

ESI (Electronic Supplementary Information)

Title:

Synergistic effect of Pd/CZO catalysts and an electric field on complete combustion of lean and humid methane at low temperatures

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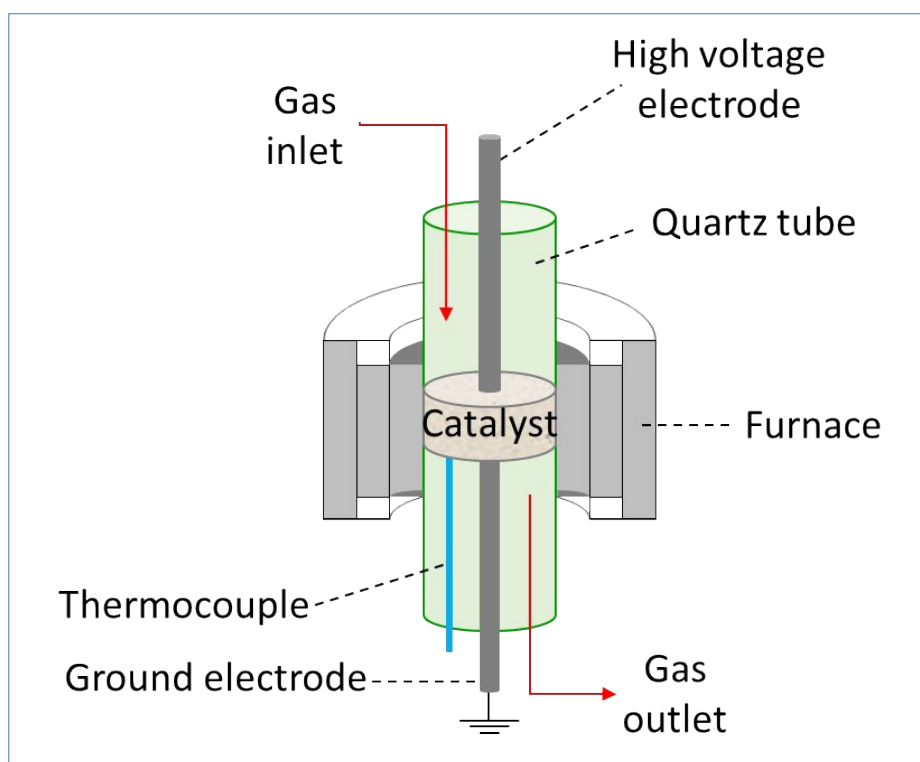


Figure S1 Schematic diagram of the reaction apparatus.

