In Situ Observation of Carbon Nanotube Layer Growth on Microbolometers with Substrates at Ambient Temperature

Vojtěch Svatoš,¹ Imrich Gablech,¹ B. Robert Ilic,² Jan Pekárek,¹ and Pavel Neužil¹,³

¹ Brno University of Technology, Technická 3058/10, 616 00 Brno, Czech Republic
² Center for Nanoscale Science and Technology, National Institute of Standards and Technology, Gaithersburg, MD, 20899, USA
³ Northwestern Polytechnical University, 127 West Youyi Road, Xi’an, Shaanxi, P.R. China

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Abstract
Carbon nanotubes (CNTs) have near unity infrared (IR) absorption efficiency, making them extremely attractive in IR imaging devices. Since CNT growth occurs at elevated temperatures, integration of CNTs with IR imaging devices is challenging and has not yet been achieved. Here we show a strategy for implementing CNTs as IR absorbers using differential heating of thermally-isolated microbolometer membranes in a C₂H₂ environment. During the process, CNTs were catalytically grown on the surface of a locally-heated membrane while the substrate was maintained at an ambient temperature. CNT growth was monitored in situ in real time using optical microscopy. During growth, we measured the intensity of light emission and the reflected light from the heated microbolometer. Our measurements of bolometer performance show that the CNT layer on the surface of the microbolometer membrane increases the IR response by a factor of (2.3 ± 0.1) (mean ± one standard deviation of the least-squares fit parameters). This work opens the door to integrating near unity IR absorption, CNT-based, IR absorbers with hybrid complementary metal-oxide-semiconductor focal plane array architectures.
Introduction

Infrared (IR) imaging in the wavelength range from 8 µm to 14 µm, also known as thermal imaging, has found widespread applications spanning disparate fields of engineering, life and physical sciences. Room-temperature and cooled thermal imagers have been used in preventive maintenance, night vision, medical imaging, security and space applications. Heat sensing, uncooled microbolometers in the form of focal plane arrays (FPAs) represents a class of integrated, two-dimensional ensemble thermal detector arrays with multiplexed readout. In contrast to cryogenically-cooled devices, uncooled IR microbolometers have numerous advantages including higher reliability, reduced power consumption and smaller physical dimensions. Modern microbolometer devices, operating at room temperature, are based on thermally isolated membrane architectures with integrated temperature sensors. The incident radiation, absorbed as heat, increases the temperature of the microbolometer membrane. The temperature change (ΔT<sub>IR</sub>), proportional to the power of incident radiation (P<sub>IR</sub>), is given by:

\[
\Delta T_{IR} = \frac{P_{IR}}{G},
\]

where \( G \) is thermal conductance of the microbolometer.

The FPA imagers are mass produced using Si-based complementary metal-oxide-semiconductor (CMOS) architectures. The imager consists of an array of pixels, with each pixel connected to a read-out integrated circuit. The microbolometer pixels are created using microelectromechanical systems (MEMS) technology. The microbolometer membranes are typically made from various CMOS-compatible materials such as silicon dioxide (SiO<sub>2</sub>), silicon nitride, amorphous Si and SiGe. Mid IR absorption (\( \eta \)), in range of interest from 8 µm to 14 µm of these materials, depends on the film thickness and composition. For instance, \( \eta \) for SiO<sub>2</sub> with thickness of \( \approx 0.6 \) µm ranges from \( \approx 35 \) % to \( \approx 48 \) %. The key challenge is then to maximize \( \eta \) while using a CMOS compatible process. Several developed strategies to increase \( \eta \) suffer from a significant increase in processing costs and complexity.

The first reported IR absorbing carbon nanotube (CNT) layer was implemented on the surface of a pyroelectric detector sparking an interest in CNTs as an electromagnetic radiation absorber. Controlled placement of vertically-aligned CNT layers require growth in a chemical vapor deposition (CVD) reactor at elevated temperatures above 550 °C, using lithographically patterned catalytic materials. The highest temperature that CMOS devices can withstand is typically below 475 °C. This value represents the eutectic temperature of the commonly used Al-Si contacts in
CMOS chips. Modern CMOS devices with titanium silicide, tantalum nitride, nickel silicide and other barrier materials between aluminum and silicon can be heated to higher temperatures. Nevertheless, exposing the CMOS chips to a temperature of 550 °C required for CNT growth is undesirable, thereby making the CNT growth on the surface of CMOS chips a major integration challenge. Shulaker et. al.\textsuperscript{11} show a complex, thermal release tape process for transferring CNTs from one substrate to another. This method is not suitable for mechanically suspended devices such as MEMS-based microbolometer FPAs.

Here we demonstrate a spatially-localized Joule heating technique, warming up only the bolometer membrane, for Fe-based catalytic CNT growth on the surface of the bolometer membrane while the rest of the substrate remains at room temperature. This method enables the integration of a highly efficient, IR-absorbing CNT layer onto existing monolithic CMOS FPA imagers.

