Localized Joule heating produced by ion current focusing through micron-size holes

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We provide an experimental demonstration that the focusing of ionic currents in a micron size hole connecting two chambers can produce local temperature increases of up to 100 °C with gradients as large as 1 °K μm⁻¹. We find a good agreement between the measured temperature profiles and a finite elements-based numerical calculation. We show how the thermal gradients can be used to measure the full melting profile of DNA duplexes within a region of 40 μm. The possibility to produce even larger gradients using submicron pores is discussed. © 2010 American Institute of Physics. [doi:10.1063/1.3399315]

The creation of local heat sources and large thermal gradients in confined aqueous environments is a challenging problem due to the rapid diffusion of heat in water. Several solutions were proposed, such as heating micro/nanoparticles by magnetic induction or by using a focused laser beam. Local thermal gradients in microchannels were used to sort and concentrate molecules. Several approaches were developed such as thermophoresis, temperature gradient focusing, field gradient focusing, and isoelectric focusing. These techniques use high dc voltages (tens to hundreds of volts) and thermal gradients in the range of 0.01 °K μm⁻¹. The typical volumes are micro to nanoliters. In this paper, we show that a few tens of picoliters can be strongly heated by focusing an ionic current through a micron size hole in a saline solution. The resulting gradients are of the order of 1 °K μm⁻¹.

We use a custom made cell composed of two chambers separated by a 50 μm thick Teflon septum (see Fig. 1). A conical hole (40° half angle, minimum diameter r₀=7.5 μm) is punctured in the center of the septum. The ac voltage (=100Vrms, 10 kHz) is applied across the chambers using platinum electrodes. We use tris buffer, 1 M KCl, pH 7.4, of electrical conductance σ=107 mS/cm. The dc value of the electrical resistance Rₖ across the hole is 2300 Ω. The septum capacitance is estimated to Cₛ=0.4 pF. It results that the current across the chambers is mostly resistive at 10 kHz. The Joule heating power Pₛ dissipated in the hole is proportional to the mean root square current iₘₛ. We measure the local temperature profiles along the vertical hole axis in the lower chamber. The temperature is derived from the confocal detection (λ=532 nm) of the calibrated fluorescence of tetramethylrhodamine (TMR) grafted at the 5’ end of DNA oligomers.

Large enough currents iₘₛ>1.9 mA, results in buffer vaporization. Smaller values of iₘₛ lead to stationary thermal profiles within a few seconds. Figure 2(a) shows the temperature increase ΔT=r−Tₐ, where Tₛ=298 °K is the room temperature, along the vertical axis for iₘₛ=1.81 mA. Over the first 40 μm, the average temperature gradient is 1 °K μm⁻¹. As a first approximation we model the heating power Pₑff being distributed uniformly in an effective sphere of radius rₑff. The temperature then reads as follows:

\[ ΔT(r) = \frac{Pₑff}{8πKRₑff} \left( 3 - \frac{r^2}{rₑff^2} \right) \quad r < rₑff, \]

\[ ΔT(r) = \frac{Pₑff}{4πKRₑff} \quad r ≥ rₑff, \quad (1) \]

where r is the distance from the hole center and K=0.6 W m⁻¹ K⁻¹ is the thermal conductivity of water. Figure 2(a) shows the measured temperature profile for hole radius r₀=7.5 μm and power Pₛ=6.5×10⁻³ W. The best fit parameters for our model are rₑff=17.8 μm and a Pₑff =5.2×10⁻³ W. This model predicts that when r>rₑff the value of (r/r₀)ΔT(r)/ΔT(r₀) is a constant independent of r₀ and Pₛ. Figure 2(b) shows the experimental curves can be scaled this way for several values of r₀ and Pₛ provided that we take rₑff=2.4r₀.

FIG. 1. (Color online) (a) Schematic representation of the experimental setup. The two chambers are separated by a 50 μm thick Teflon septum punctured by a 7.5 μm radius conical hole (b) Close up on the hole region. (c) Model set up used for our finite element modeling. (d) Close up on the pore region. The ion current lines are represented and the map of the temperature profile is color coded.

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the heating source by a spherical source of uniform heating power 
steady-state solutions were obtained with an ac voltage at the 
current is 
triangles 
calculation. 

is the local dielectric constant, 

\( \Delta T / °C \)

\( z/r \)

\( \Delta T \)

\( \Delta T / °C \)

1/z

The dotted line is the fit obtained when modeling the heating source by a spherical source of uniform heating power \( P,=5.2 \times 10^{-3} \) W and radius \( r,=17.8 \) μm. The solid line is the finite element calculation. (b) Rescaling of all temperature profiles obtained with a hole of 7.5 μm (open circles) and 20 μm (full triangles) for various currents (0.5 mA < \( i, < 1.85 \) mA). We used \( r,=2.4 r, \). The straight line corresponds to the scaling of the analytical model. Inset: Values of \( \Delta T(r,0) \) as a function of \( I, \) for the hole of 7.5 μm (open circles) and 20 μm (full triangles).

