Supporting Information

Synthesis and Characterization of Individual High-Entropy Alloy Particles for Electrocatalytic Water Oxidation

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Table of Contents:

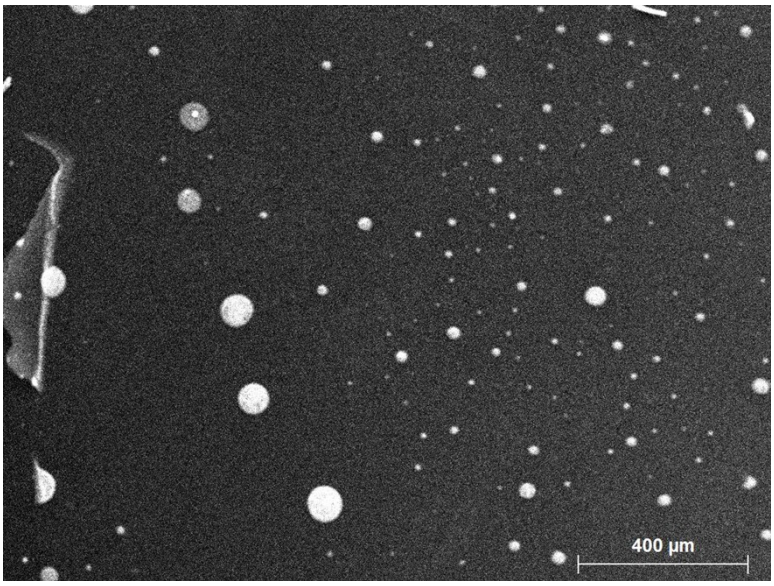


Fig. S1. SEM image of HEA particles on HOPG surface using backscattered electron detector (BSE) mode.

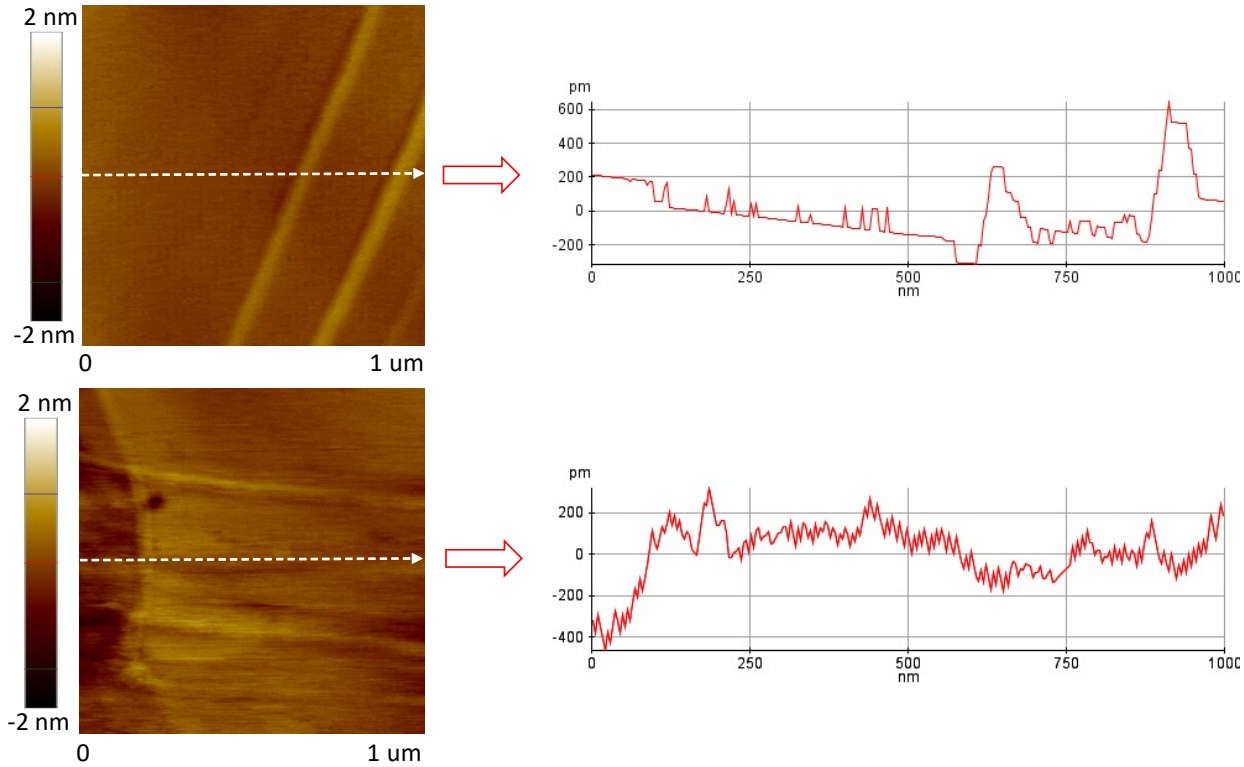


Fig. S2. AFM characterization of HOPG roughness before (top) and after (bottom) microwave irradiation. The dashed lines correspond to the shown cross sections.