Results and Discussion

Microbolometer design and fabrication

Using the Nanolithography Toolbox\textsuperscript{12}, we designed 40 \(\mu\text{m} \times 40 \mu\text{m}\) microbolometer membranes\textsuperscript{4} using a fabrication flow outlined in Fig. 1. Microbolometers, with an embedded Ti resistive temperature detector (RTD) and Fe layer for catalytic CNT growth, were fabricated using conventional Si bulk-micromachining techniques. Prior to membrane release, we measured the RTD resistance and the temperature coefficient of resistance (\(\alpha\)) using a probe station with a heated wafer chuck. The corresponding measured ambient temperature resistance (\(R_0\)) and \(\alpha\) values were (7.726 \(\pm\) 0.250) k\(\Omega\) and (0.53 \(\pm\) 0.01) mK\(^{-1}\), both represent (mean \(\pm\) standard deviation measured from), respectively. The experimental procedure and uncertainties are described in the supplementary section 1. Membranes were then released by removing the underlying Si using XeF\(_2\) vapors. Subsequently, CNT growth was carried out on individual chips.

Microbolometer characterization

Following membrane release, microbolometer chips were mounted onto a leadless chip carrier with 68 pads and electrically connected with gold wires using wire-bonding. Packaged bolometer devices were then placed into a vacuum chamber with a Ge window. Using a method described by Gu et. al.\textsuperscript{13}, at a pressure of \(\approx 7.7 \times 10^{-4}\) Pa and an applied voltage amplitude (\(V_b\)) of \(\approx 0.6\) V with
known values of $R_0$ and $\alpha$, we determined the thermal capacitance ($H$), $G$ and thermal time constant ($\tau = H/G$). Our measurements over three sets of bolometer devices show $H = (3.113 \pm 0.009) \ \text{nJ} \cdot \text{K}^{-1}$, $G = (160.9 \pm 2.7) \ \text{nW} \cdot \text{K}^{-1}$ and $\tau = (19.35 \pm 0.34) \ \text{ms}$ (mean ± standard deviation). The experimental procedure and uncertainties are described in the supplementary section 2. At a constant pressure of $\leq 5 \ \text{Pa}$ and ambient temperature of $\approx 25 \ \text{°C}$, the convective heat transfer and radiation loss are negligible and consequently, $G$ is solely determined by the thermal conduction of the microbolometer legs ($P_C$). Once the microbolometer membrane warms up by the dissipated Joule heat ($P_J$), the radiation heat loss ($P_R$), based on the Stefan-Boltzmann law, cannot be neglected. The amplitude of $P_J$, distributed between $P_R$ and $P_C$, is given by:

$$P_J = P_R + P_C = A \cdot \varepsilon \cdot \sigma \cdot T^4 + G \cdot \Delta T,$$

where $A = 2 \cdot a^2$ is the top and bottom area of the square microbolometer membrane of length $a$, $\varepsilon$ is the emissivity of the microbolometer membrane, $\sigma$ is the Stefan-Boltzmann constant, $T$ is the thermodynamic temperature, and $\Delta T$ is the temperature difference between the microbolometer membrane and the substrate chip. We measured the bolometer response to a square wave pulse over a range of $V_b$ values (Fig. S4A in supplementary section 3). In the vicinity of $V_b = 0$, where self-heating effects are neglected, our calculations show $G = (180.4 \pm 1.2) \ \text{nW} \cdot \text{K}^{-1}$, $H = (3.602 \pm 0.024) \ \text{nJ} \cdot \text{K}^{-1}$ and $\tau = (19.96 \pm 0.02) \ \text{ms}$ (all are mean ± fitting error). Here and below, the fitting error corresponds to one standard deviation of the least-squares fit parameters. The values were in good agreement with the ones determined at $V_b \approx 0.6 \ \text{V}$. Collectively this data shows that self-heating plays a minor role in the bolometer performance.

**Monitoring of CNT growth**

The packaged bolometer devices were then placed into a vacuum chamber equipped with a glass window, tubes for gas and vacuum connections, and an electrical feedthrough, as schematically shown in Fig. 2A. The chamber was then placed under an optical microscope equipped with $\approx 9 \ \text{mm}$ working distance, $50 \times$ objective with a numerical aperture (NA) of 0.55. The $\approx 7 \ \text{mm}$ distance between the bolometer membrane and the glass window allowed for high-resolution, *in situ* observation of the subsequent CNT growth processes. Thermal conditioning of the catalytic layer was accomplished by heating the microbolometer using
$P_J \approx 200 \, \mu W$ in the 5 % H$_2$ + 95 % N$_2$ forming gas (FG) environment for $\approx 15$ min. To determine the Joule heat threshold required for growth, we increased $P_J$ until the onset of CNT growth. Further increase of $P_J$ revealed an upper limit of 750 $\mu W$ above which the bolometer membrane was physically damaged from the excessive power dissipation. To circumvent the membrane damage, $P_J$ was set to 650 $\mu W$. Using a closed-loop feedback configuration, $P_J$ was maintained by the controlling voltage $V_{BOL}$ while measuring the current $I_{BOL}$ through the microbolometer resistor.

