

Fig. S1 (a) Bright-field (BF) STEM image of 500 CV-Pt. (b) TEM-EDS mapping image shows flower-like morphology belongs to Pt of 500 CV-Pt. (c) EDS map spectrum of 500 CV-Pt. Scale bar : 200 nm. (d) Bright-field (BF) STEM image of CP-Pt. (e) TEM-EDS mapping image shows flower-like morphology belongs to Pt of CP-Pt. (f) EDS map spectrum of CP-Pt. Scale bar : 200 nm.

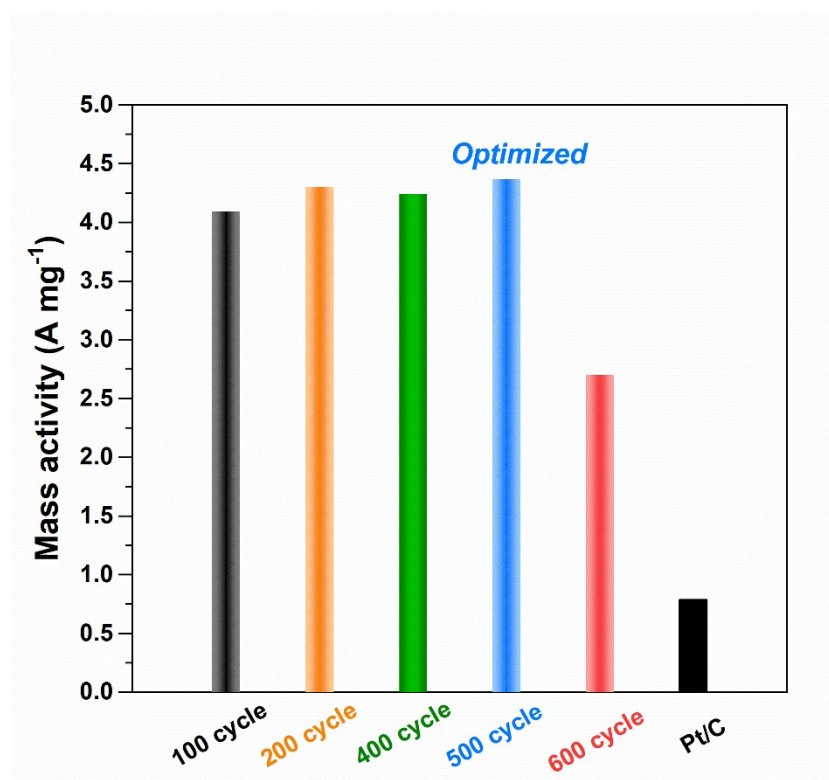


Fig. S2 Mass activity of peak current for 100 CV-Pt, 200 CV-Pt, 400 CV-Pt, 500 CV-Pt, 600 CV-Pt, and commercial Pt/C catalyst.

