

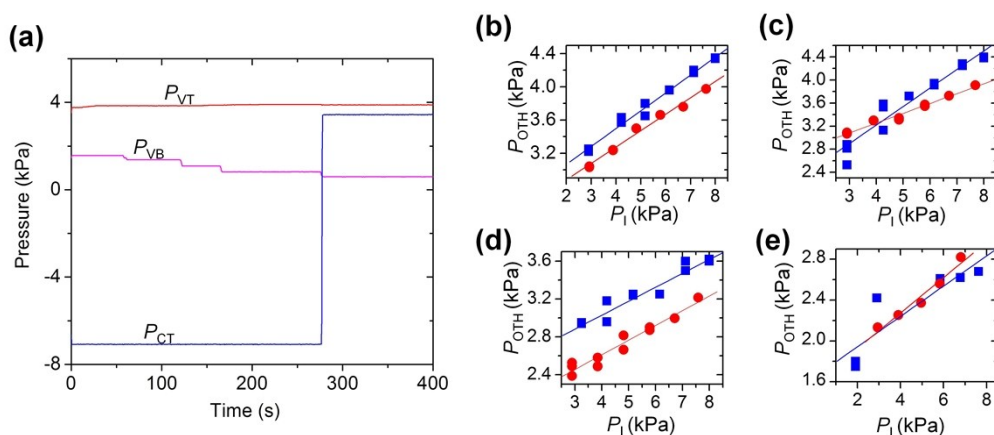
## Electronic supplementary information For

### Analyzing water-head-driven microfluidic oscillators for autonomous control of period and flow rate

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#### I. Open condition of the valve and its threshold pressure

The open condition of valve  $m$  is  $P_{VTm} - P_{VBm} > P_{OTHm}$ . Because  $P_{VTm} = P_1$  and  $P_{VBm} = P_{CBm}$ , the open condition is rewritten as  $P_{CBm} < P_1 - P_{OTHm}$ . We measured the open threshold pressure of valve  $m$  ( $m = 1, 2$ ) with the oscillator. To obtain  $P_{OTHm}$ , we measured  $P_{VTm}$ ,  $P_{VBm}$ , and  $P_{CTm}$  of the oscillator with pressure sensors (PX309-015G5V, Omega Eng). We maintained  $P_{VTm}$  at 3.7 kPa and decreased  $P_{VBm}$ . At the moment valve  $m$  opened,  $P_{CTm}$  rose rapidly (Fig. S1a).

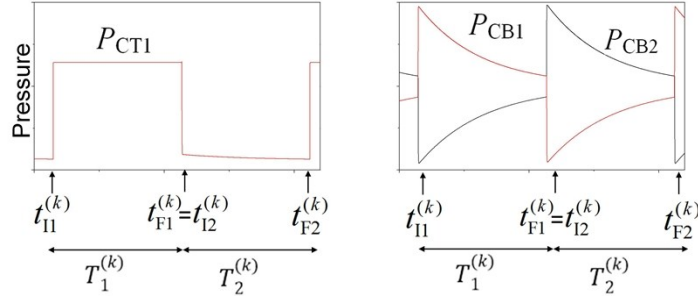


**Fig. S1.** Measurement of open threshold pressure. (a) Pressure profiles of  $P_{VB}$  and  $P_{CT}$ . (b to e) Calibration curves of  $P_{OTHm}$  with respect to  $P_1$ .  $R_F$  is  $3.2 \times 10^{11}$ ,  $1.2 \times 10^{12}$ ,  $2.5 \times 10^{13}$ , and  $1.3 \times 10^{15}$  N s m<sup>-5</sup> from (b) to (e), respectively. The blue and red lines correspond to valves 1 and 2, respectively.