During CNT growth, we maintained a chamber pressure of $\approx 5$ Pa by adjusting partial pressures of individual gases at $\approx 1.5$ Pa and $\approx 3.5$ Pa for FG and C$_2$H$_2$, respectively. Under the above conditions, the CNT thickness was controlled by growth time. Following CNT growth, surface passivation was not carried out. Fig. 2B and 2C show optical micrographs of the microbolometer membrane before and after growth, at $\approx 65$ % of the field of view, respectively. The typical microbolometer membrane with grown CNTs is shown in Fig. 3A and 3B. A video (Figure 2D) (Multimedia view) shows real time CNT growth on the surface of the bolometer membrane.

We used finite element modelling (FEM) to show the temperature distribution within the thermally isolated membrane and the surrounding substrate. For the computation, we considered a silicon volume of 160 $\mu m \times 180 \, \mu m \times 75 \, \mu m$ with a suspended bolometer membrane and materials of dimensions resembling the fabricated devices. We used 20-node thermal solid elements for the Si substrate and 10-node tetrahedral thermal solid elements for Ti and SiO$_2$ layers. The initial boundary condition for the Si substrate was set to 25 °C. Directly following, we applied a voltage of 0.8 V between the ends of the two Ti metal traces. The FEM results (Figures 3C and 3D) show heat concentration within the suspended membrane with the substrate at approximately room temperature.

The measured photocurrent ($I_p$) from the silicon photodiode during the CNT growth was related to the emitted radiation from the heated membrane (Fig. 4A, supplementary section 5). During the measurements, the microscope light source was turned off. $I_p$ was converted into a voltage ($V_p$) using a current preamplifier with an active low-pass filter. The amplitude of $V_p$ was monitored using an oscilloscope. $I_p$ as function of time ($t$) during the growth phase, initiating at $t \approx 112$ s, can be approximated by a first order exponential decay in combination with a linear decay:

$$I_p = I_0 + I_1 \cdot e^{-\frac{t-t_0}{\tau_e}} + I_2 \cdot t,$$  \hspace{1cm} (3)
where \(I_0\) is the steady state photocurrent offset, \(I_1\) is the maximum photocurrent, \(t_0\) is time at the onset of amplitude decay (\(\approx 112\) s in Fig. 4A), \(\tau_e\) is the emission time constant, and \(I_2\) is the coefficient of linear \(I_p\) decay. Our measurements show values of \(\tau_e = (34.91 \pm 1.26)\) s and \(I_2 = (-15.94 \pm 2.02)\) fA⋅s\(^{-1}\), both values are (mean ± one standard deviation measured from 3 devices) (Fig. 4A). The uncertainties are calculated from the functional fitting error using Eq. (3). Details of the measurement results are in the supplementary section 4. The value of \(\tau_e\) can serve as a guide to minimize the CNT growth time with saturated absorbance value.

Since the amplitude of \(I_p\) is proportional to \(P_J\), the exponential decay of the \(I_p\) as a function of \(t\) can be derived from Eq. (2), while accounting for the geometric change of the radiation emitting area during CNT growth. The \(G\) value of a microbolometer is determined by the cross-sectional areas, length and thermal conductivity of the two microbolometer legs. Since the CNT layer was grown from the lithographically-defined catalyst spanning the square shaped microbolometer membrane with an area of \(A\) (see Fig. 1G and 1H), the value of \(G\) remains constant during CNT growth.

During the growth, since \(P_J\) was held constant at \(\approx 650\) µW, terms \(A\) and \(G\) in Eq. (2) can be also considered as constant. Consequently, the CNT layer on the surface of the membrane increased \(\varepsilon\) and decreased \(T\). With increasing \(\varepsilon\), the membrane emits radiation in all directions thereby lowering the temperature. Consequently, the power emitted per unit area of the microbolometer membrane decreases (supplementary section 5). Measured resistance during the CNT growth allowed a qualitative determination of the microbolometer temperature change \(\Delta T\):

\[
R_b = \frac{V_{\text{BOL}}}{I_{\text{BOL}}}; \quad R_b = R_0 \left( 1 + \alpha \cdot \Delta T \right); \quad \text{and} \quad \Delta T = \frac{1}{\alpha} \left( \frac{V_{\text{BOL}}}{R_0} - 1 \right),
\]

where \(R_0\) and \(\alpha\) are the corresponding ambient resistance at \(T \approx 298\) K and temperature coefficient of resistance of the temperature sensor, respectively. The \(\Delta T\) amplitude dropped once \(P_J\) reached an amplitude of \(\approx 650\) µW at the onset of CNT growth.

