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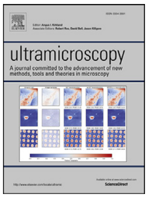

Advanced microheater for *in situ* transmission electron microscopy; enabling unexplored analytical studies and extreme spatial stability

J. Tijn van Omme*, Marina Zakhozheva, Ronald G. Spruit, Mariya Sholkina, H. Hugo Pérez Garza

DENSsolutions B.V., Informaticalaan 12, Delft 2628 ZD, The Netherlands

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ABSTRACT

In this work we present our advanced *in situ* heating sample carrier for transmission electron microscopy (TEM). The TEM is a powerful tool for materials characterization, especially when combined with micro electro-mechanical systems (MEMS). These deliver *in situ* stimuli such as heating, in which case temperatures up to 1300 °C can be reached with high temporal stability without affecting the original TEM spatial resolution: indeed, atomic resolution imaging can be routinely performed. Previously, the thermal expansion of suspended microheaters caused vertical displacement of the sample (bulging). As a result, changing temperatures required either continuous focus or stage adjustments, inducing resolution loss or mechanical drift, respectively. Moreover, those actions hinder the possibility to capture fast dynamic events. This new MEMS-based sample carrier, however, keeps the sample at constant *z*-position (no bulging) up to 700 °C. Furthermore, it enables energy dispersive x-ray spectroscopy (EDS) acquisition in the TEM up to an unmatched temperature of 1000 °C, with a drift rate down to 0.1 nm/min. Its viewable area of 850 µm^2 features a temperature homogeneity up to 99.5%.

1. Introduction

Materials research is vital in addressing challenges in a wide range of research topics, both from the fundamental and industrial point of view [1–3]. Having the capability to observe and understand dynamic events as a consequence of a given stimulus (i.e. heat) is necessary to determine how to manipulate and optimise materials and processes. Several techniques are commonly available to characterise and visualize events such as changes in morphology, failure, catalysis and synthesis of nanomaterials [4–6]. Among them, the TEM enables imaging and analysis at sub-ångström spatial and sub-eV energy resolution [7,8]. However, traditional TEM restricts operation to static conditions in high vacuum and room temperature [9,10].

The first TEM heating experiments used furnace-like configurations or filaments, which led to massive sample drifts due to thermal expansions and long stabilization times [11,12]. Therefore, the traditional copper sample grids were replaced by MEMS devices, which became the new consumable sample carriers that only manipulate the environment just around the sample [13]. By placing the microheater on a suspended membrane, it is isolated from the silicon frame and the drift is minimized [14–17]. Together with the development of aberration corrected electron optics and fast cameras and detectors, this caused a boost in the number of published papers about *in situ* TEM research over the past years [10,18–21], with *in situ* heating being one of the most popular [22].

Nevertheless, new limitations arose, most importantly the bulging of the membrane due to thermal expansion, causing the sample to move out of focus upon temperature variation. This severely limits the resolution especially in aberration corrected scanning TEM (STEM) or requires meticulous stage alignment, nonetheless inducing mechanical drift. Moreover, bulging makes it impossible to capture fast dynamics in heating/quenching experiments. Secondly EDS analysis, a central tool in analytical electron microscopy, was so far comfortably possible only below 700 °C in S/TEM due to the infrared radiation emitted by the heater [23,24].

In this work we present our innovative microheater for *in situ* TEM (Fig. 1(a)) that was optimized in terms of bulging, sample drift and EDS analysis. On this MEMS device, referred to as the Nano-Chip, other important performance parameters such as temperature homogeneity, viewable area and ease of sample preparation were also improved. Overall, this Nano-Chip represents a considerable advantage over commercial and custom-designed MEMS devices for *in situ* TEM purposes [14–17,23,25–27].

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* Corresponding author.

E-mail addresses: tijn.vanomme@denssolutions.com (J.T. van Omme), marina.zakhozheva@denssolutions.com (M. Zakhozheva), ronald.spruit@denssolutions.com (R.G. Spruit), mariya.sholkina@denssolutions.com (M. Sholkina), hugo.perez@denssolutions.com (H.H. Pérez Garza).

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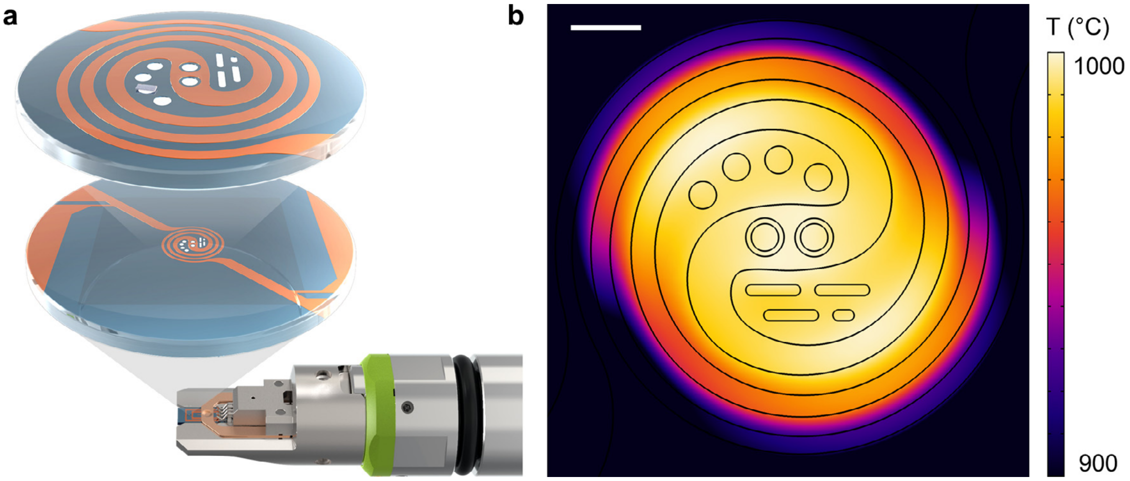