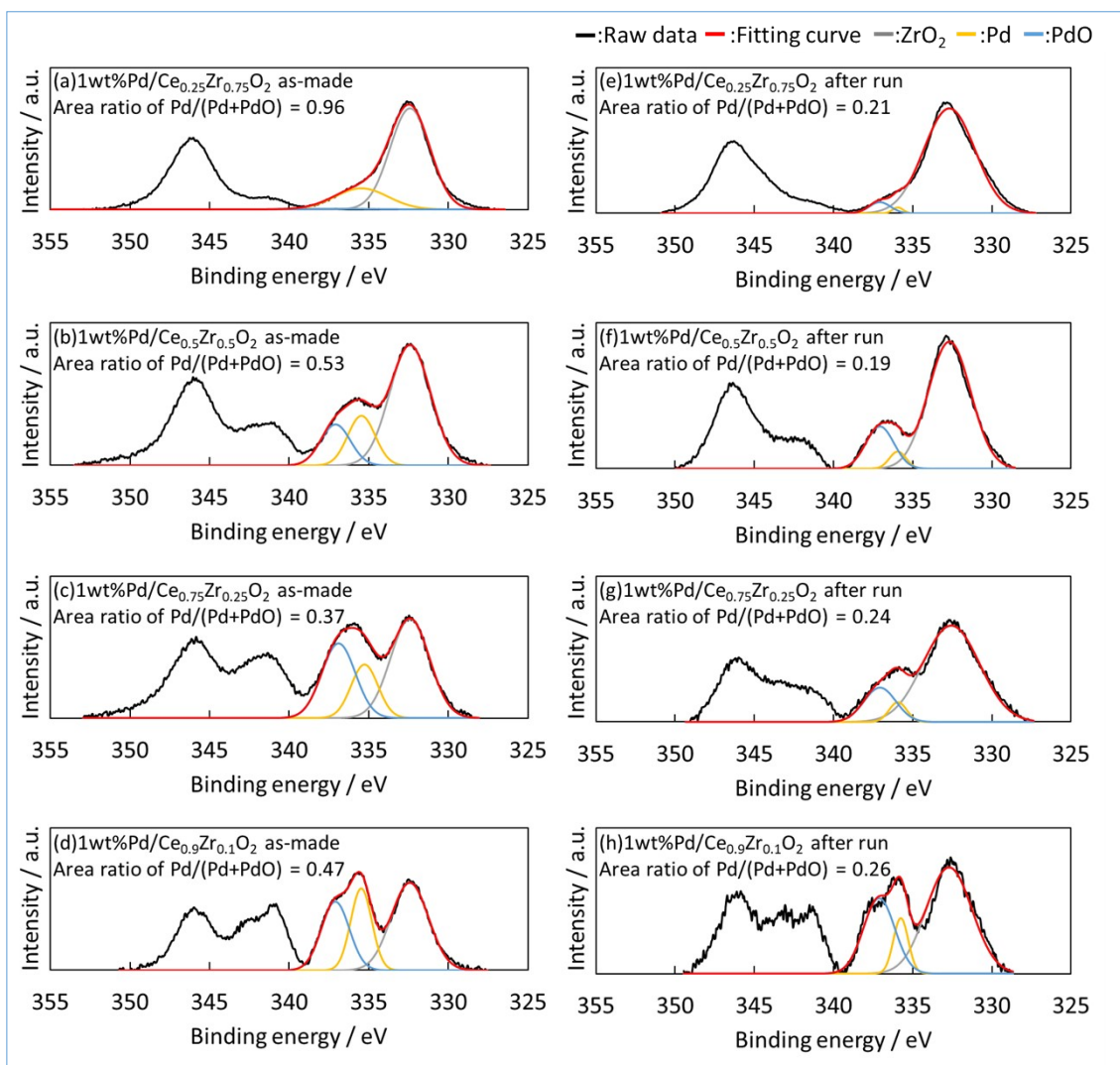


Figure S2 XPS profiles of 1wt%Pd/Ce_xZr_{1-x}O₂ in as-made and after reaction under the humid conditions with the electric field (1 mA) up to 723 K.

No significant correlation was observed between the chemical state of Pd in after reaction and the order of activity test results.

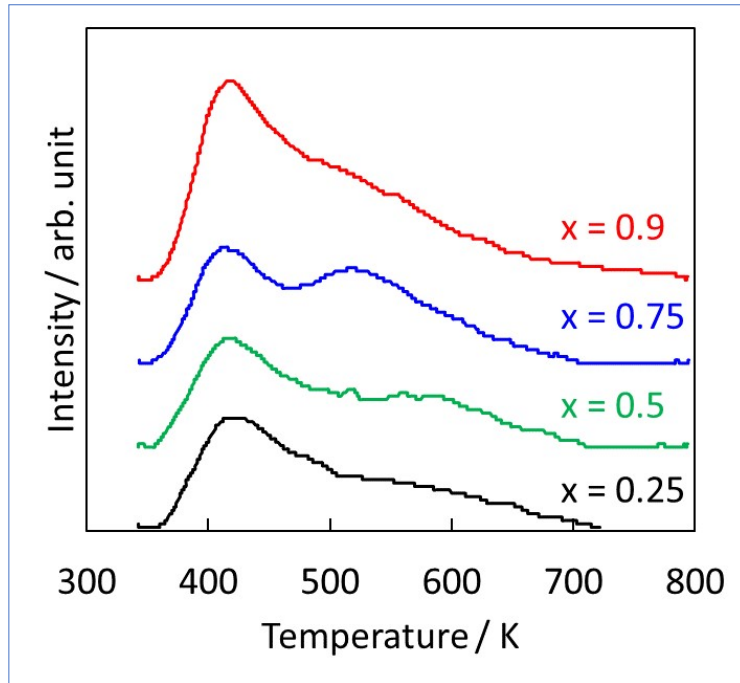


Figure S3 H₂O-TPD profiles over 1wt%Pd/Ce_xZr_{1-x}O₂.

