrostatic levitation suspends samples between two electrode plates while employing laser heating to melt materials at high temperatures, enabling simultaneous thermophysical property measurements. We compare several typical methods for measuring thermophysical properties, elucidate the advantages of electrostatic levitation, and provide a detailed description of how electrostatic levitation measures melt density, thermal expansion coefficient, surface tension, viscosity coefficient, and specific heat.

Keywords: electrostatic levitation; thermophysical property measurements; density; thermal expansion coefficient; surface tension; coefficient of viscosity; specific heat

0 Introduction

The drop tube represents a free-fall deep undercooling rapid solidification technique.

1 Development of Classical Containerless Measurement Methods

The development of novel materials is crucial for national defense, aerospace, and advanced industrial applications. During materials exploration, measuring physical properties at high temperatures is essential, yet conventional container-based methods suffer from sample contamination by container walls, significantly affecting measurement accuracy. Containerless techniques suspend samples, eliminating wall effects during melting and solidification, and enable real-time measurement of internal structure and physical properties through non-contact optical methods. Containerless processing represents a key research direction in space materials science, with active investigations by NASA, Japan, and the European Space Agency [1-4].

Containerless methods primarily measure melt density, thermal expansion coefficient, surface tension, viscosity coefficient, and specific heat under deep undercooling. Density, a fundamental material property, varies with temperature and pressure and plays a vital role across numerous applications. The thermal expansion coefficient is a basic thermophysical parameter critical for engineering design, precision instrument manufacturing, welding, and materials processing. Surface tension governs surface thermodynamics and fluid dynamics, influencing casting, laser surface remelting, welding, and rotational liquid processing. Viscosity reflects liquid alloy fluidity and microstructure [5], significantly affecting melting, mold filling, and solidification synthesis [6]. Specific heat, a thermodynamic property reflecting temperature-energy relationships [7], is essential for thermodynamic calculations of metal solidification and phase transformation studies, and serves as a prerequisite for nucleation rate calculations. Consequently, specific heat and total hemispherical emissivity are two critical parameters for studying materials in deeply undercooled states.

Primary containerless techniques include free-fall and levitation methods [8]. Electrostatic levitation represents an advanced measurement approach among containerless methods. The drop tube, the earliest containerless technique, dates to 1782 when William Watts patented a free-fall method for improving lead shot sphericity—the first application of weightlessness for materials processing. In 1956, Turnbull built the world' s first prototype drop tube for Fe-Ni alloy undercooling studies [9]. After decades of development, drop tubes enable containerless solidification in microgravity, combining deep undercooling with rapid quenching [10-12], typically achieving substantial undercooling for studying liquid metal rapid solidification [11,13]. Currently, dozens of drop tubes operate worldwide. Despite being an excellent microgravity and containerless method, drop tubes require large facilities, incur high costs, and offer limited experiment duration, imposing significant constraints.

Electromagnetic levitation originated in 1923 when Mack proposed suspending metals using high-frequency electromagnetic fields, patenting "electromagnetic levitation melting" [14]. Okress conducted the first suspension experiments and preliminary theoretical studies [15]. In the late 1950s, Comenetz successfully levitated ~10g metal spheres of dozens of types using a 450kHz, 10kW high-frequency source [16]. In 1971, Lu and colleagues at McMaster University first applied electromagnetic levitation droplet oscillation for surface tension measurement [17]. Today, electromagnetic levitation primarily suits metals with high magnetic permeability and semiconductors with high electrical conductivity [18,19], using alternating magnetic fields for simultaneous levitation and heating in vacuum or protective atmospheres, with inherent electromagnetic stirring. However, achievable undercooling is limited by sample purity, and melt vaporization during levitation affects specific heat measurements of undercooled melts.

Acoustic levitation, first explained by L.V. King in 1934 [20], uses strong acoustic fields to generate radiation pressure balancing gravity. It finds broad applications in thermophysical property measurement, solidification kinetics, and liquid motion studies. Compared to other methods, acoustic levitation imposes no electrical or magnetic property constraints, enabling processing of diverse materials. However, its levitation capability and stability are significantly affected by ambient gas temperature, restricting studies primarily to low-melting-point materials. While acoustic levitation eliminates container wall effects on nucleation and enables nanofluid undercooling experiments [21], it requires gas environments that cannot prevent external contamination, offers weak levitation forces, and introduces mechanical interference from acoustic waves, making it unsuitable for high-temperature liquid thermophysical property measurements.