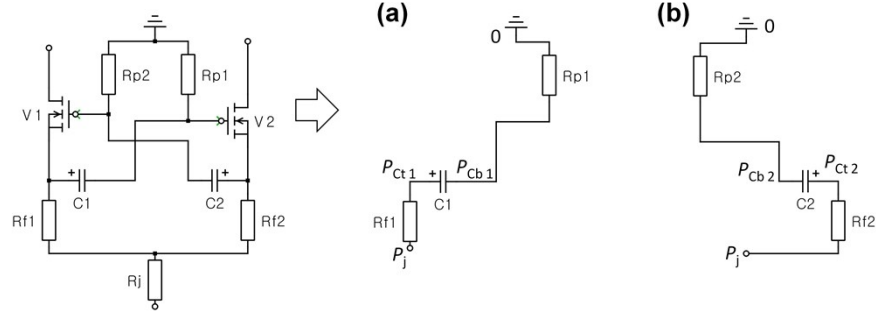
At this time, we obtained  $P_{OTHm}$  from  $P_{OTm} = P_{VTm} - P_{VBm}$ . Figures S1b to S1e show the calibration curves of  $P_{OTHm}$  with respect to  $P_{VTm}$  ( $=P_1$ ) under different  $R_F$ . From the curves of Figs. S1b to S1e, we obtained  $P_{OTH1} = -341.37 \log_{10}(R_F) + 7428.40$  and  $P_{OTH2} = -298.02 \log_{10}(R_F) + 6683.15$  at  $P_1 = 3.7$  kPa, and applied them to Figs. 4 and 5 in the main text. The unit of  $P_{OTH1}$  and  $P_{OTH2}$  is Pa.

## II. Theoretical derivation of equations for $P_{CBn}$ and $P_{CTn}$

One oscillation period includes the sequential open-process of the two valves. At the  $k^{\text{th}}$  oscillation period ( $k = 0, 1, 2, \dots$ ), valve 1 opens for  $t_{11}^{(k)} \leq t \leq t_{F1}^{(k)}$  and then valve 2 opens for  $t_{12}^{(k)} \leq t \leq t_{F2}^{(k)}$ . Figure S2 shows the pressure profiles of  $P_{CT1}$ ,  $P_{CB1}$ , and  $P_{CB2}$  at the  $k^{\text{th}}$  period. To derive equations, we used the circuit diagram that depicts the fluidic connection at the state when valve 1 is open and valve 2 is closed (Fig. S3).



**Fig. S2.** Pressure profiles of  $P_{CT1}$ ,  $P_{CB1}$ , and  $P_{CB2}$  at the  $k^{\text{th}}$  oscillation period.



**Fig. S3.** For the state when valve 1 is open, the relation between  $P_{CT1}$  and  $P_{CB1}$  is obtained in (a) and the relation between  $P_{CT2}$  and  $P_{CB2}$  is obtained in (b).

While valve 1 is open,

$$P_{CT1} = P_1. \quad (S1)$$

From Fig. S3a, we obtain

$$C_1 \frac{d}{dt} (P_{CT1} - P_{CB1}) = \frac{P_{CB1} - 0}{R_{P1}}. \quad (S2)$$

By the initial condition of  $P_{CB1}$  at  $t_{II}^{(k)}$ ,  $P_{CB1}(t)$  is

$$P_{CB1}(t) = P_{CB1}(t_{II}^{(k)}) e^{-\frac{t-t_{II}^{(k)}}{C_1 R_{P1}}}. \quad (S3)$$

From Fig. S3b, we obtain

$$\frac{0 - P_{CB2}}{R_{P2}} = C_2 \frac{d}{dt} (P_{CB2} - P_{CT2}) = \frac{P_{CT2} - P_J}{R_{F2}}. \quad (S4)$$

After removing  $P_{CT2}$  in eqn (S4),

$$-\left(\frac{R_{F2}}{R_{P2}} + 1\right) \frac{d}{dt} P_{CB2} = \frac{1}{C_2 R_{P2}} P_{CB2}. \quad (S5)$$

By the initial condition of  $P_{CB2}$  at  $t_{II}^{(k)}$ , eqn (S5) is

$$P_{CB2}(t) = P_{CB2}(t_{II}^{(k)}) e^{-\frac{t-t_{II}^{(k)}}{C_2 (R_{F2} + R_{P2})}}. \quad (S6)$$

Then from eqn (S4),

$$P_{CT2}(t) = P_J - \frac{R_{F2}}{R_{P2}} P_{CB2}(t). \quad (S7)$$

The equations for the open state of valve 2 can be derived in the same manner. Table S1 summarizes equations for the changes of pressures and duration times of each valve's openings.