Above we assumed a negligible emission contribution from the membrane sidewalls, and considered the source of emission solely from \(A\). However, during CNT growth, the total surface area as a function of the CNT layer thickness \((t_c)\) increases by a factor of \(4 \cdot a \cdot t_c\) (supplementary section 6) while the sidewall emission increases with \(t_c\). This increase is not negligible and consequently results in a decrease of \(I_p\) at a rate of \((-15.70 \pm 1.38)\) fA⋅s\(^{-1}\) (mean ± fitting error) (Fig. 4A).
Using a ≈100 W tungsten lamp, we illuminated the bolometer membrane and measured the reflectance during CNT growth (Fig. 4B and supplementary section 7). The dynamics of the reflection-based experiment differed from the emissivity measurement. Once the CNT growth initiates at $P_J \approx 420 \mu W$, the amplitude of light reflected from the membrane decreased following an exponential decay of the first order with time constant $\tau_r = (13.44 \pm 0.07) s$ (mean ± one standard deviation from 3 devices). The faster $\tau_r$, in comparison $\tau_e$, shows that the related reflection from the membrane in visible spectrum of electromagnetic radiation is sensitive to small changes in surface properties, whereas a much thicker layer of CNTs is required to cause significant changes to $\tau_e^6$.

Inserting the parameters obtained from the above microbolometer characterization into Eq. (2) allowed us to calculate the amplitude of $P_J$ as a function temperature, without the CNT layer and for various CNT thickness values up to 30 μm (Fig. 5A). In our calculations, we used $\varepsilon_{SiO_2} = 0.48$ and $\varepsilon_{CNT} = 1$ for the emissivity of membranes without$^4$ and with$^{11,15}$ CNTs, respectively. Furthermore, the area of the CNT membrane was $A = 2 \cdot a^2 + 4 \cdot a \cdot t_c$ (supplementary section 6). The results in Fig. 5A show that the $P_J$ amplitude required to heat up bolometer membrane without and with the CNT layer to the same value of $T$ increases with increasing thickness of the CNT layer.

We then calculated the power of emitted radiation $P_{ER} = b \cdot \varepsilon \cdot \sigma \cdot T^4$ from a 1 μm$^2$ area ($b$) of the bolometer membrane without CNTs and for various CNT layer thicknesses. Fig. 5B shows a significant emission dependence with respect to the CNT layer thickness.

Experimental data in Fig. 6 show that membranes with CNTs are cooler and emit $\approx 0.23$ of the power emitted by membranes without CNTs. Our results further show that the CNT layers increased the value of thermal capacitance $H$ by a factor $\approx 5$, to $(14.83 \pm 0.53)$ nJ·K$^{-1}$ (mean ± standard deviation, 3 measurements).

**CNT layer performance**

We placed the bolometer, connected in a balanced Wheatstone bridge configuration, into a vacuum chamber and evaluated the $P_{IR}$ response. The amplitude of $P_{IR}$ was controlled by distance ($D$) between black body at $T \approx 673.15$ K and bolometer from $D \approx (59.5, 74.5, 89.5, 104.5$, and 119.5) mm. The $P_{IR}$ was estimated using$^4$: 

\[
\text{CNT layer performance}
\]
\[ P_{IR} = a^2 \cdot \varepsilon_{bb} \cdot \tau_F \cdot M \left[ \frac{\left( \frac{d}{2} \right)^2}{D^2 + \left( \frac{d}{2} \right)^2} \right], \quad (5) \]

where \( \varepsilon_{bb} \) is the emission of black body with amplitude of \( \approx 0.95 \) (manufacturer’s datasheet value), \( \tau_F \) is the average experimentally determined transmittance of the Ge window with value of \( \tau_F \approx 0.76 \), \( M \) is the integrated radiant exitance of black body (supplementary section 8) according to Planck’s radiation law with an amplitude of \( \approx 2588 \, \text{W} \cdot \text{m}^{-2} \) and \( d \approx 10.5 \, \text{mm} \) is the diameter of the black body radiation source. The value of \( \tau_F \) was determined by a ratio of an output signal of a commercial microbolometer based IR camera with and without the Ge window. The microbolometer responsivity to IR radiation \( (\Re_{IR}) \) is defined as:

\[ \Re_{IR} = \frac{\Delta V_{\text{out}}}{P_{IR}}. \quad (6) \]

Our results show that, by adding the CNT absorbing layer, the \( \Re_{IR} \) increased by a factor of \( (2.3 \pm 0.1) \) (mean \( \pm \) fitting error) (Fig. 7).

**Conclusions**

Our work presented here, for the first time demonstrates CNT growth on the surface of suspended microbolometer membranes while keeping the device substrate at room temperature. By taking advantage of the thermally isolated nature of the released microbolometer membrane, the developed CMOS fabrication compatible method allows for integration of IR absorbing CNTs with variety of active device architectures including FPAs. Our results show \( \approx 2.3 \) fold IR responsivity increase with grown CNT absorbers and reached the \( \Re_{IR} \) of \( (16.35 \pm 0.48) \, \text{kV} \cdot \text{W}^{-1} \). Furthermore, the IR absorption near unity is determined solely by the CNT layer regardless of the membrane material and thickness, cavity and underlying IR reflector. The promising growth technique of highly absorbing CNT layers using spatially localized heating on the surface of suspended membranes opens opportunities for complex integration of these absorbing layers for efficient IR imaging and FPA emission applications.