Aerodynamic levitation achieves stable suspension through controlled gas flow and optimized nozzle design, offering simple equipment and convenient operation for various materials. In 1982, spherical solid samples were levitated under low-pressure free jets and laser-heated [20]. Aerodynamic levitation provides excellent single-droplet manipulation for studying cooling rate effects. Rapid cooling enables substantial undercooling, altering microstructure formation. However, latent heat release reduces undercooling at solidification interfaces, causing transitions from irregular to regular eutectics in both single grains and macroscopic samples [22]. While aerodynamic levitation offers superior temperature control, achieving stable heat balance for temperature regulation, it requires gas environments that disturb solidification and complicate precise position control.

2 Development of Electrostatic Levitation Measurement Methods

Electrostatic levitation employs Coulomb forces in an electric field to suspend samples, providing a clean, interference-free environment without convection from gas or electromagnetic effects [23]. It suits all materials capable of maintaining sufficient surface charge, including metals, semiconductors, and insulators. In 1959, Langmuir et al. [24] studied electrostatic levitation of micron-sized particles without feedback control, establishing the technique's materials science foundation. In 1984, Rhim and colleagues at NASA' s Jet Propulsion Laboratory first proposed electrostatic levitation for containerless materials processing for space experiments, designing three electrode configurations with feedback control to achieve stable suspension of 5mm silver-coated styrene spheres and ~10mm water droplets in parabolic flight low-gravity environments [25].

Electrostatic levitation matured through the 1990s. In 1993, Rhim et al. developed ground-based electrostatic levitation equipment for high-temperature containerless materials processing [26], using position-sensitive detectors (PSD) for three-dimensional position sensing. In 2000, Nakamura et al. detailed electrostatic levitation furnace control systems [27], employing two vertically-mounted 500Hz high-speed CCD cameras for 3D position detection. In 2001, Meister et al. investigated gain-scheduled control methods [28]. In 2005, Ishikawa et al. described JAXA' s electrostatic levitation furnace [29], which suspends 0.9-3mm samples using PSD with PID control. JAXA has since developed ground-based and aircraft-mounted electrostatic levitation systems [30-34], targeting space containerless processing applications. NASA' s electrostatic levitation capabilities are comparable [1].

Domestic electrostatic levitation research, while nascent, is rapidly advancing. After over a decade of development, Northwestern Polytechnical University' s Space Materials Science and Technology Laboratory has successfully developed ground-based electrostatic levitation systems. The National Space Science Center has achieved suspension, heating control, and classical thermophysical property measurements after several years of research. Figure 1 illustrates the thermophysical property measurement principle: a stainless steel vacuum chamber sits on a vibration isolation table, with the sample positioned between two plates and observation, measurement, and heating equipment arranged around the chamber. Two orthogonal lasers and 2D position detectors monitor sample position, while three diode laser arrays spaced at 120° intervals heat the sample. Dual-wavelength pyrometers measure temperature across different ranges, UV sources and density-measurement CCD cameras determine sample volume for melt density calculations, position lasers and amplitude sensors measure sample oscillations for surface tension and viscosity determination, and dual-wavelength pyrometry combined with single-wavelength measurements calculates emissivity for specific heat determination under deep undercooling.