**Table S1.** Equations at the open state of each valve.

Valve 1 open state: $t_{i1}^{(k)} \leq t \leq t_{f1}^{(k)}$	Valve 2 open state: $t_{i2}^{(k)} \leq t \leq t_{f2}^{(k)}$
$P_{CT1} = P_1 \quad (S1)$	$P_{CT2} = P_1 \quad (S8)$
$P_{CB1}(t) = P_{CB1}(t_{i1}^{(k)}) e^{-\frac{t-t_{i1}^{(k)}}{C_1 R_{P1}}} \quad (S3)$	$P_{CB2}(t) = P_{CB2}(t_{i2}^{(k)}) e^{-\frac{t-t_{i2}^{(k)}}{C_2 R_{P2}}} \quad (S9)$
$P_{CB2}(t) = P_{CB2}(t_{i1}^{(k)}) e^{-\frac{t-t_{i1}^{(k)}}{C_2 (R_{F2} + R_{P2})}} \quad (S6)$	$P_{CB1}(t) = P_{CB1}(t_{i2}^{(k)}) e^{-\frac{t-t_{i2}^{(k)}}{C_1 (R_{F1} + R_{P1})}} \quad (S10)$
$P_{CT2}(t) = P_J - \frac{R_{F2}}{R_{P2}} P_{CB2}(t) \quad (S7)$	$P_{CT1}(t) = P_J - \frac{R_{F1}}{R_{P1}} P_{CB1}(t) \quad (S11)$

### III. Theoretical derivation of equations of $P_{CBn}$ at times $t_{In}^{(k)}$ and $t_{Fn}^{(k)}$

We derive equations for  $P_{CB1}(t_{i2}^{(k)})$ ,  $P_{CB1}(t_{f1}^{(k)})$ ,  $P_{CB2}(t_{i1}^{(k)})$ , and  $P_{CB2}(t_{f2}^{(k)})$ . The open condition of valve 2 is  $P_{VT2} - P_{VB2} > P_{OTH2}$ . At the moment valve 2 is opened in the  $k^{\text{th}}$  period, the condition is

$$P_{VT2}(t_{f1}^{(k)}) - P_{VB2}(t_{f1}^{(k)}) = P_{OTH2},$$

with

$$P_{VT2}(t_{f1}^{(k)}) = P_1 \text{ and } P_{VB2}(t_{f1}^{(k)}) = P_{CB1}(t_{f1}^{(k)}). \quad (S12)$$

Thus,

$$P_{CB1}(t_{f1}^{(k)}) = P_1 - P_{OTH2}. \quad (S13)$$

From the continuity of pressure differences across capacitor 1,

$$P_{CT1}(t_{F1}^{(k)}) - P_{CB1}(t_{F1}^{(k)}) = P_{CT1}(t_{I2}^{(k)}) - P_{CB1}(t_{I2}^{(k)}). \quad (\text{S14})$$

From Eqs. (S1) and (S13), eqn (S14) is

$$P_{CT1}(t_{I2}^{(k)}) - P_{CB1}(t_{I2}^{(k)}) = P_{OTH2}. \quad (\text{S15})$$

When valve 2 opens, valve 1 is closed. From Fig. S2(a), we obtain:

$$\frac{0 - P_{CB1}(t_{I2}^{(k)})}{R_{P1}} = \frac{P_{CT1}(t_{I2}^{(k)}) - P_J}{R_{F1}}. \quad (\text{S16})$$

From Eqs. (S15) and (S16), we derive an equation for  $P_{CB1}$  at the moment valve 2 opens:

$$P_{CB1}(t_{I2}^{(k)}) = \frac{R_{P1}}{R_{F1} + R_{P1}} (P_J - P_{OTH2}). \quad (\text{S17})$$

Similarly, at the moment valve 1 opens in the  $k^{\text{th}}$  period, the condition of valve 1 is

$$P_{CB2}(t_{F2}^{(k)}) = P_1 - P_{OTH1}. \quad (\text{S18})$$

From the continuity of pressure differences across capacitor 2,

$$P_{CB2}(t_{I1}^{(k)}) = \frac{R_{P2}}{R_{F2} + R_{P2}} (P_J - P_{OTH1}). \quad (\text{S19})$$

Table S2 summarizes the equations of  $P_{CBn}$  at times  $t_{In}^{(k)}$  and  $t_{Fn}^{(k)}$ .