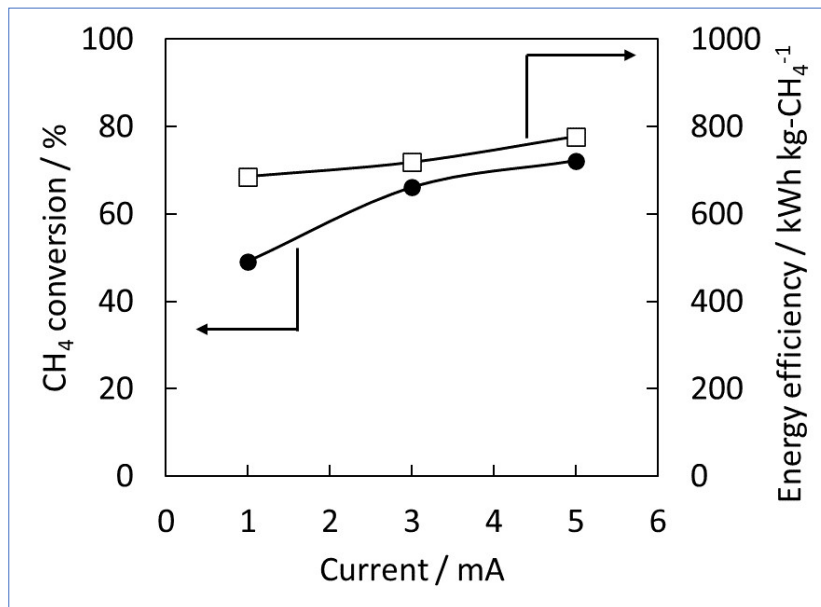


Figure S4 The results of current variation test and the energy efficiency over 1wt%Pd/Ce_{0.25}Zr_{0.75}O₂

in the EF (1, 3, 5 mA) at 473 K, CH₄: 2000ppm, O₂: 10%, H₂O: 10%, N₂ balance.

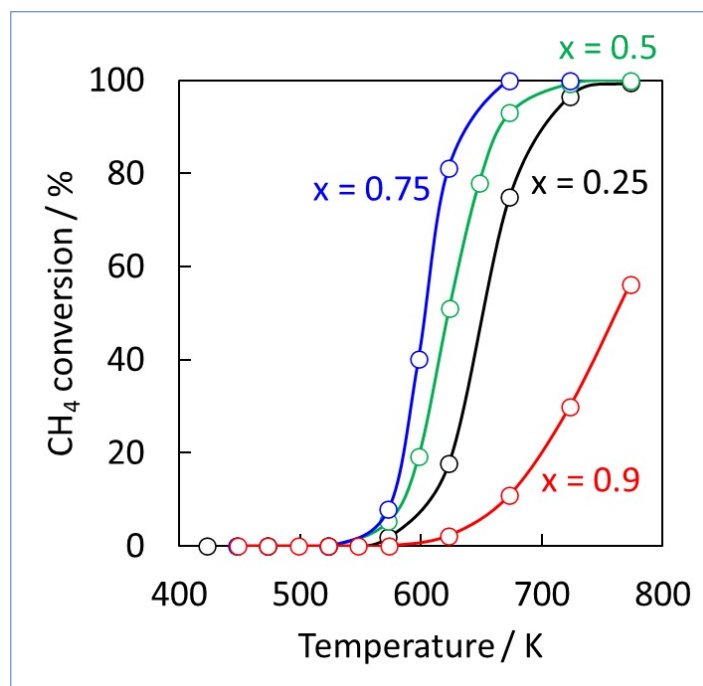


Figure S5 Methane conversion over 1wt%Pd/Ce_xZr_{1-x}O₂ without the EF, CH₄: 2000ppm, O₂: 10%,
H₂O: 10%, N₂ balance.