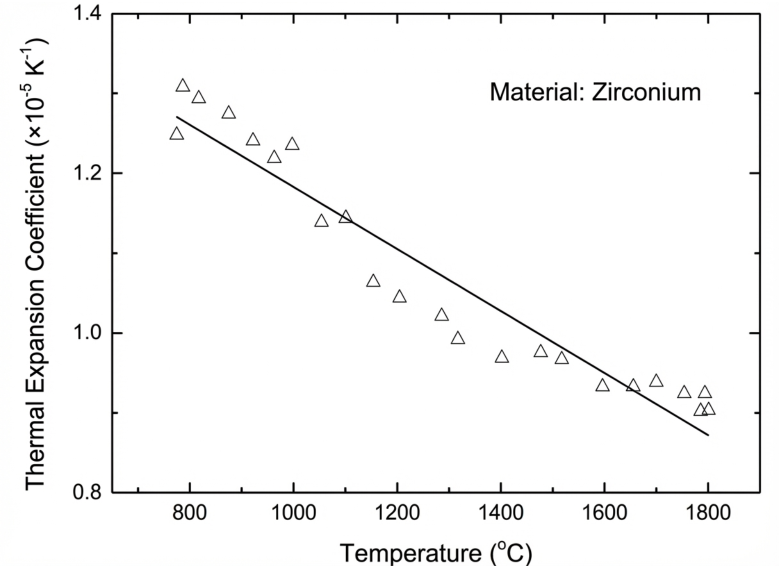

Figure 1: Figure 1

3 Measurement of Classical Thermophysical Properties Under Electrostatic Levitation

Electrostatic levitation enables effective thermophysical property measurement for numerous high-melting-point metallic and non-metallic materials. The entire process occurs under high vacuum, avoiding medium interference. Independent control of heating and levitation maintains suspension across wide temperature ranges, while feedback control keeps samples stationary, facilitating heating and deep undercooling [35]. This contamination-free environment is crucial for surface tension measurements of high-temperature, highly reactive conductive materials, as liquid metals readily react with container walls at elevated temperatures. Containerless processing eliminates heterogeneous nucleation from walls, enabling deep undercooling and accurate measurement of high-temperature liquid thermophysical properties.

3.1 Melt Density and Thermal Expansion Coefficient Measurement

For high-melting-point materials in electrostatic levitation furnaces, density-measurement CCD cameras capture spherical sample profile areas when heated to melting. The volume V is calculated from the magnified spherical sample images. Since levitated melts form perfect spheres, the projected image yields a circular outline from which radius r is determined and volume V calculated. Combined with measured mass, the density of molten high-melting-point materials is obtained. The thermal expansion coefficient is then calculated using:

[FIGURE:2] and [FIGURE:3] show sample volume and thermal expansion coefficient versus temperature curves, respectively. Traditional methods struggle to obtain accurate values without container contact or interference from gases and other conditions.

3.2 Surface Tension and Viscosity Coefficient Measurement

Under electrostatic levitation, surface tension and viscosity measurements become feasible even for high-melting-point materials where conventional methods fail. Based on the surface tension calculation formula:

where ω is the natural oscillation frequency, σ is surface tension, is melt density, and r is sample radius. With and r obtained from prior density measurements, determining the natural vibration frequency (resonant frequency) ω yields surface tension σ:

JAXA has measured extensive surface tension and viscosity data across temperature ranges including deep undercooling. For pure metals, melt surface tension exhibits a linear temperature relationship, decreasing with increasing temperature. Viscosity data at melting points remain scarce for many metals, but electrostatic levitation fills this gap. Data show viscosity also inversely correlates with temperature in pure molten metals.

The relationship between vibration decay time and viscosity coefficient is:

where τ is the free decay time after external vibration excitation removal, is viscosity coefficient, is melt density, and r is sample radius. With and r known from density measurements, determining τ yields viscosity :

Thus, knowing both ω and τ determines surface tension and viscosity. For low-viscosity liquids, the droplet vibration method [36] is employed. This method, predicated on spherical test materials, uses controlled DC electrostatic fields near the droplet' s resonant frequency to excite oscillations. After excitation stops, the droplet continues vibrating at its natural frequency (determined by surface tension) with decay time (determined by viscosity). Electrostatically levitated melts are nearly spherical with stable position control, enabling surface tension and viscosity determination from vibration decay analysis. Position lasers and amplitude sensors measure sample vibrations, producing time-amplitude curves. A bandpass Fourier filter removes low-frequency vibrations from sample wobbling, and the filtered data is fitted to damped waveform templates to calculate surface tension and viscosity.

Figure 6 [FIGURE:6] and Figure 7 [FIGURE:7] show surface tension and viscosity coefficient versus temperature for yttrium samples [37]. These measurements can span wide temperature ranges including deep undercooling stages.

3.3 Specific Heat Measurement

Specific heat measurement is crucial in materials science, yet conventional methods suffer from external interference, preventing accurate determination of high-melting-point material values. Electrostatic levitation fills this gap, enabling relatively accurate specific heat measurements. The deep undercooling specific heat calculation principle is:

where m is sample mass, M is molar mass, C__p is specific heat, (T) is emissivity at temperature T, A is sample area, σ_s is Stefan-Boltzmann constant, T is sample temperature, T__0 is ambient temperature, and dT/dt is temperature change rate. Sample area A is obtained from density measurement images, temperature change rate from deep undercooling cooling curves, enabling specific heat calculation under deep undercooling:

The specific heat measurement system obtains cooling curves and emissivity parameters from solidifying suspended samples to calculate deep undercooling stage specific heat. Temperature and emissivity measurement units employ dual-wavelength pyrometry for temperature, single-wavelength pyrometry combined with temperature for emissivity, and thermocouples for ambient temperature. The system controls laser heater output to melt samples, then shuts off the laser to allow cooling and solidification, obtaining deep undercooling cooling curves as shown in Figure 9 [FIGURE:9]. Note the two distinct stages in Figure 9 due to different emissivities for liquid and solid samples—one during heating/melting, another during cooling. Analyzing only the deep undercooling curve using the specific heat formula yields the desired values.