In order to explore the influence of the pore geometry on thermal profiles we use a finite elements approach with various hole radius \( r, \) and pore lengths \( L, \) The numerical calculation accounts for mass transport (2), electrostatic potential (3), and heat dissipation (4) in the following way:

\[
\partial_t C^\pm - \nabla (D^\pm \nabla C^\pm \pm e \mu^\pm C^\pm \nabla \phi) = 0, \tag{2}
\]

\[
\partial_t (\nabla (e \nabla \phi)) + \nabla (\sigma \nabla \phi) = 0, \tag{3}
\]

\[
\rho C_p \alpha T - \nabla (\kappa \nabla T) - \sigma (\nabla \phi)^2 = 0. \tag{4}
\]

\( C^\pm, D^\pm, \) and \( \mu^\pm \) are the number densities, the diffusion coefficients, and the electrophoretic mobilities of the positive and negative ions, respectively. \( \phi \) is the electric potential, \( e \) is the local dielectric constant, \( \sigma \) is the electrical conductivity, \( e \) is the electron’s charge, and \( C_p \) is the heat capacity. The steady-state solutions were obtained with an ac voltage at the electrodes. 6

Figure 3 shows the calculated temperature \( \Delta T(z) \) and gradient profiles \( \nabla T(z) \) along the z axis at a fixed heating power \( P, \) for several values of the aspect ratio \( r, /L, > 0.1 \) the temperature is well described by \( \Delta T(z) \sim z^{-1} \) for \( z \) values larger than \( 2 r, \). In the limit of small aspect ratio the heating is mainly localized in the hole and smoothly spreads in the lower chamber producing smaller temperature gradients. We conclude that \( r, /L, \) should ideally be between 0.1 and 1 to obtain the sharpest gradients and largest temperature increases.

The experimental thermal gradients generated by a hole of diameter \( r,=20 \) μm are used to determine the melting profile of a DNA duplex: strand 1: TMR 5’–TCAGACCG(TC)15–3’, strand 2: 5’–CGGTCTGA–3’ IowaBlack. The DNA was gel purified to obtain a 95% hybridization efficiency. The fluorescence intensity of the TMR is quenched 20-fold on average upon hybridization with IowaBlack. With a proper baseline calibration the fluorescence intensity measured at the laser spot can be used to determine the fraction of hybridized duplexes. We measure the fluorescence profiles at various values of \( i, \) for strand 1 only, and for the hybridized duplex with a 1:1 ratio of both strands (see Fig. 4). Assuming (i) a local thermal equilibrium, (ii) a two state model where the eight mers are either fully hybridized or completely open, and (iii) an efficient quenching for all temperatures, we can extract the dissociation coefficient \( \alpha \) of the hairpin as a function of temperature and distance from the pore. Figure 4 shows the full melting profile obtained over a distance of 45 μm for a hole of radius \( r,=20 \) μm. Following the typical melting curve analysis for bimolecular equilibrium, 10 we extract the thermodynamical melting parameters of the DNA structure.
find $\Delta H = -66 \pm 6$ kcal mole$^{-1}$ and $\Delta S = 182 \pm 25$ Cal mole$^{-1}$ K$^{-1}$, in good agreement with the thermodynamical parameters calculated with MFOLD (Ref. 11) under similar salt conditions ($\Delta H = -62$ kcal mole$^{-1}$ and $\Delta S = 169$ Cal mole$^{-1}$ K$^{-1}$). Spatial temperature gradients have the advantage over traditional melting curves techniques that all temperatures can be probed simultaneously. This method works if the diffusion and/or drift of the DNA molecule across the thermal profile is slow enough to allow thermal equilibration. As derived in Ref. 16, electrophoretic, and electro-osmotic drifts are negligible in our experiments. The Brownian diffusion coefficient$^{12}$ for our DNA molecules is of order $D_{\text{DNA}} \approx 10^{-7}$ cm$^2$ s$^{-1}$. The distance over which the temperature changes by $1 \degree$ C is $d = 1$ $\mu$m. The diffusion time across this distance is $\pi d^2 / D_{\text{DNA}} = 0.01$ s. Since small molecular beacons are reported to open over a characteristic time of $10^{-4}$ s,$^{13}$ the approximation of local equilibrium is satisfied.

We briefly discuss the influence of various parameters on the applicability of our device. For frequencies $\omega < (R_s C_s)^{-1} = a r_0^2 / (\sigma C_s)$, where $a$ is a geometry-dependent factor, the ionic current is mostly resistive. It is reasonable to assume that electro-osmotic flow are negligible due to the use of ac fields. Electro-osmosis can be significantly enhanced in smaller ($\approx$100 nm) or charged pores when current rectification occurs.$^{14}$ In the lower chamber natural convection is minimized since the hot spot (the hole) is located above the cold region. In addition, there is negligible fluid transfer between the upper and lower chambers. The small extent of the lower chamber also increases the instability threshold for natural convection. This situation contrasts with focused laser heating for which convective rolls are observed along the optical axis.$^{7}$ Convection can be externally applied to drive the molecule through the thermal profile. Our conclusions would still hold in the limit of small Peclet numbers, i.e., if the convective velocity is small: $v < \kappa / \rho C_p r_0 = 10^{-5} / r_0$ m s$^{-1}$. In this limit thermal quenching rate of $1 \degree$ K/µs can still be achieved.$^{16}$ We believe that this approach facilitates the creation of large thermal gradients in submicron regions with potential applications for fast denaturation and thermal quenching$^{15}$ to study local chemical reactions.

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16. See supplementary material at http://dx.doi.org/10.1063/1.3399315 for a detailed discussion of the approach limitations.