**Supplementary materials**

See supplementary materials for microbolometer resistance and temperature coefficient of resistance measurement, evaluation of microbolometer thermal parameters, microbolometer response to varying \( V_b \), details of emission measurement during CNT growth, resistance measurement during CNT growth, emission measurement from the microbolometer with CNTs,
reflection measurement from bolometer membrane during CNT growth and calculation of blackbody radiant exitance.

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References

Fig. 1 Fabrication process flow of the MEMS based microbolometer. (A) Deposition of ≈0.25 μm plasma enhanced CVD (PECVD) SiO₂ on the Si substrate. (B) Lithographic definition of ≈0.9 μm thick Al bond pads. (C) ≈30 nm sputter deposited Ti temperature sensor definition using optical lithography and reactive ion etching. (D) Deposition of ≈0.25 μm PECVD SiO₂ for protection of metal layers. (E) SiO₂ patterning to create access to bonding pads and etch holes for the microbolometer membrane release. (F) Deposition of the catalytic Fe layer using a photoresist lifting-off process. (G) Cross section of a microbolometer chip following microbolometer membrane release using XeF₂ vapors. (H) Catalytic CNT growth on the surface of the membrane.
Fig. 2 (A) Schematic illustration of the experimental setup. The microbolometer was placed into a vacuum chamber with an inlet for gas delivery and a pumping port. Gas flow was controlled by needle valves (NV). Chamber evacuation was accomplished using a combination of a turbo molecular pump (TMP) with a backing dry vacuum pump (DP). Using an optical microscope with a 50× objective lens (OL), the optical window flange allowed for real-time monitoring of the CNT growth process. Reflectance measurements during CNT growth were accomplished using a tungsten lamp (TL) in conjunction with a beam splitter (BS) to illuminate the microbolometer membrane. Emission and reflection during CNT growth were measured using a silicon photodiode (PD). The $I_p$ was converted to a voltage using a current preamplifier (Amp) that was monitored using an oscilloscope (Osc). The $P_J$ amplitude dissipated in the microbolometer membrane was set using a closed-loop feedback system by controlling the amplitude of $V_{BOL}$ while measuring $I_{BOL}$ and keeping the $P_J$ at a predetermined value. Optical micrographs of a microbolometer (B) before and (C) after the CNT growth. The scale bars in (B), (C) and (D) represent a length of $\approx 40\, \mu m$. (D) A video (Multimedia view) showing a real-time the CNT growth at the bolometer membrane captured with microscope equipped with 50× objective lens.
Fig. 3 Scanning electron micrographs of fabricated microbolometer showing (A) a thermally isolated, undercut microbolometer membrane following CNT layer growth. (B) Zoomed-in view of the CNTs near the membrane edge. The scale bars in (A) and (B) represent lengths of ≈ 20 µm and ≈ 5 µm, respectively. FEM analysis with (C) perspective and (D) top-down views of the temperature distribution within the structure. The heat is localized within the suspended, membrane region with the surrounding Si substrate at 25 °C. The scale bars in (C) and (D) represent lengths of ≈ 40 µm. The annotations in (D) represent temperatures at various locations, denoted by the corresponding arrows, along the structure.
Fig. 4 (A) Typical optical emission from the microbolometer membrane as a function of $t$ during CNT growth. $P_J$ was gradually increased at a rate of $\approx 8 \, \mu\text{W} \cdot \text{s}^{-1}$ up to 650 $\mu\text{W}$. At $t \approx 53 \, \text{s}$ we observed light emission from the membrane. CNT growth, initiating at $t \approx 112 \, \text{s}$ with $P_J \approx 650 \, \mu\text{W}$, increased the microbolometer emissivity and thereby lowered the microbolometer membrane temperature. At $t \approx 850 \, \text{s}$, heating was switched off ($P_J = 0 \, \text{W}$) and the CNT growth ceased. Directly following, the light emission quickly decayed to zero. We determined the value of $\tau_e$ using Eq. (3) as $(32.03 \pm 0.82) \, \text{s}$ (mean ± fitting error). Solid line represents a first-order exponential decay functional fit. (B) Under similar conditions, we measured the optical reflection from the microbolometer. At $t \approx 40 \, \text{s}$, an increase from a steady state of $P_J \approx 90 \, \mu\text{W}$ caused the membrane reflectivity to drop. The CNT growth initiated at $t \approx 86.5 \, \text{s}$ with $P_J \approx 420 \, \mu\text{W}$. The newly grown CNT layer lowered the membrane reflection with a time constant $\tau_r = (13.44 \pm 0.07) \, \text{s}$ (mean ± fitting error). The maximum $P_J \approx 650 \, \mu\text{W}$ occurred at $t \approx 96 \, \text{s}$. At $t \approx 250 \, \text{s}$, the heating was switched off, following which the reflection amplitude changed marginally. Solid line represents a first-order exponential decay functional fit.
Fig. 5 (A) Calculated values of dissipated Joule heat $P_j$ as function of $T$ for microbolometer without CNTs (thick black line) and for various CNT thickness values. The CNT membranes require a larger $P_j$ to remain at a distinct $T$. (b) Calculated values of $P_{ER}$ as a function of $P_j$ for microbolometers without CNTs (thick black line) and for various $t_c$ values.
Fig. 6 (A) Measured light emission amplitude from bare (black squares) and CNT covered (red circles) microbolometer membrane as a function of dissipated $P_j$. The two insets are scanning electron micrographs of microbolometers with and without the CNT layer. Scale bars represent 40 $\mu$m. Solid lines represent a 4th order polynomial curve fitting with cubic coefficient set to zero. The radiation power amplitude of the microbolometer with the CNT is $\approx 0.23$ of the one without. Filled in markers are average values and vertical bars represent one standard deviation resulting from repeated measurements across a device. (B) Optical micrograph showing light emission from the membrane at $P_j \approx 650$ $\mu$W. The scale bar represents 40 $\mu$m.
Fig. 7 Wheatstone bridge output voltage change $\Delta V_{\text{OUT}}$ as a function of incident $P_{\text{IR}}$ for microbolometer with (red circles) and without (black squares) a CNT layer. The amplitude of $P_{\text{IR}}$ was calculated from black body–bolometer distance as per Eq. (5) and supplementary section 8. The bridge was biased with a square wave pulse of amplitude $\approx 0.6$ V and duration $\approx 200$ ms. The $\Delta V_{\text{OUT}}$ amplitude was captured at $t \approx 200$ ms. Using linear regression, the microbolometer $\mathcal{R}_{\text{IR}}$ with and without CNT was $(16.35 \pm 0.48)$ kV·W$^{-1}$ and $(7.08 \pm 0.24)$ kV·W$^{-1}$, both (mean ± standard deviation, across 3 measurement), respectively. Filled in markers are average values and vertical bars represent one standard deviation resulting from repeated measurements across three devices. Overall, with CNTs, $\mathcal{R}_{\text{IR}}$ increases by a factor of $(2.3 \pm 0.1)$ (mean ± fitting error).
SUPPLEMENTARY INFO:

Supplementary section 1: Microbolometer Resistance and Temperature Coefficient of Resistance

The microbolometer chip with an embedded Ti resistance temperature detector (RTD) was placed onto a probe station hotplate chuck and electrically connected using micromanipulators. The temperature coefficient of resistance ($\alpha$) measurements were conducted prior membranes release. This condition ensures a minimal temperature gradient within the microbolometer chip. We measured the microbolometer resistance ($R_b$) by measuring the current with an applied voltage ranging from -1 V to +1 V in increments of $\approx 0.05$ V at a set temperature. The thermodynamic temperature ($T$) was monitored using a type-K thermocouple with an accuracy of 1.0 K in the range from 298 K to 473 K. Fig. S1 shows the $R_b$ as function of $T$ for three microbolometer chips. The corresponding measured resistance at ambient temperature ($R_0$) and $\alpha$ values were performed on three different microbolometers to obtain the measurement uncertainties. The ambient temperature was $\approx 298$ K.

Fig. S1 Measured $R_b$ as a function of $T$ for three different microbolometers prior membranes release.
$R_b$, as a function of temperature change $\Delta T$ is given by,

$$R_b = R_0(1 + \alpha \cdot \Delta T).$$  \hspace{1cm} (S1)

The RTD resistance change is then defined as

$$\Delta R = R_0 \cdot \alpha \cdot \Delta T.$$  \hspace{1cm} (S2)

For a measured value of $R_0$, $\alpha$ is given by the slope of the $R_b$ vs. $T$ curve (Fig. S1):

$$\alpha = \frac{1}{R_0} \cdot \frac{dR_b}{dT}.$$  \hspace{1cm} (S3)

Supplementary section 2: Evaluation of thermal parameters

The microbolometer, connected in a balanced Wheatstone bridge configuration\(^1\), was biased by a voltage pulse of amplitude $V_b$. The Wheatstone bridge consisted of a microbolometer and three $R_0$ resistors. The output voltages $V_1$ and $V_2$ were fed into a differential amplifier (Fig. S2). The voltage difference $\Delta V_{\text{OUT}} = V_1 - V_2$ at the output was monitored with an oscilloscope.

Fig. S2 Schematic illustration showing the microbolometer as part of a balanced Wheatstone bridge. The application of $V_b$ induces heat within the membrane that in turn changes the resistance. The consequent unbalanced bridge signals were fed into a differential amplifier and monitored by an oscilloscope.

Dynamics of the thermal processes are governed by the heat balance equation:
\[
H \frac{d\Delta T}{dt} + G \cdot \Delta T = P_j,
\]
where \(t\) is the time, and \(\Delta T, P_j, H\) and \(G\) are the temperature change, Joule heat, thermal capacitance and thermal conductance of the membrane, respectively.

Solution of Eq. (S4) provides the following expression for the \(\Delta T\) as a function of the applied \(V_b\):
\[
\Delta T = \frac{V_b^2}{4G \cdot R_0} \left[ 1 - \exp \left( -\frac{t}{\tau} \right) \right],
\]
where \(\tau = H/G\) is the microbolometer thermal time constant.