Fig. 1. Nano-Chip design. (a) Overview of the Nano-Chip in the Wildfire *in situ* heating holder. Samples are placed in the centre of the metallic microheater spiral, which is located on the suspended membrane of the MEMS device. The Nano-Chip is loaded into the dedicated TEM holder with four needles to read the resistance and supply the current. Accurate temperature control is realized with the four-point-probe system. (b) Simulation of the temperature distribution across the microheater at a power of 10 mW, reaching a temperature of 1000 °C at the central windows. The varying linewidth results in a homogeneous microheater with a temperature uniformity of 98% across the window area and 99.5% across the two central windows (also shown in the zoomed-in temperature simulation of Fig. 2(d)). Scale bar is 30 µm.

Comparable to our previous efforts, the metallic microheater spiral on the suspended membrane is encapsulated in silicon nitride [14,15]. The Nano-Chip is inserted into the TEM using a dedicated holder (Fig. 1(a)) with four needles that make ohmic contact with the contact pads. Two contacts deliver the current that induces Joule heating, while the two others measure the microheater's resistance, which linearly depends on temperature. This four-point-probe configuration excludes the influences of contact resistances and wires [28], enabling the closed loop feedback system to guarantee a temperature stability in the millikelvin range. Furthermore, this configuration effectively handles changes in the environment, for example when the heat balance is disrupted through the introduction of a (different) gas in an environmental TEM.

2. Limitations of existing microheaters for *in situ* purposes

Current state of the art MEMS-based sample carriers opened up new fields of research by enabling low sample drift and atomic resolution while maintaining extreme temperature stability [29–31]. However, the bulging of the membrane upon changing temperatures represents an important limitation [17,32,33]. As a microheater experiences higher temperatures it will start expanding, and as the movement in the in-plane directions is restricted by the silicon frame, the membrane will start to bulge out-of-plane [34,35]. This movement causes the sample to move vertically, causing the following issues:

1. The sample is moving out of the optimum focus. Even though this may be compensated by manually adjusting the focus, the highest resolution can only be obtained with the sample at eucentric position [36]. Especially for spherical aberration (Cs) corrected STEM, a change in *z*-height in the order of micrometres compensated with focus effectively cancels out the benefits of corrected optics [37]. The vertical displacement of the sample could also be counteracted using the stage, but this slow and tedious correction would result in undesired stage drift [38].
2. In dynamic experiments the TEM operator is forced to constantly adjust the image focus or move the stage as the temperature changes. Any event taking place during these adjustments cannot be captured. Fast dynamics such as quenching or rapid annealing thus cannot be recorded [39].
3. When the sample carrier is tilted to an angle (for example to facilitate tomography or zone-axis alignment), bulging creates an additional undesired sample movement that appears as a lateral shift.
4. For parallel beam diffraction experiments (e.g. nanobeam), bulging causes an undesirable change in the diffraction pattern focus if the beam is not perfectly parallel, complicating quantitative analysis [40].

A second fundamental performance indicator is the spatial stability of the sample in the horizontal plane which relates to both the displacement after a temperature step and the sample drift rate once thermal stability has been reached. It is important to minimize the displacement to prevent losing the region of interest during a temperature change and to reduce the time to image. Similarly, minimal drift is important to ensure the highest resolution and stable imaging when longer exposure times are needed (in case of for example beam sensitive samples), and is regarded as an important quality measure of *in situ* microheaters [17,20,22]. Both displacement and drift are largely influenced by the thermal expansion of the system in its entirety, and therefore by the power consumption of the device. By minimizing the power dissipated in the heater, the silicon frame and the holder will experience less thermal expansion, therefore displacement and drift are reduced [41].

EDS analysis is a valuable addition to many TEM experiments, especially when combined with high angle annular dark field (HAADF) STEM imaging to obtain chemical mapping with atomic resolution [42]. However, EDS analysis is limited in temperature range due to the infrared radiation emitted by the heater, to which the EDS detector is sensitive. To the best of our knowledge, no S/TEM EDS data were reported at temperatures higher than 700 °C [23,24,43–45].

A fourth concern is the thermal gradient across the spiral, which is limiting the effective area of homogeneous temperature [22,26]. Together with the inaccuracies that are introduced by the calibration procedure of the microheater, inhomogeneity limits the temperature accuracy that can be guaranteed at the sample. As the temperature is controlled using the resistance of the entire microheater, only a single temperature is measured, which is the temperature at the central windows. Ideally, the viewable area that is at homogeneous temperature is maximized.