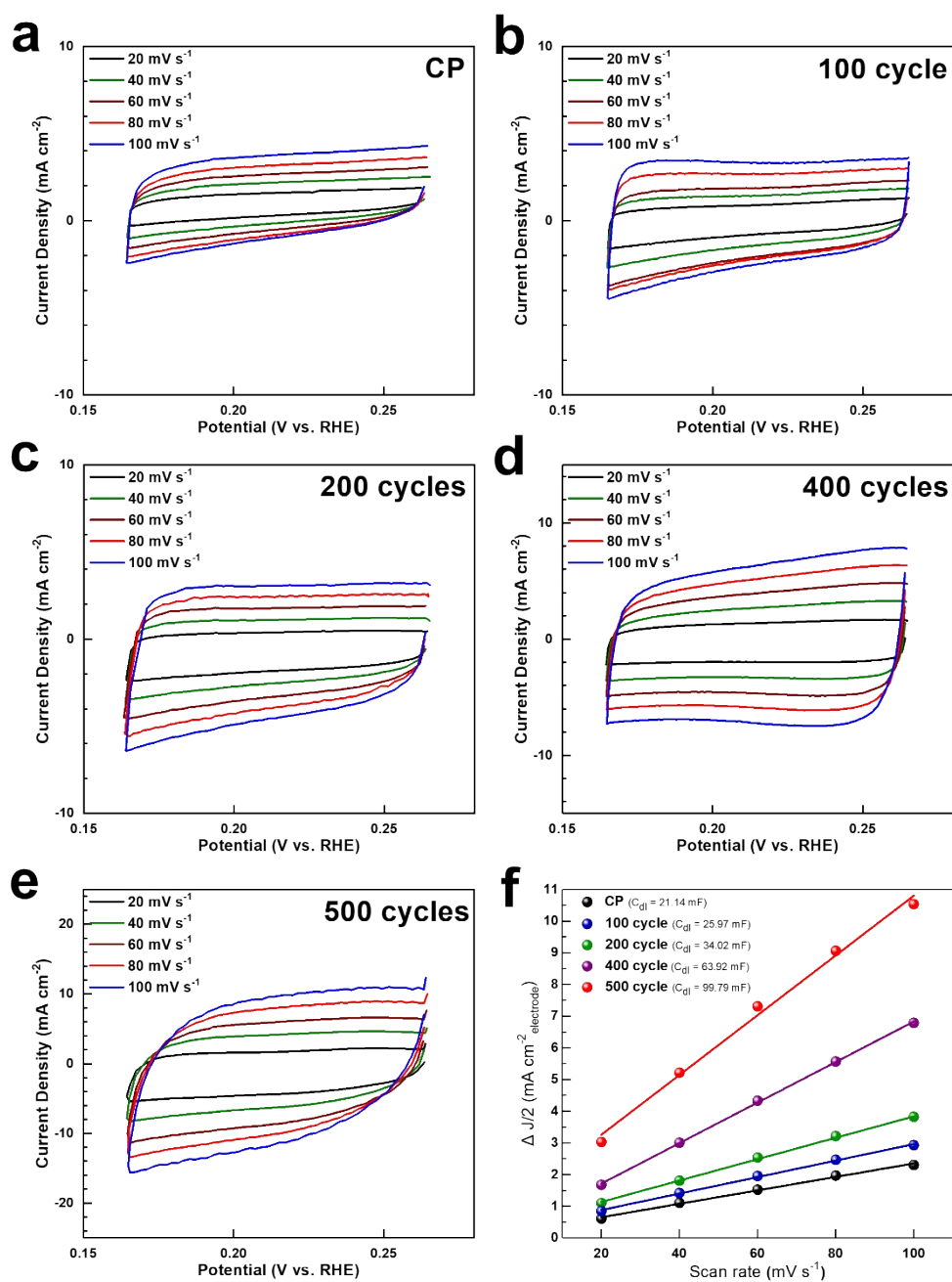


Fig. S3 Electrochemical CV scans recorded for each electrode; Scan rates are 20, 40, 60, 80, 100, and 120 mV s^{-1} . The selected potential range where no faradic current was observed from 0.165 to 0.265 V vs. RHE. (a) CP (b) CV-100 cycles (c) CV-200 cycles (d) CV-400 cycles (e) CV-500 cycles and (f) Capacitive current of each electrode at 0.215 V with different scan rates.

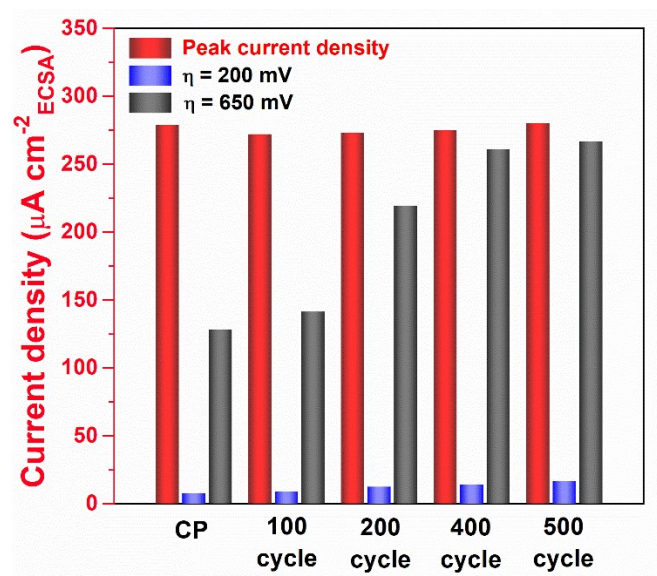


Fig. S4 Peak current density and current density at specific potential of each electrode. η means overpotential for AOR.