For silicon samples under electrostatic levitation, multiple heating-cooling cycles produce stable time-temperature curves. Analysis can estimate the ratio of specific heat to hemispherical emissivity. Polynomial fitting further derives temperature-dependent functions for both specific heat and hemispherical emissivity. Following Y.S. Sung' s work on measuring Si specific heat and hemispherical emissivity under electrostatic levitation using 99.999% pure spherical Si samples [38], the values can be determined.

4 Summary and Outlook

Electrostatic levitation enables containerless processing without additional energy input to samples or interference with solidification processes, facilitating integration with other non-contact measurement devices for thermophysical property determination [39,40]. Compared to other containerless methods, electrostatic levitation is indispensable in materials science. Currently, domestic systems can measure several typical thermophysical properties of high-temperature molten samples, establishing parameter variation relationships for different materials. However, compared to mature US and Japanese technologies, domestic experiments require further research and improvement, including enhancing suspension success rates through algorithm refinement, improving measurement precision, and correcting for charge and shape variation interferences. These challenges warrant continued investigation and breakthrough.

References

[1] Zhou Yangyang. Design of Electrostatic Levitation Position Control System[D]. Graduate University of Chinese Academy of Sciences (Center for Space Science and Applied Research), 2010.
[2] Sung Y S, Takeya H, Togano K. Containerless solidification of Si, Zr, Nb, and Mo by electrostatic levitation[J]. Review of Scientific Instruments, 2001, 72(12):4419-4423.
[3] Rulison A J, Watkins J L, Zambrano B. Electrostatic containerless processing system[J]. Review of Scientific Instruments, 1997, 68(7):2856-2863.
[4] Meister T, Werner H, Lohoefer G, et al. Gain-scheduled control of an electrostatic levitator[J]. Control Engineering Practice, 2003, 11(2):117–128.
[5] Paradis P F, Ishikawa T, Yoda S. Electrostatic Levitation Research and Development at JAXA: Past and Present Activities in Thermophysics[J]. International Journal of Thermophysics, 2005, 26(4):1031-1049.
[6] Paradis P F, Ishikawa T, Koike N. Non-contact measurements of surface tension and viscosity of molybdenum using an electrostatic levitation furnace[J]. International Journal of Refractory Metals & Hard Materials, 2007, 25(1):95-100.
[7] Rhim W K, Chung S K, Barber D, et al. An electrostatic levitator for high-temperature containerless materials processing in 1-g[J]. Review of Scientific Instruments, 1993, 64(10):2961-2970.
[8] Lü Yongjun. Deep Undercooling and Rapid Solidification Under Acoustic

Levitation and Free Fall Conditions[D]. Northwestern Polytechnical University, 2002.
[9] Qin Zhicheng, Yang Haiqing, Wang Wenkui, et al. Drop tube—A ground simulation method for space experiments[J]. Modern Physics, 1991(2).
[10] Ludwig A, Wagner I, Laakmann J, et al. Undercooling of superalloy melts: basis of a new single-crystal manufacturing technique for turbine blades[J]. Materials Science & Engineering A, 1994, 178(1):299-303.
[11] Hofmeister W H, Evans N D, Bayuzick R J, et al. Microstructures of niobium-germanium alloys processed in inert gas in the 100 meter drop tube[J]. Metallurgical & Materials Transactions A, 1986, 17(8):1421-1428.
[12] Vinet B, Cortella L, Favier J J, et al. Highly undercooled W and Re drops in an ultrahigh-vacuum drop tube[J]. Applied Physics Letters, 1991, 58(1):97-99.
[13] Jansen R, Sahm P R. Solidification under microgravity[J]. Materials Science Engineering B, 1987, 26(1-2):199-212.
[14] Muck. Electromagnetic levitation: German. 422004[P].1923
[15] Okress E C, Wroughton D M, Comenetz G, et al. Electromagnetic levitation of solid and molten metals[J]. J.APPL.Phys., 1952, 23(5);525-547.
[16] Begley R T, Comenetz G, Flinn P A, et al. Vacuum Levitation Melting[J]. Review of Scientific Instruments, 1959, 30(1):38-38.
[17] Han Xiujun. Thermophysical Properties of Metastable Metallic Melts and Their Rapid Solidification[D]. Northwestern Polytechnical University, 2002.
[18] Wei Bingbo, Barth. Thermodynamic Driving Force for Rapid Crystallization of Liquid Metals[J]. Acta Metallurgica Sinica, 1994(7):289-293.
[19] Wang Haipeng, Luo Bingchi, Chang Jian, et al. Study on specific heat and related thermophysical properties of liquid Fe-Cu-Mo alloy[J]. Science in China, 2007, 37(3):273-281.
[20] Xiang Heng. Aerodynamic Levitation Solidification and Dynamics Study of $BaTiO_3$ and $BaTiO_5$ Ceramics[D]. China University of Geosciences (Beijing),
[21] Hu Pengfei. Study on Undercooling and Nucleation Characteristics of Graphene Oxide Nanofluids Under Acoustic Levitation[D]. Chongqing University, 2014.
[22] Geng Delu, Xie Wenjun, Hong Zhenyu, et al. Containerless solidification of Ag-Cu eutectic alloy under acoustic levitation[J]. Science in China: Physics, Mechanics & Astronomy, 2011(3):227-235.
[23] Wei B, Yang G, Zhou Y. High undercooling and rapid solidification of Ni 32.5% Sn eutectic alloy[J]. Acta Metallurgica Et Materialia, 1991, 39(6):1249-1258.
[24] Wuerker R F, Shelton H, Langmuir R V. Electrodynamic Containment of Charged Particles[J]. Journal of Applied Physics, 1959, 30(3):342-349.
[25] Wang T G, Trinh E, Rhim W K et al. Development of an electrostatic positioner for space material processing [J]. Space material processing, 1984, 56(6):42-84
[26] Won-Kyu Rhim, Sang K. Chung, Daniel Barber et al. An electrostatic levitator for high-temperature containerless materials processing in l-g [J]. Rev Sci Instrum 1993, 64(8):46-832
[27] TAI NAKAMURA Y A, HARUHIKO SHIMOJI ET AL. CONTROL SYS-