**Table S2.** Equations of  $P_{CBn}$  at times  $t_{in}^{(k)}$  and  $t_{Fn}^{(k)}$ .

$P_{CB1}$	$P_{CB2}$
$P_{CB1}(t_{F1}^{(k)}) = P_1 - P_{OTH2}$ (S13)	$P_{CB2}(t_{F2}^{(k)}) = P_1 - P_{OTH1}$ (S18)
$P_{CB1}(t_{I2}^{(k)}) = \frac{R_{P1}}{R_{F1} + R_{P1}} (P_J - P_{OTH2})$ (S17)	$P_{CB2}(t_{I1}^{(k)}) = \frac{R_{P2}}{R_{F2} + R_{P2}} (P_J - P_{OTH1})$ (S19)

#### IV. Theoretical derivation of equations for fluidic switching periods

From eqn (S3), we obtain the duration time of the opening of valve 1:

$$T_1^{(k)} = t_{F1}^{(k)} - t_{I1}^{(k)} = C_1 R_{P1} \ln \left[ \frac{P_{CB1}(t_{I1}^{(k)})}{P_{CB1}(t_{F1}^{(k)})} \right]. \quad (S20)$$

Similarly, the duration time of the opening of valve 2 can be written as

$$T_2^{(k)} = t_{F2}^{(k)} - t_{I2}^{(k)} = C_2 R_{P2} \ln \left[ \frac{P_{CB2}(t_{I2}^{(k)})}{P_{CB2}(t_{F2}^{(k)})} \right]. \quad (S21)$$

The  $k^{\text{th}}$  switching period of the two valves is  $T^{(k)} = T_1^{(k)} + T_2^{(k)}$ . Equations (S20) and (S21) show that  $T^{(k)}$  includes  $P_{CB1}(t_{I1}^{(k)})$ ,  $P_{CB1}(t_{F1}^{(k)})$ ,  $P_{CB2}(t_{I2}^{(k)})$ , and  $P_{CB2}(t_{F2}^{(k)})$ . Because we obtained  $P_{CB1}(t_{F1}^{(k)})$  and  $P_{CB2}(t_{F2}^{(k)})$  (Table S2), we need  $P_{CB1}(t_{I1}^{(k)})$  and  $P_{CB2}(t_{I2}^{(k)})$  for  $T^{(k)}$ .

At  $k = 0$ , we assume valve 1 opens and valve 2 closes. Thus,

$$P_{CB1}(t_{I1}^{(0)}) = P_{CT1}(t_{I1}^{(0)}) = P_1. \quad (S22)$$

From eqn (S4),

$$P_{CB2}(t_{I1}^{(0)}) = P_{CT2}(t_{I1}^{(0)}) = \frac{R_{P2}}{R_{F2} + R_{P2}} P_J. \quad (S23)$$

From Eqs. (S13), (S20), and (S22), the duration time of valve 1 at the 0<sup>th</sup> period is

$$T_1^{(0)} = t_{F1}^{(0)} - t_{I1}^{(0)} = C_1 R_{P1} \ln \left[ \frac{P_1}{P_1 - P_{OTH2}} \right]. \quad (S24)$$

When valve 2 opens, the continuity of pressure differences across capacitor 2 should be met:

$$P_{CT2}(t_{F1}^{(0)}) - P_{CB2}(t_{F1}^{(0)}) = P_{CT2}(t_{I2}^{(0)}) - P_{CB2}(t_{I2}^{(0)}). \quad (S25)$$

Applying Eqs. (S6)–(S8), and (S24) to eqn (S25), we obtain  $P_{CB1}$  at the moment valve 2 opens in the 0<sup>th</sup> period:

$$P_{CB2}(t_{I2}^{(0)}) = P_1 - P_J + P_J \left( \frac{P_1 - P_{OTH2}}{P_1} \right)^{a_1}, \quad (S26)$$

with

$$a_1 = \frac{C_1 R_{P1}}{C_2 (R_{F2} + R_{P2})}. \quad (S27)$$

At  $k \geq 1$ , we can obtain  $P_{CB1}(t_{I1}^{(k)})$  and  $P_{CB2}(t_{I2}^{(k)})$ .

At the moment valve 1 opens in the  $k^{\text{th}}$  period, the continuity condition of pressure difference across capacitor 1 is

$$P_{CT1}(t_{F2}^{(k-1)}) - P_{CB1}(t_{F2}^{(k-1)}) = P_{CT1}(t_{I1}^{(k)}) - P_{CB1}(t_{I1}^{(k)}). \quad (S28)$$

From eqn (S10) and (S17),

$$P_{CB1}(t_{F2}^{(k-1)}) = \frac{R_{P1}}{R_{F1} + R_{P1}} (P_J - P_{OTH2}) e^{-\frac{t_{F2}^{(k-1)} - t_{I2}^{(k-1)}}{C_1 (R_{F1} + R_{P1})}}. \quad (S29)$$

