

Supporting information

Programs on UNIX environment for QCM-D

In this study we made programs for treating 2M data. These programs were useful for not only QCM-D but also analyzing damping function which has large data set measured by any sensor devices.

Fig S1 (a) was the raw data of damping plots of Type (iii). These figures were revised as bellow. Fig S1 (b) showed the cycle related to the difference between the QCM period (100.xxx ns) and the sampling period of the oscilloscope (80–400ns). This difference created another cycle. In this case, the sampling period was 80 ns and the other cycle was 100 μ s. We chose maximum values from each cycle in program 1 for calculation constant V_{max} . Fig S1 (c), the voltage was larger than V_{max} (obtained from Program1) and was used for estimating “T” which is the difference between the origin and the damping start time. Then Program 2 adjusted the time origin to the damping start time. Fig S1 (d) showed modified figure through program 1, 2 and 3. The decay time constant τ was calculated when the voltage was V_{max}/e . The programs for calculating “ τ ” from above explanation were constituted as follows.

```
<Program1 >
$ tail -2000000 QCMDdata.csv > QCMDdata2.csv // (1)
$ Split QCMDdata2.csv -l 1250 newfiles // (2)
$ awk -F, '{print $2}' newfiles** | sort -gr | head -n 1352 > Vmaxs // (3)
$ awk 'BEGIN {} { x+= $1 } END { print x/NR }' Vmaxs // (4)
<Program2 >
$ xinit // (5)
$ gnuplot // (6)
gnuplot> set datefile separator "," // (7)
gnuplot> plot "QCMDdata2.csv" using 1:2 // (8)
$ awk -F, '{ print $1 - T, $2 }' QCMDdata2.csv > QCMDdata3.csv // (9)
<Program3 >
$ awk 'if( $2 > Vmax/e ) print $1' QCMDdata3.csv | tail -n1 > QCMDdata4.csv // (10)
```

(1) The QCMDdata.csv files (2M csv file data) used seventeen lines for documentation. Those lines were deleted and the remaining 2M lines were saved as QCMDdata2.csv. (2) When $t < 0$, the data was separated by 1250 plots and 676 new files were made to check for periodicity. (3) The data were sorted in descending order and the upper 1352 points were chosen. (4) The average value was calculated and V_{max} value was obtained. (5), (6) Xinit was started on Cygwin and Gnuplot was started for plotting. (7) The csv format was separated with commas; that is why the separation mode is a comma. (8) The damping graph was plotted from column1 and column2 data. A maximum value of voltage from the entire area was chosen and was shifted with respect to the QCM turn-off time (start time of damping); the difference of origin and turn-off time was named as “T”. (9) Using the calculated Column1 $\pm T$, the damping start time was set to 0 sec, and QCMDdata2.csv was changed to QCMDdata3.csv. (10) For the values from column1 which correlated with Column2 $> V_{max}/e$, the last value was τ , which was stored in QCMDdata4.csv.

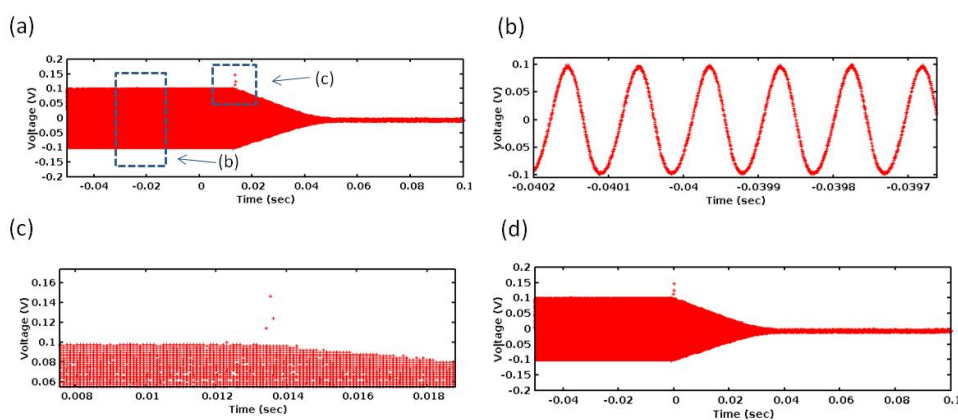


Fig S1. QCM-D (treated with program1, 2 and 3). (a) A Type (iii) film was measured by QCM-D and the UNIX system. (b) This cycle is a function of the difference of the QCM period (100.xxx ns) and the sampling period (80–400 ns). We chose maximum values from each cycle in program1. (c) At the damping start time, the voltage was larger than V_{max}

because the QCM had just been turned off. (d) After adjusting decay start time by program 2, the decay time constant τ was calculated when the voltage was V_{max} / e by program 3.

GDOES raw data

When we measured the elements of Si, Ti and C, the peak of Ti was observed even if there was not Ti for example substrate regions. That is why we used the minimum voltage value of Ti for base plot.

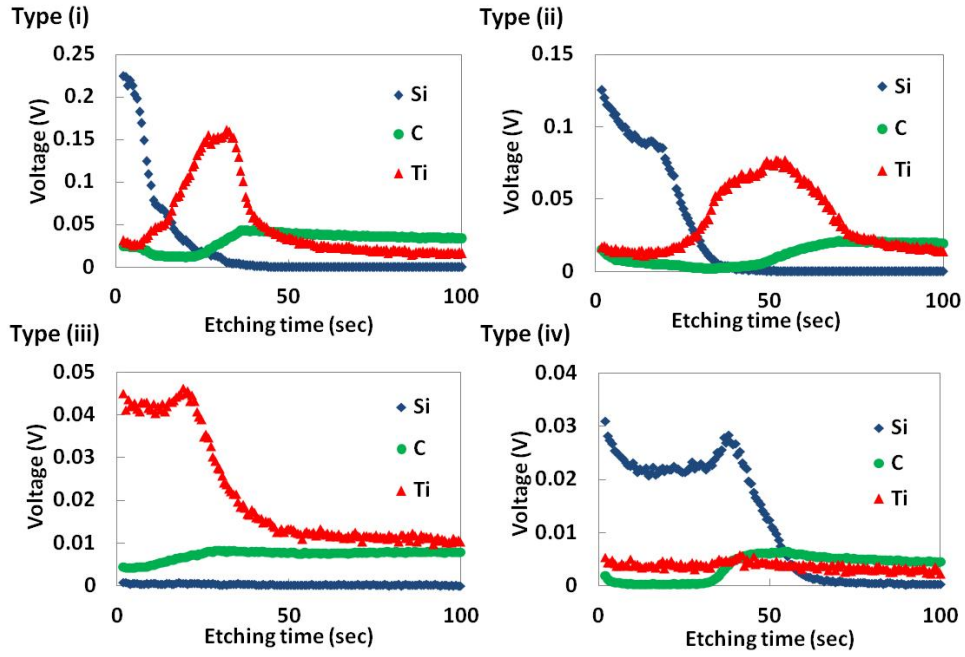


Fig S2. Raw data of GDOES for Type (i), Type (ii), Type (iii) and Type (iv).