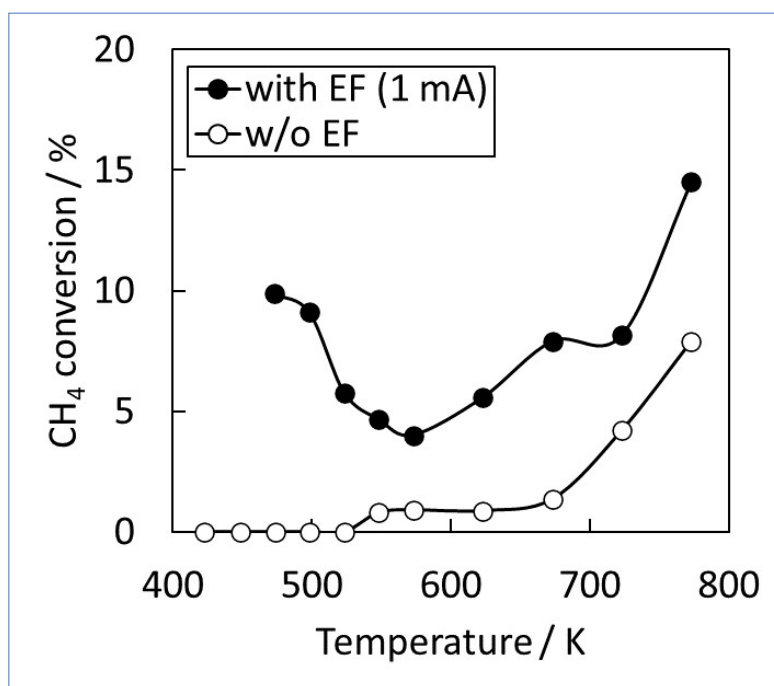


Figure S6 Methane conversion over Ce_{0.25}Zr_{0.75}O₂ with and without the EF.

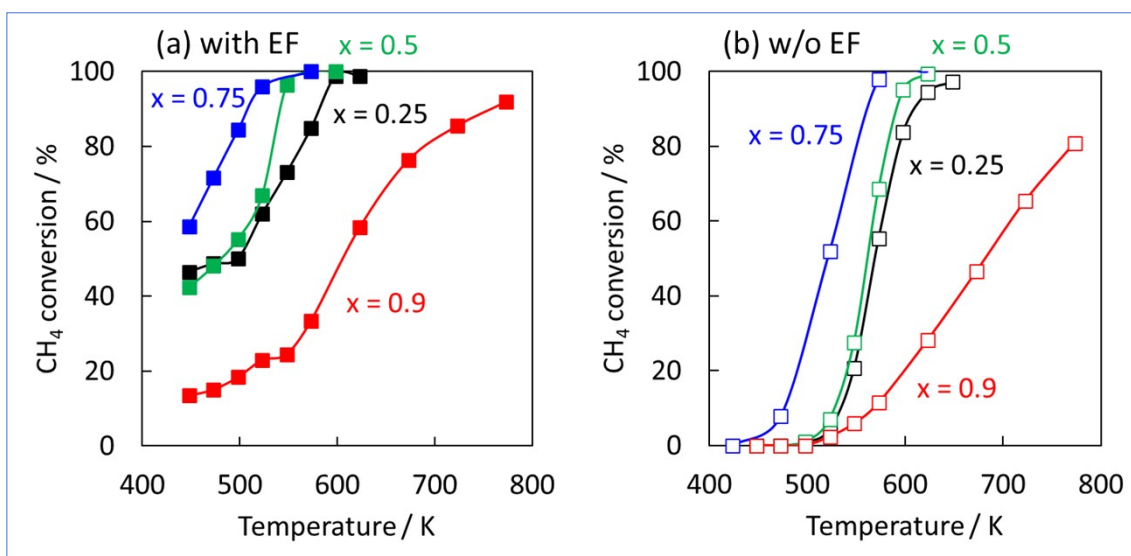


Figure S7 Methane conversion over 1wt%Pd/Ce_xZr_{1-x}O₂, (a) with the EF and (b) without the EF, CH₄: 2000ppm, O₂: 10%, N₂ balance.