From eqns (S18) and (S21), eqn (S29) is

$$P_{CB1}(t_{F2}^{(k-1)}) = \frac{R_{P1}}{R_{F1} + R_{P1}} (P_J - P_{OTH2}) \left( \frac{P_1 - P_{OTH1}}{P_{CB2}(t_{I2}^{(k-1)})} \right)^{a_2} \quad (S30)$$

with

$$a_2 = \frac{C_2 R_{P2}}{C_1 (R_{F1} + R_{P1})}. \quad (S31)$$

From eqn (S11),

$$P_{CT1}(t_{F2}^{(k-1)}) - P_{CB1}(t_{F2}^{(k-1)}) = P_J - \left( \frac{R_{F1}}{R_{P1}} + 1 \right) P_{CB1}(t_{F2}^{(k-1)}). \quad (\text{S32})$$

Applying eqns. (S1), (S30), and (S31) to eqn (S28), we obtain:

$$P_{CB1}(t_{11}^{(k)}) = P_1 - P_J + (P_J - P_{OTH2}) \left( \frac{P_1 - P_{OTH1}}{P_{CB2}(t_{12}^{(k-1)})} \right)^{a_2}. \quad (\text{S33})$$

Similarly, we can get

$$P_{CB2}(t_{12}^{(k)}) = P_1 - P_J + (P_J - P_{OTH1}) \left( \frac{P_1 - P_{OTH2}}{P_{CB1}(t_{11}^{(k)})} \right)^{a_1}. \quad (\text{S34})$$

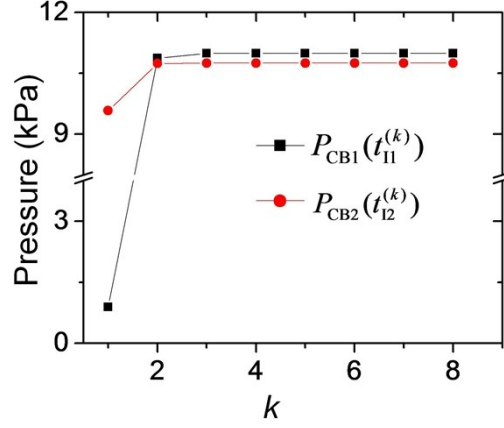
We used  $C_1 = C_2$ ,  $R_{F1} = R_{F2}$ , and  $R_{P1} = R_{P2}$  in the experiment, and  $T^{(k)} = T_1^{(k)} + T_2^{(k)}$ . Thus, the final equations for the period are summarized in Table S3.

**Table S3.** Theoretical equations for period and related pressures

$T^{(k)} = CR_p \ln \left[ \frac{P_{CB1}(t_{11}^{(k)}) P_{CB2}(t_{12}^{(k)})}{(P_1 - P_{OTH1})(P_1 - P_{OTH2})} \right]$	(S13, S18, S20, S21)
$P_{CB1}(t_{11}^{(k)}) = P_1 - P_J + (P_J - P_{OTH2}) \left( \frac{P_1 - P_{OTH1}}{P_{CB2}(t_{12}^{(k-1)})} \right)^a$	(S33)
$P_{CB2}(t_{12}^{(k)}) = P_1 - P_J + (P_J - P_{OTH1}) \left( \frac{P_1 - P_{OTH2}}{P_{CB1}(t_{11}^{(k)})} \right)^a$	(S34)
with $a = \frac{1}{1 + R_F / R_p}$	(S27, S31)
and $P_{CB2}(t_{12}^{(0)}) = P_1 - P_J + P_J \left( \frac{P_1 - P_{OTH2}}{P_1} \right)^a$	(S26)