TEM OF ELECTROSTATIC LEVITATION FURNACE [J]. Acta Astronautica, 2000, 50(5):1-34
[28] T. Meister, H. Werner, G. Lohoefer et al. Gain-scheduled Control of an Electrostatic Levitator [J]. Control Engineering Practice, 2003, 27(3):3-35
[29] P.-F. Paradis, T. Ishikawa, S. Yoda. Electrostatic Levitation Research and Development at JAXA: Past and Present Activities in Thermophysics [J]. International Journal of Thermophysics, 2005, 26(1):23-34
[30] Jianding Yua, Naokiyo Koshikawa et al. Containerless solidification of oxide material using an electrostatic levitation furnace in microgravity [J]. Journal of Crystal Growth, 2001, 231(9): 3-77)
[31] Yasutomo Arai, Naokiyo Koshikawa, Takehiko Ishikawa et al. Specifications of a ground-based electrostatic levitation furnace and applications to the study of liquid properties [J]. Journal of Non-Crystalline Solids, 2002, 314(4):75-388
[32] Yang Xiaoguang, Lu Xiaoyu, Cao Chongde. Design and Application of Material Electrostatic Levitation Control System [J]. Journal of Northwestern Polytechnical University, 2009, 27(4):25-255
[33] Guo Jingjie, Fu Hengzhi. Alloy Melts and Their Treatment. Beijing: Mechanical Industry Press, 2005.24
[34] Takehiko Ishikawa. Measurement of Thermal Properties of Refractory Metals Using Electrostatic Levitation Technique [D]. Tokyo; Tokyo Institute of Technology, 2001
[35] Paul-Franc, ois Paradis, Takehiko Ishikawa et al. Experiments in materials science on the ground and in reduced gravity using electrostatic levitators [J]. Advances in Space Research, 2008, 41(3):2-56
[36] Guo Jingjie, Fu Hengzhi. Alloy Melts and Their Treatment. Beijing: Mechanical Industry Press, 2005.24
[37] Zhou Lu. Physical Chemistry Tutorial. Beijing: Science Press, 2006.13
[38] Paradis P F, Ishikawa T, Koike N. Thermophysical Properties of Molten Yttrium Measured by Non-contact Techniques[J]. Microgravity Science and Technology, 2009, 21(1-2):113-118.
[39] Sung Y S, Takeya H, Hirata K, et al. Laboratory: High-Temperature Materials Science 2000-2004[J]. List of Publications 2000-2004, 2003, 83(6):1122 - 1124.
[40] Ishikawa T, Paradis P F, Watanabe Y, et al. Thermophysical Properties of Liquid and Supercooled Rare Earth Elements Measured by an Electrostatic Levitator[J]. Jaxa Research & Development Report, 2009, 8:1-14.
[41] Hu Hanqi. Principles of Metal Solidification. Beijing: Mechanical Industry Press, 2000.255