The balanced Wheatstone bridge output voltages \(V_1\) and \(V_2\) can be described by equations:
\[
V_1 = \frac{R_b}{R_0 + R_b} V_b; \quad V_2 = \frac{R_0}{R_0 + R_b} V_b = \frac{1}{2} V_b.
\]

The differential voltage \(\Delta V_{\text{OUT}}\) is then given by:
\[
\Delta V_{\text{OUT}} = \frac{\Delta R}{4R_0 + \Delta R} V_b.
\]

Assuming \(4R_0 >> \Delta R\) the Eq. (S7) can be further simplified after substituting \(\Delta R\) using Eq. (S2) as:
\[
\Delta V_{\text{OUT}} = \frac{\Delta R}{4R_0} V_b = \frac{R_0 \cdot \alpha \cdot \Delta T}{4R_0} V_b = \frac{\alpha \Delta T}{4} V_b.
\]

Direct substitution of \(\Delta T\) using Eq. (S5) yields the following functional form of \(\Delta V_{\text{OUT}}\) as function of \(V_b\):
\[
\Delta V_{\text{OUT}} = \frac{\alpha V_b^3}{16G \cdot R_0} 1 - \exp \left( -\frac{t}{\tau} \right).
\]

First derivative of Eq. (S9) at \(t = 0\ s\) (Fig. S3) is inversely proportional to the value of \(H\):
\[
\frac{d\Delta V_{\text{OUT}}}{dt} = \frac{\alpha V_b^3}{16R_0 \cdot H}.
\]

\(H\) can be then described as:
\[
H = \frac{\alpha V_b^3}{16R_0 \cdot \frac{d\Delta V_{\text{OUT}}}{dt}}.
\]
Fig. S3 Measured $\Delta V_{\text{OUT}}$ as function of $t$. The slope at $t = 0$ is inversely proportional to $H$, and the amplitude of $\Delta V_{\text{OUT}}$ at the steady state is inversely proportional to $G$. Microbolometer’s $\tau$ can be calculated as $H/G$ or directly extracted from a first order exponential functional fit of the measured data.

At $t \gg \tau$, Eq. S9 gives the steady state $\Delta V_{\text{OUT}}$:

$$\Delta V_{\text{OUT}} = \frac{\alpha V_b^3}{16G R_0}$$, \hspace{1cm} (S12)

leading to an expression for $G$:

$$G = \frac{\alpha V_b^3}{16R_0 \Delta V_{\text{OUT}}}$$.

(S13)
Supplementary section 3: Microbolometer response to varying $V_b$

To highlight effects of self-heating, we performed a set of measurements as described in supplementary section 2 with variable $V_b$ ranging from $\approx 0.2$ V to $\approx 0.6$ V with values of (0.20, 0.25, 0.30, 0.35, 0.40, 0.42, 0.46, 0.48, 0.50, 0.52, 0.54, 0.56, 0.58 and 0.60) V. Fig. S4A shows the influence of $V_b$ on the Wheatstone bridge output.

![Figure S4A](image)

Fig. S4 (A) The measured Wheatstone bridge output as a function of $V_b$ with voltage pulse lengths of $\approx 200$ ms ranging from $\approx 0.2$ V to $\approx 0.6$ V. (B) The amplitude of the saturation value ($V_s$) as a function of $V_b$ was determined from the functional fit using Eq. (S14). The statistical uncertainties based on the functional fit are smaller than the data markers. The solid line represents a third order polynomial fit. Value of $V_s$ at $V_b = 0$ represents a region where self-heating effects are eliminated.

We performed curve fitting using modified Eq. (S9):

$$\Delta V_{\text{OUT}} = V_s \cdot e^{-\frac{t}{\tau}} + F_1,$$  (S14)

where the fitting coefficient $V_s = \frac{\alpha V_b^3}{16 \cdot G \cdot R_0}$ and $F_1$ is an offset caused by original Wheatstone bridge setting. Figure S4B shows the mean value of $V_s$ as a function of $V_b$. With an extrapolated value of $V_s$ at $V_b = 0$, a region where self-heating is neglected, we calculated $G = (180.4 \pm 1.2)$ nW·K$^{-1}$ (mean ± standard deviation). Using Eq. (S11) and Eq. (S14), we calculated $\tau = (19.96 \pm 0.02)$ ms (mean ± standard deviation measured from 3 devices) and the $H = (3.602 \pm 0.024)$ nJ·K$^{-1}$ (mean ± fitting error), respectively.
Supplementary section 4: Emission during CNT growth

During CNT growth, we monitored the emitted power from the heated microbolometer membrane (Fig. 3A). Fig. S5 shows the measured photocurrent from the silicon photodiode ($I_p$) for three microbolometer samples. We grew CNT layer on three samples while monitoring amplitude of microbolometer emission as shown by blue, red and black lines in Fig. S5. Using a first order exponential decay in combination with linear decay (Eq. (4)) with our measured data, we determined the emission time constant ($\tau_e$) and the coefficient of linear $I_p$ decay ($I_2$).