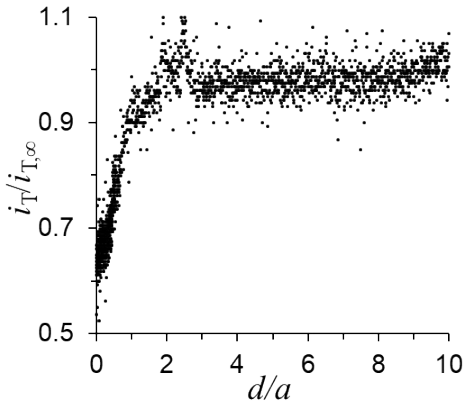


Fig. S3. Experimental dimensionless current *vs*. distance curve obtained with a microforged Pt nanotip approaching a 29-micron HEA particle on unbiased substrate in 0.1 M NaOH solution containing oxygen. E_T = − 0.7 V *vs.* Ag/AgCl; a = 355 nm; RG = 1.1.

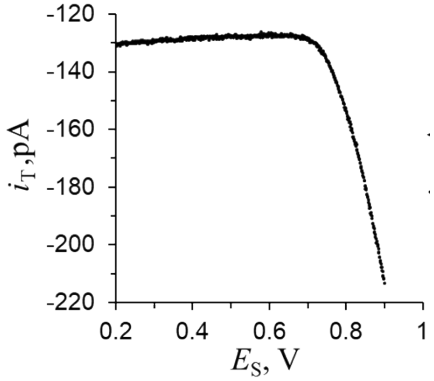


Fig. S4. i_T *vs* E_S curve recorded at a 200 nm-radius Pt nanoelectrode positioned approximately 85 nm above a 50-µm-radius HEA particle attached to HOPG. Substrate potential was scanned, and E_T was -0.7 V *vs.* Ag/AgCl. Solution contained 0.1 M NaOH (pH 13); v = 0.1 V/s.

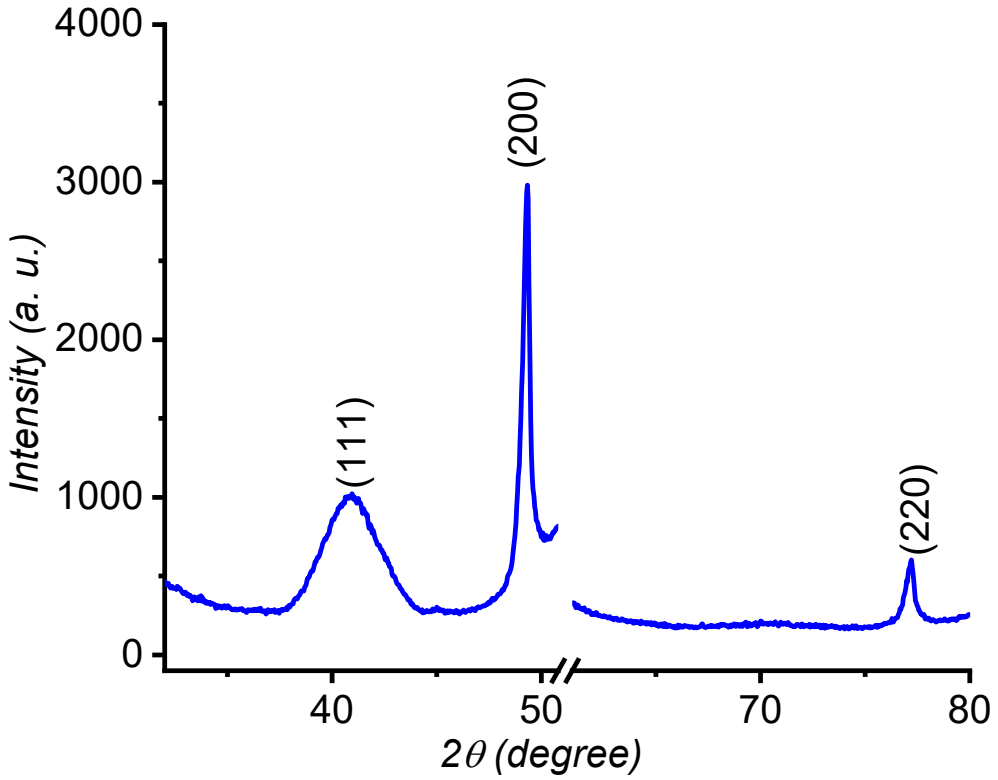


Fig. S5. XRD patterns of FeCoNiCuMnCr HEA particles on HOPG after electrochemical measurements.