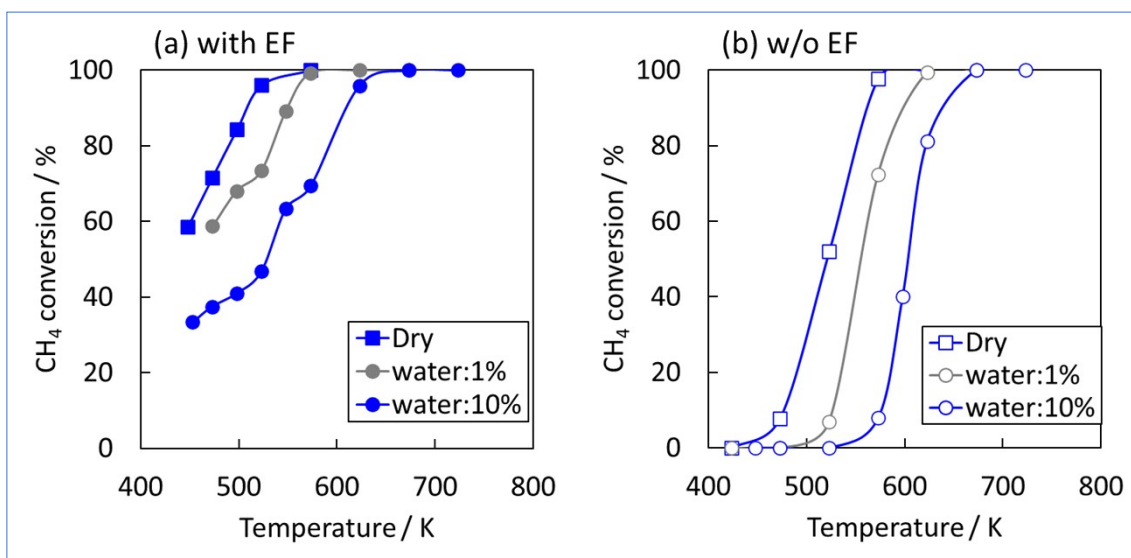


Figure S8 Methane conversion over 1wt%Pd/Ce_{0.75}Zr_{0.25}O₂ in various water vapour pressures, (a) with the EF and (b) without the EF.

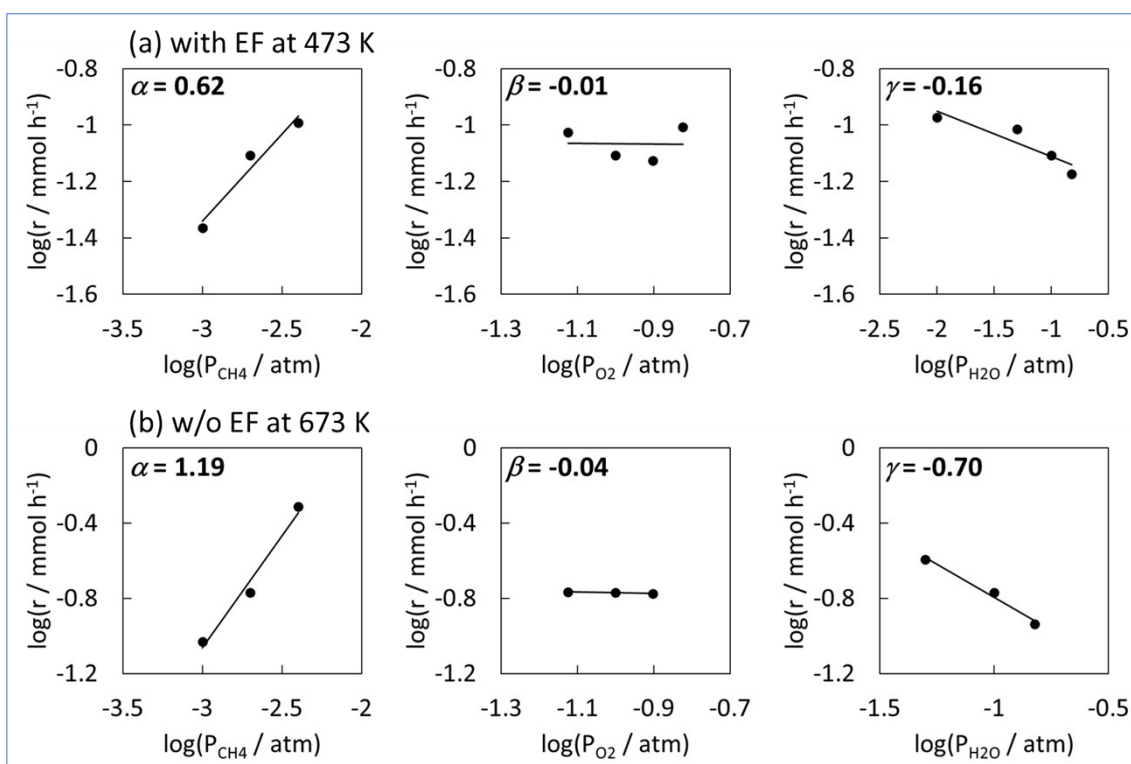


Figure S9 Methane, oxygen, and water pressure dependencies on the reaction rate of methane combustion in a kinetic range, (a) with the EF at 473 K and (b) without the EF at 673 K.

Catalyst: 60mg, total flow rate: 150 SCCM.