Fig. S5 Measured light emission from the microbolometer membrane during CNT growth. As $P_J$ increased gradually, at $t \approx 53$ s we observed the onset of light emission from the microbolometer membrane. CNT growth, initiating at $t \approx 112$ s (sample 3) with $P_J \approx 650$ $\mu$W, increased the microbolometer emissivity and thereby lowered the microbolometer membrane temperature. At $t \approx 850$ s, heating was switched off ($P_J = 0$). The CNT growth ceased and the light emission decayed to zero as the microbolometer membrane cooled. The solid line curve fit was performed using Eq. (3) from the main body of the manuscript. The extracted emission time constant and the coefficient of linear decay are (34.91 ± 1.26) s and (-15.94 ± 2.02) fA·s$^{-1}$, respectively, both values represent (mean value ± one standard deviation measured from 3 devices).
Supplementary section 5: Resistance measurement during CNT growth

Fig. S6 shows the measured light emission from the microbolometer membrane during CNT growth. Overall, the resistance decrease of the integrated Ti resistor implies a drop in the membrane temperature at the initiation of CNT growth.

![Graph showing measured light emission and resistance changes](image)

Fig. S6 Measured $I_p$ (blue circles) and $R_b$ (black squares) during the CNT growth. As predicted by Eq. (3), a decrease of $I_p$ at the onset of CNT growth (black arrow at $t \approx 75$ s) is correlated with a decrease of the microbolometer membrane temperature.
Supplementary section 6: Emission from the microbolometer with CNTs

Fig. S7 Schematic illustration representation of the emission from a microbolometer with CNT.

The emitted radiation results from the entire CNT surface ($S$). The total surface area $S$ of this cube is:

$$S = A + 4 \cdot a \cdot t_c,$$

where $A = 2 \cdot a^2$ accounts for the top and bottom areas of length $a$ and thickness $t_c$.

This omnidirectional emitted radiation from the CNT surface lowers the membrane temperature as predicted by Eq. (2). Using an objective lens with $a \approx 9$ mm working distance and a numerical aperture of 0.55, we monitored the emitted power from an area defined by a cone with top angle of $\approx 1.164$ rad, as schematically depicted in Fig. S7.
Supplementary section 7: Reflection from bolometer membrane during CNT growth

During the CNT growth, we used an incident 100 W tungsten lamp and monitored reflected light from the microbolometer membrane. Fig. 3B shows one of the measured membranes. Fig. S8 shows representative results from two microbolometer samples.

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Fig. S8 Membrane reflection measurements during two distinct CNT growth cycles. We ramped the $P_J$ from $\approx 0$ µW to $\approx 650$ µW at rate of $\approx 8$ µW s$^{-1}$. At $P_J \approx 90$ µW and $t \approx 40$ s (sample 2), we observed a drop in the membrane reflected power. The CNT growth initiated at $t \approx 86.5$ s with $P_J \approx 420$ µW. The grown CNT layer further lowered the membrane reflection with $\tau_r = (13.44 \pm 0.07)$ s (mean ± fitting error). The maximum $P_J \approx 650$ µW occurred at $t \approx 96$ s. At $t \approx 250$ s, the heating was switched off, following which the reflection amplitude changed marginally.
Fig. S8 shows an offset in time and reflected power between the two runs. Time offset is a result of a manual switching of the oscilloscope recording. The reflection amplitude depends on the precise position of the microbolometer in the field of view of the microscope. The manual placement of the microbolometer most likely caused the difference in reflected power from the microbolometer membrane.

**Supplementary section 8: Calculation of blackbody radiant exitance**

The integrated radiant exitance of black body $M$ was calculated according to the Planck’s radiation law as\(^2\):

$$M = \int_{\lambda_{\text{min}}}^{\lambda_{\text{max}}} M(\lambda) d\lambda = \int_{\lambda_{\text{min}}}^{\lambda_{\text{max}}} \left( \frac{2\pi c^2}{\lambda^5} \right) \left( \frac{\frac{hc}{\lambda^2}}{e^{\left(\frac{hc}{\lambda\kappa B T_{\text{bb}}}ight)} - 1} \right) d\lambda, \quad (S16)$$

where $\lambda$ is the wavelength of incident radiation in the range from $\lambda_{\text{min}} = 8 \, \mu m$ to $\lambda_{\text{max}} = 14 \, \mu m$, $c$ is speed of light in a vacuum ($c \approx 2.998 \times 10^8 \, \text{m}\cdot\text{s}^{-1}$), $h$ is the Planck’s constant ($h \approx 6.626 \times 10^{-34} \, \text{J}\cdot\text{s}$), $k_B$ is the Boltzmann’s constant ($k_B \approx 1.381 \times 10^{-23} \, \text{J}\cdot\text{K}^{-1}$) and $T_{\text{bb}}$ is blackbody thermodynamic temperature ($T_{\text{bb}} \approx 673 \, \text{K}$).

**Supplementary References**