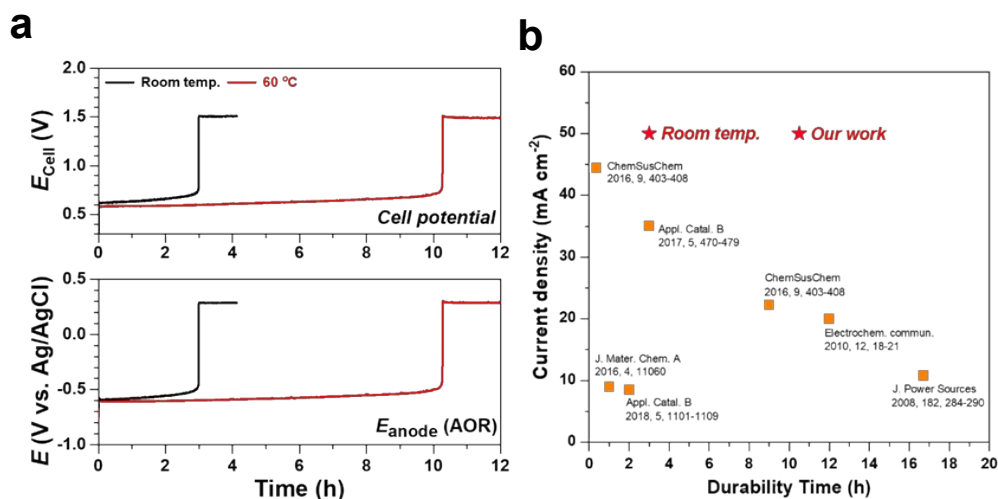


Fig. S5 (a) Stability test of the CV-Pt electrodes at 60°C and room temperature at constant current density of 50 mA cm^{-2} using 5 M KOH containing 1 M ammonia. (b) Maximum durability time and operating current density for the present work with that of previous studies for the electro-oxidation of ammonia.

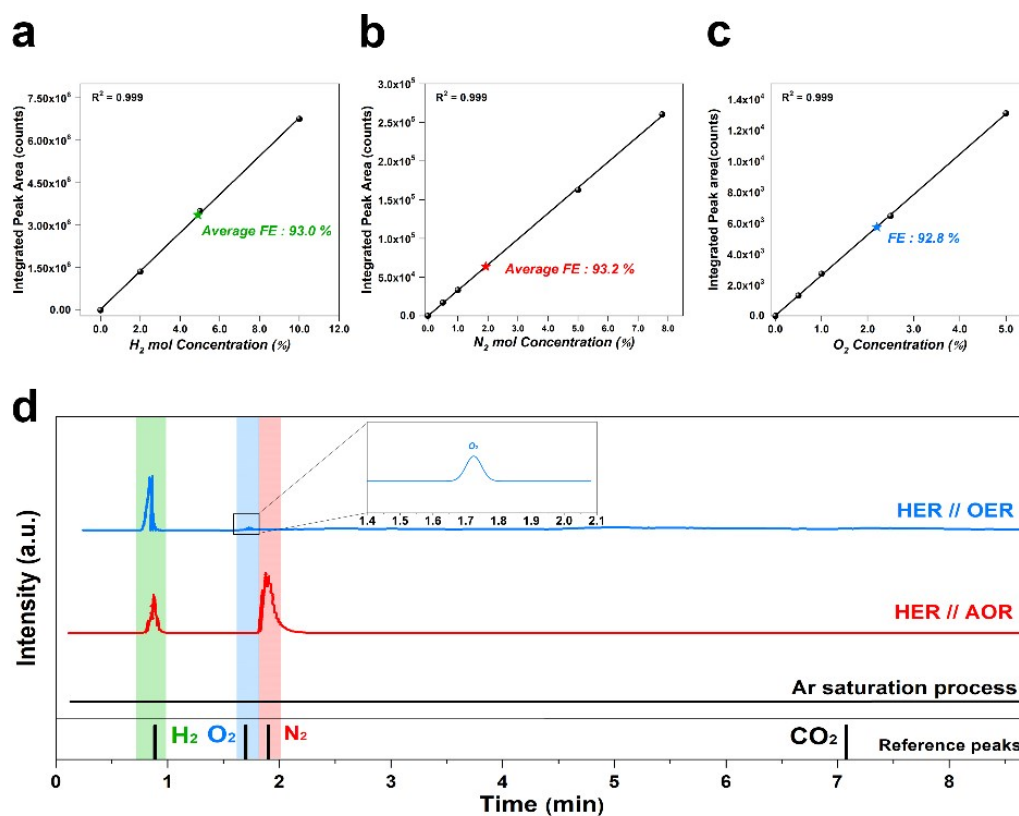


Fig. S6 Integrated peak areas of each gas and faradaic efficiency of (a) H_2 , (b) O_2 , and (c) N_2 . (d) GC profiles of Ar saturation process, HER/AOR, and HER//OER configuration. After Ar cleaning process, there is no residue gas in the ammonia electrolysis system.