## V. Convergence of $P_{CB1}(t_{11}^{(k)})$ and $P_{CB2}(t_{12}^{(k)})$

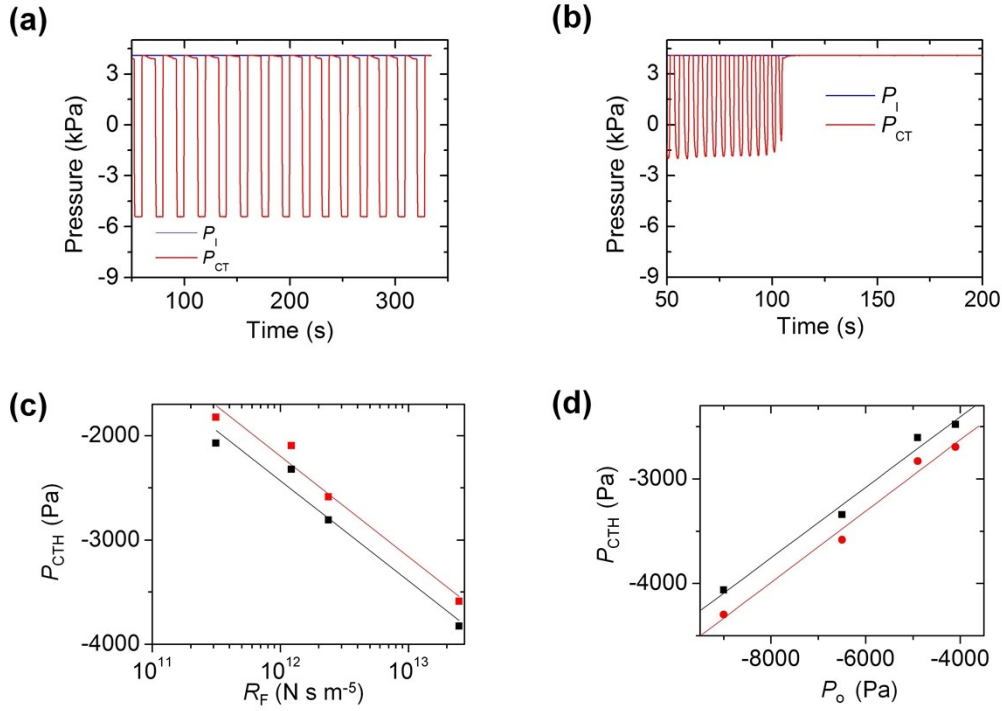


**Fig. S4.**  $P_{CBn}(t_{ln}^{(k)})$  converges at  $k = 3$ . Here,  $P_I$  and  $P_O$  are 3.7 and  $-8.5$  kPa, respectively, and  $R_F$  and  $R_P$  are  $1 \times 10^{13}$  and  $5 \times 10^{13}$  N s  $m^{-5}$ , respectively.

## VI. Closed threshold pressure

We obtained the closed threshold pressures of valve  $m$  ( $P_{CTHm}$ ) with working oscillators. To obtain  $P_{CTHm}$  for Figs. 4 and 5, we used  $R_P = 6.1 \times 10^{13}$  N s /  $m^5$  and  $P_I = 3.7$  kPa.  $R_F$  and  $P_O$  are variables. For example, when  $R_F$  is  $2.4 \times 10^{12}$  N s /  $m^5$  and  $P_O$  was increased from  $-8.5$  to  $-3.3$  kPa, the oscillation stopped at  $P_O = -3.3$  kPa (Figs. S2a and S2b). As  $P_O$  increased,  $P_{CBn}(t_{ln}^{(k)})$  decreased and the condition of eqn (4) in the main text was not satisfied at  $P_O = -3.3$  kPa. At such a critical  $P_O$ ,  $P_{CBn}(t_{ln}^{(k)})$  is equal to  $-P_{CTm} + P_I$ . This can be rewritten as  $P_{CTn} = -P_{CBn}(t_{ln}^{(k)}) + P_I$ . Here,  $P_{CBn}(t_{ln}^{(k)})$  is obtained from eqn (2) in the main text, and  $k$  is  $\geq 3$ . As we changed  $R_F$ , calibration curves of  $P_{CTHm}$  for Figs. 3 and 4 were obtained, as shown in Fig. S5c, where  $P_{CTH1} = -418.7\ln(R_F) + 9372.5$  and  $P_{CTH2} = -416.9\ln(R_F) + 9086.6$ .

To obtain  $P_{CTHm}$  for Fig. 5, we used  $R_p = 6.1 \times 10^{13}$  and  $R_F = 2.4 \times 10^{12} \text{ N s m}^{-5}$ .  $P_I$  and  $P_O$  are variables. Figure S5d shows the calibration curves:  $P_{CTH1} = 0.3371 P_O - 1057.9$  and  $P_{CTH2} = 0.3412 P_O - 1261.1$ .



**Fig. S5.** Measurement of closed threshold pressure. (a and b) Pressure profiles of  $P_{CTm}$ .  $P_O$  is  $-8.5$  kPa in (a) and  $-3.3$  kPa in (b). (c) Calibration curves of  $P_{CTHn}$  with respect to  $R_F$ . (d) Calibration curves of  $P_{CTHm}$  with respect to  $P_O$ . The black and red lines correspond to valves 1 and 2, respectively, in (c) and (d).