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Miniaturized System for Vapor Pressure Measurement using a Combination of Knudsen Cell and Nanobalance

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Summary:
A miniaturized system for measurement of vapor pressure is presented. It consists of a Knudsen cell and a langasite based resonant nanobalance. The molecular beam from the cell hits the surface of the nanobalance and forms a thin film which results in an increase of the mass and thus in a shift of the resonance frequency. The mass sensitivity of the high-temperature stable nanobalance enables even detection of single monolayers of deposited material. The resonance data is analyzed using impedance spectroscopy as well as frequency counting techniques.

Keywords: piezoelectricity, Knudsen effusion, impedance spectroscopy, microgravity

Introduction and Objectives

Precision data for the free enthalpy of pure substances and alloys, which is essential for development of new materials, improvements in process control as well as determination of application limits of new material systems, can be obtained through vapor pressure measurements [1]. It is commonly determined by means of mass spectrometry on a molecular beam produced by a resistively heated Knudsen cell [2,3]. Conventional systems exhibit however several drawbacks. The sample stays in contact with the crucible and thus a risk of contamination is given. Furthermore the investigated material cannot evaporate equally in all directions. Finally, the size of the experimental setup is not suited to be applied in systems like e.g. microgravity levitation systems in which the abovementioned disadvantages do not exist.

The miniaturized system presented here overcomes the aforementioned limitations as the conventional mass spectrometer is replaced by a nanobalance, resulting in significant reduction of size. It allows the measurement of the Knudsen effusion during e.g. electromagnetic levitation experiments. The capabilities of the system are validated during German Aerospace Center (Deutsches Luft- und Raumfahrt, DLR) and European Space Agency (ESA) parabolic flight campaigns (PF) in 2018, 2019 and 2020.

Description of the System

The nanobalance system for Knudsen effusion measurements is shown schematically in Fig. 1. It consists of a regular Knudsen cell and a high-temperature stable nanobalance based on piezoelectric langasite resonators ($La_3Ga_5SiO_{14}$, LGS). The molecular beam from the cell hits the surface of the nanobalance and forms a thin film which results in an increase of the mass and thus in a shift of the resonance frequency. An additional shutter placed between Knudsen cell and resonator (not shown in the schema) allows to interrupt the deposition process during the experiment.

Due to the proximity of the resonator, heating by infrared radiation from the sample has to be taken into account. Therefore, the system uses

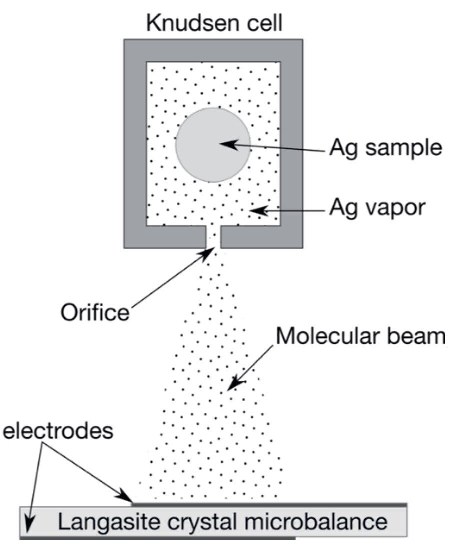

Fig. 1. Function principle of a nanobalance-based set-up for the measurement of a Knudsen effusion.

a Pt-100 resistive temperature sensor placed in the vicinity of the resonator. In combination with temperature compensation techniques, such as application of the 3rd overtone [4], shifts of the resonance frequency due to increasing mass load are decoupled from disturbing temperature effects.

Experimental

The system is tested in combination with the TEMPUS electromagnetic levitation facility from the DLR. There, the material systems Ag and $Ag_{60}Cu_{40}$ are annealed in the temperature range from 850 to 950 °C. The shutter between Knudsen cell and nanobalance is opened as soon as the sample is levitating and its temperature stabilizes. The temperature of the LGS resonator is recorded using a Keithley DVM-2000 digital voltmeter. The duration of the experiment is limited to approximately 20 seconds, which is determined by the length of the microgravity condition during parabolas.

The frequency shift is measured using two different approaches. The first is the impedance measurement of LGS in the vicinity of its resonance frequency (both fundamental mode and the overtones). The data acquired using a network analyzer (Agilent E5100A) is calibrated in order to exclude the stray capacitances caused by the resonator support and the wiring. Subsequently, a Lorentz curve is fitted to the conductance peak, giving information about the resonance frequency and Q-factor of the resonator. This method is however limited to a sampling rate of about 1 Hz.

Another measurement method incorporates an oscillator circuit driven by the LGS resonator. The oscillator circuit operates at the series resonance frequency. The latter is recorded using a calibrated and temperature compensated 10 ns time base. Finally, the data is analyzed and stored in form of a resonance frequency as a function of time. This system allows recording of resonance frequency at a rate of up to 100 Hz.

Results and Discussion

The Ag samples are analyzed in microgravity conditions during PF in 2018 and 2019. Thereby, the data is acquired at a temperature ranging from 1173 to 1373 K (see Fig. 2). The lowest measured mass flow rate of Ag is 1.5 ng/s at 1193 K. The results are compared with numerical simulations of the system made using Comsol Multiphysics 5.3a [5] and found to be in good agreement.

The $Ag_{60}Cu_{40}$ sample is analyzed during PF of ESA in 2020.

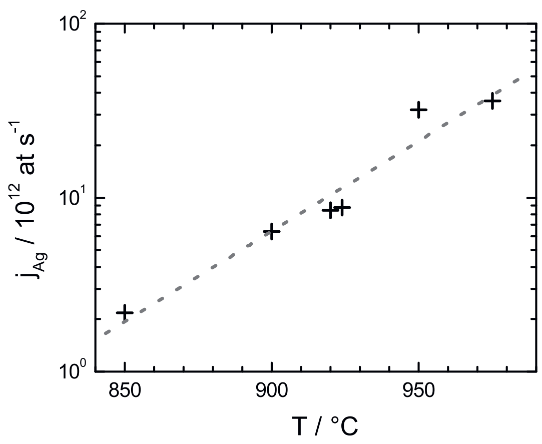

Fig. 2. Ag molecular flux as a function of temperature.

Conclusions

The miniaturized system presented here enables vapor pressure measurement in microgravity conditions. The vapor pressure of pure Ag and $Ag_{60}Cu_{40}$ are determined and confirm the usability of the system. The measurements using an oscillator circuit allow determination of the resonance frequency at high sampling rates.

Acknowledgment

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