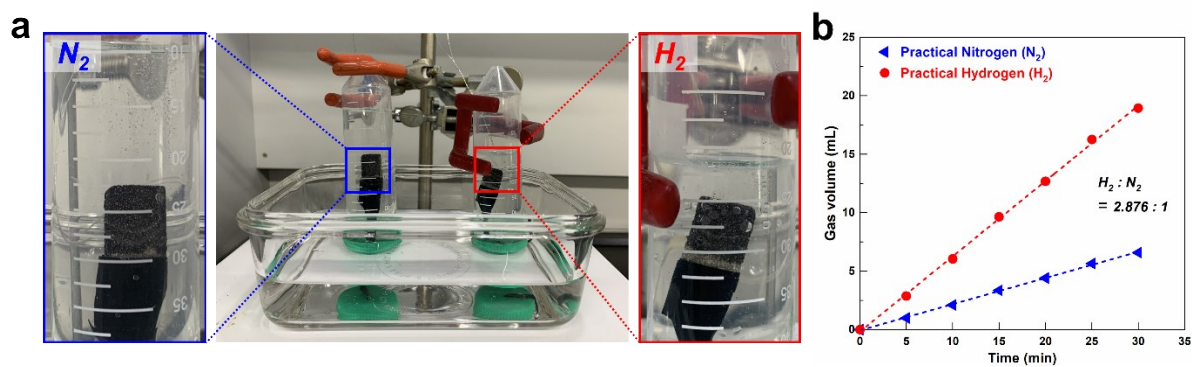


Fig. S7 a) A digital image of the ammonia electrolysis system for b) the production volume of H_2 and N_2 at 0.8V using 500 CV-Pt electrode.

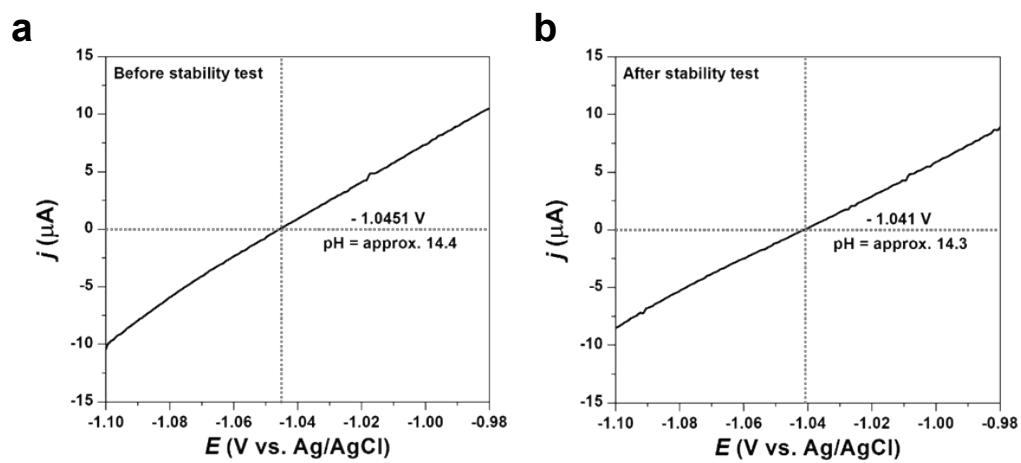


Fig. S7 RHE calibration test (a) before and (b) after stability test.

Supplementary Note 1. Activation Energy (Ea) Calculation

(In Figure 3c-d)

With the CVs results, we can obtain the apparent activation energy (E_a) calculated using the Arrhenius equation as follows.

$$k = A e^{(-E_a/RT)}$$

where k , A , R , and T represent the reaction rate constant, frequency factor, gas constant, and temperature, respectively. Between the logarithm of current density and the reciprocal of the temperature, the linear increase of current density was confirmed with increased temperatures. Taking the natural logarithm of Arrhenius equation yield.

$$\ln k = \ln A - \frac{E_a}{RT}$$

Rearranging yields.

$$\ln k = -\frac{E_a}{1000R} \left(\frac{1000}{T} \right) + \ln A$$

This has the same form as an equation for a straight line ($y = mx + b$). Then, we can calculate the E_a using the equation. For example, with the peak current density corresponding the different temperature, the slope of CV electrode was estimated as - 2.84. Then, from the value