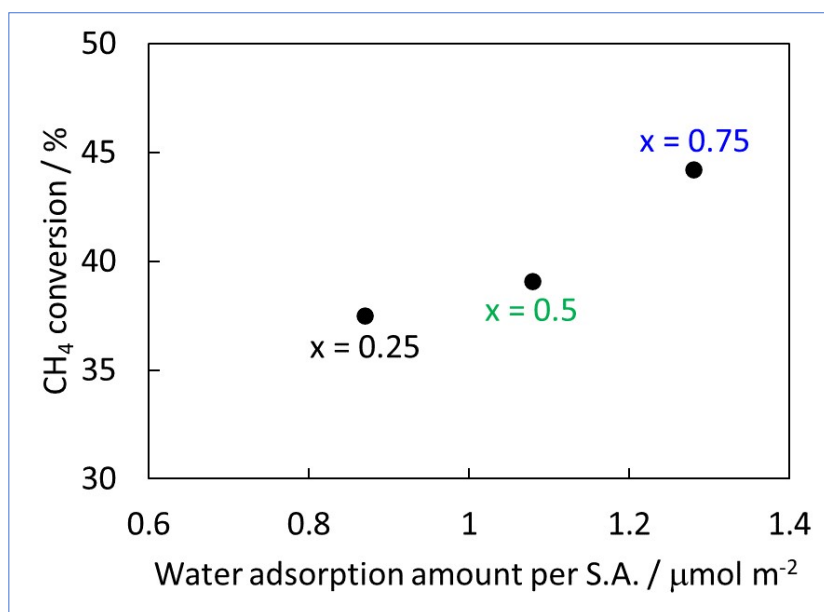


Figure S10 The relationship between water adsorption amount per surface area and the activity of the methane reaction with steam over 1wt%Pd/Ce_xZr_{1-x}O₂ in the EF, 473 K, fixed electric field power (1 W).

Table S1 Calculation results of the Ce-Zr-O (111) lattice spacing and the lattice constant.

Support	(111) Lattice spacing / nm	Lattice constant / nm
Ce _{0.25} Zr _{0.75} O ₂	0.300	0.450
Ce _{0.5} Zr _{0.5} O ₂	0.305	0.457
Ce _{0.75} Zr _{0.25} O ₂	0.309	0.463
Ce _{0.9} Zr _{0.1} O ₂	0.311	0.466

Table S2 Results of methane complete oxidation in a humid condition with and without the electric field over 1wt%Pd/Ce_{0.25}Zr_{0.75}O₂ catalyst, CH₄: 2000ppm, O₂: 10%, H₂O: 10%, N₂: balance.

EF	Temperature / K	Voltage / kV	Methane conversion / %	Selectivity / %	
				CO	CO ₂
With (1 mA)	473	1.93	58.9	0.7	99.3
	498	1.94	61.0	0.6	99.4
	523	1.97	63.7	0.2	99.8
	548	1.90	68.8	0.2	99.8
	573	1.69	77.3	0.0	100.0
	623	1.47	92.3	0.0	100.0
	673	1.45	99.0	0.0	100.0
	723	1.40	100.0	0.0	100.0
	373		0.0	-	-
	423		0.0	-	-
	473		0.0	-	-
	523		0.0	-	-
	Without	573	-	1.9	0.2
623			17.7	0.0	100.0
673			74.8	0.0	100.0
723			86.5	0.0	100.0
773			99.5	0.0	100.0

Table S3 Results of pressure changing tests with/without the electric field.

Temp. / K	P_{CH_4} / atm	P_{O_2} / atm	P_{H_2O} / atm	r / mmol h ⁻¹	V / kV	$r = kP_{CH_4}^\alpha P_{O_2}^\beta P_{H_2O}^\gamma$			
						α	β	γ	
473 with EF (1 mA)	0.001			0.0433	1.14				
	0.002	0.100		0.0782	0.98				
	0.004			0.1020	0.80				
			0.075	0.100	0.0943	1.15			
			0.100		0.0782	0.98			
			0.125		0.0748	1.03	0.62	-0.01	-0.16
	0.002	0.150		0.0983	1.12				
				0.010	0.1067	1.17			
			0.100	0.050	0.0969	1.10			
				0.100	0.0782	0.98			
			0.150	0.0670	1.02				
673 w/o EF	0.001			0.0932					
	0.002	0.100		0.1704					
	0.004			0.4866					
			0.100	0.1707					
			0.010	0.1704	-	1.19	-0.04	-0.70	
	0.002	0.125		0.1675					
			0.050	0.2542					
		0.100	0.100	0.1704					
			0.150	0.1156					