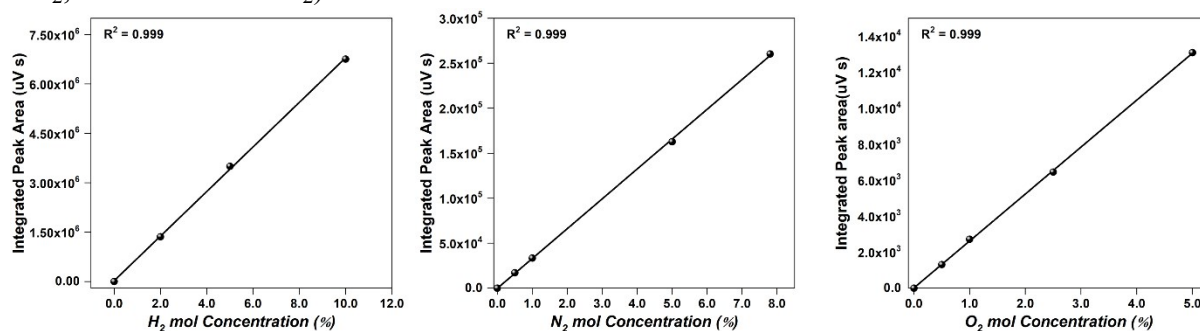
of slope, we can calculate the E_a ($-2.82 = -\frac{E_a}{1000R}$, $R = 8.31 \text{ J K}^{-1} \text{ mol}^{-1}$). The activation energy of CV electrode was 23.46 KJ mol⁻¹.

Supplementary Note 2. Suggested *In-operando* GC test protocol with example

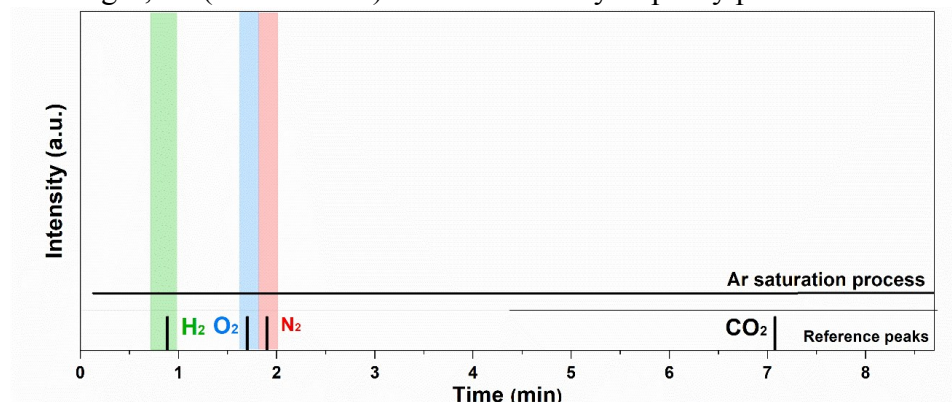
We implement a systematic bench-marking protocol that also quantifies the electrolysis of ammonia with an example of our real test as follows.

(1) Make calibration of H_2 , O_2 , and N_2 standard gas corresponding gas molar concentration versus integrated peak area.

(At least 3~4 point should be evaluated a mixture of standard gas and carrier gas. we have used 10 % of each gas with Ar based standard gas. the lower or higher concentration of each gas was mixed and used with high purity of gas such as 99.999 % Ar, 99.995 % O_2 , 99.999% N_2 , and 99.9999 % H_2)

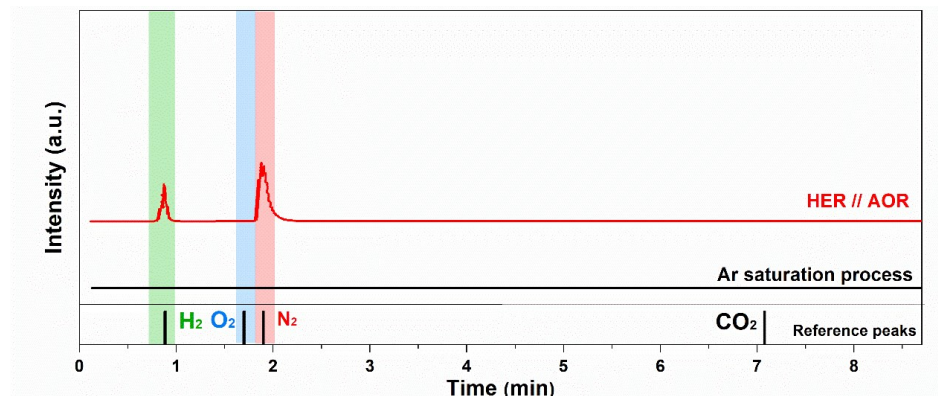


(2) Connect ammonia electrolysis system toward GC instrument with the constant flowing of carrier gas, Ar (50 mL min^{-1}) until without any impurity peaks on GC.



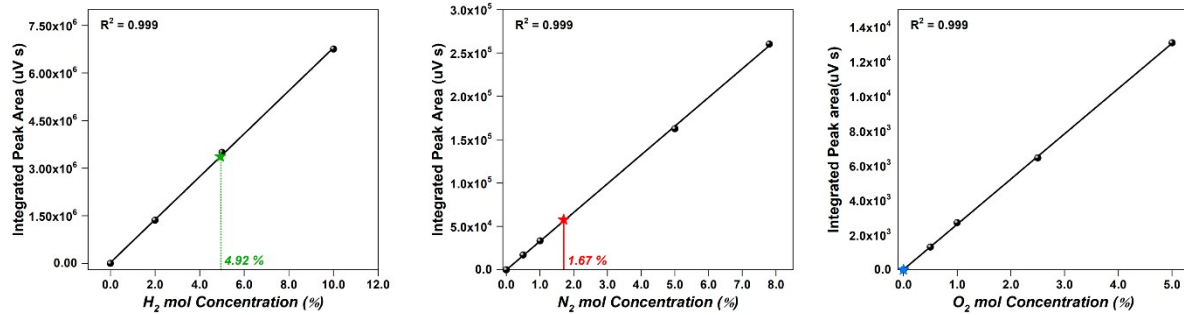
(3) With the operating of ammonia electrolysis, measure the produced gas with flowing the carrier gas, Ar (25 mL min^{-1}) into GC instrument.

(Example. H_2 // N_2 generated condition)



(4) With the GC results, we have estimated the integrated peak area of each gas. Then, we can calculate the mol concentration of each gas as follows.

(Example. H_2 // N_2 generated condition. Integrated Peak Area of H_2 = 3365490 uVs. Integrated Peak Area of N_2 = 63709.2 uVs.)



(5) Denote the mol concentration of H_2 , N_2 , and O_2 as A, B, and C, respectively. And denote the flow rate of produced gas ($mL\ min^{-1}$) of H_2 , N_2 , and O_2 as a, b, and c, respectively. Then, following the equations, the Faradaic efficiency could be calculated.

$$A = \frac{a}{25 + a + b + c} \times 100, \quad B = \frac{b}{25 + a + b + c} \times 100, \quad C = \frac{c}{25 + a + b + c} \times 100$$

(From the GC results of example, $A = 4.92$, $B = 1.67$, and $C = 0$. Then, value of a and b is calculated as 1.31, and 0.44 $mL\ min^{-1}$, respectively, indicating the value of FE is 94 %, and 96 %, respectively)

Table S1. FWHM and crystallite size extracted from the XRD data for electrodeposited Pt electrodes prepared with CC and CV methods

Sample	Theta (rad)	Cos θ	β , FWHM (rad)	λ , Cu K α (nm)	Particle size (nm)
500 cycle	0.353952	0.93801	0.015374509	0.15406	10.04174
	0.411007	0.916719	0.014589506	0.15406	10.82782
	0.600044	0.825311	0.017476807	0.15406	10.04010
400 cycle	0.351353	0.938908	0.016154846	0.15406	9.547553
	0.408695	0.91764	0.01562143	0.15406	10.10240
	0.597299	0.826857	0.017461752	0.15406	10.02996
200 cycle	0.351878	0.938727	0.014104084	0.15406	10.93789
	0.410338	0.916986	0.015462694	0.15406	10.21338
	0.597159	0.826936	0.018876891	0.15406	9.27716
100 cycle	0.351971	0.938695	0.014000307	0.15406	11.01934
	0.410114	0.917075	0.017184	0.15406	9.189430
	0.597581	0.826699	0.015794198	0.15406	11.09105
CC	0.352607	0.938476	0.008769777	0.15406	17.59568
	0.409807	0.917198	0.008017641	0.15406	19.69283
	0.597907	0.826515	0.011548777	0.15406	15.17157