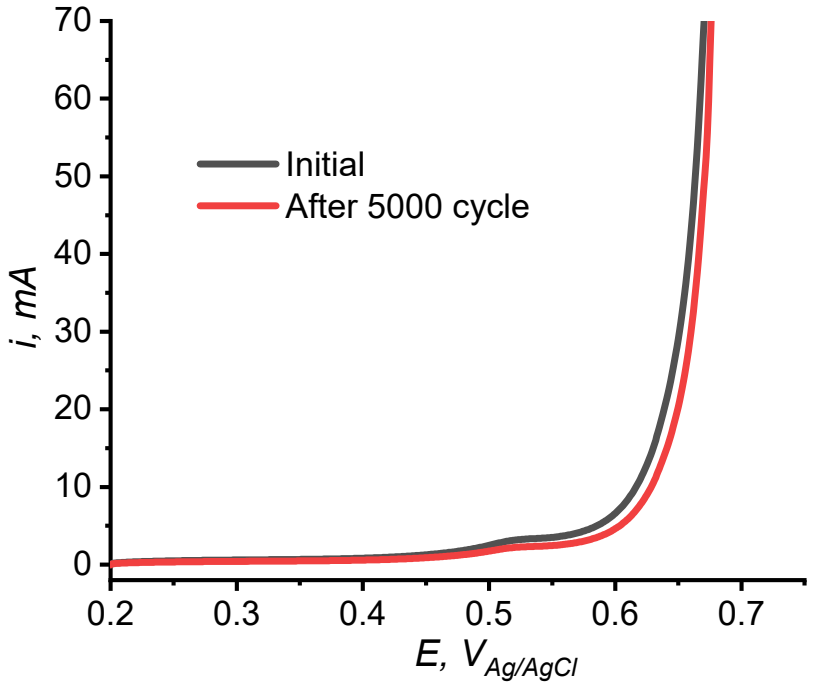


Fig. S6. A Comparison of OER polarization curves of HEA/HOPG, measured in 0.1 M NaOH aqueous solutions at a scan rate of 5 mV s^{-1}.

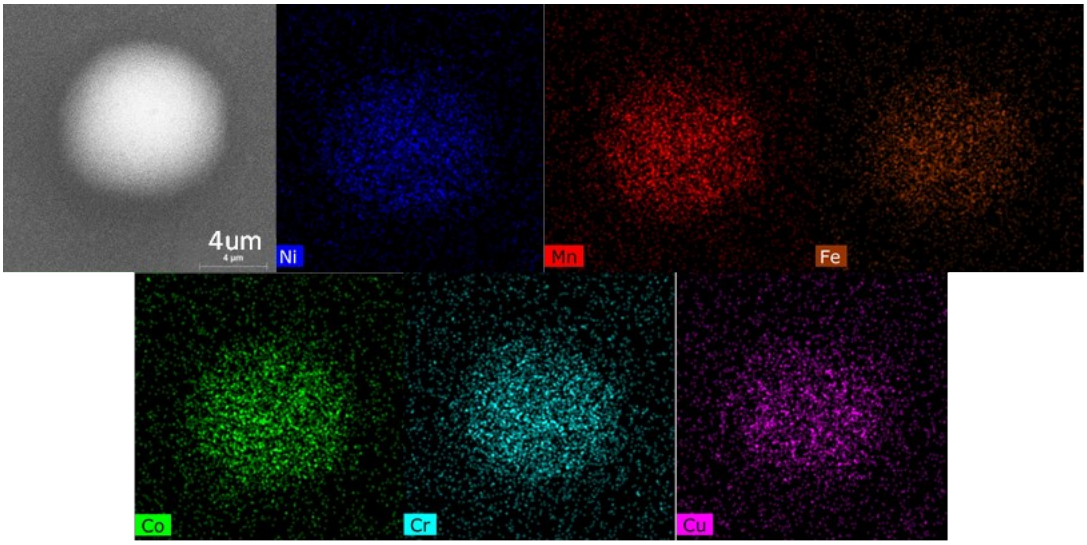


Fig. S7. SEM images of an individual ~8 μm HEA particle A using SE detector and its EDS elemental mapping.

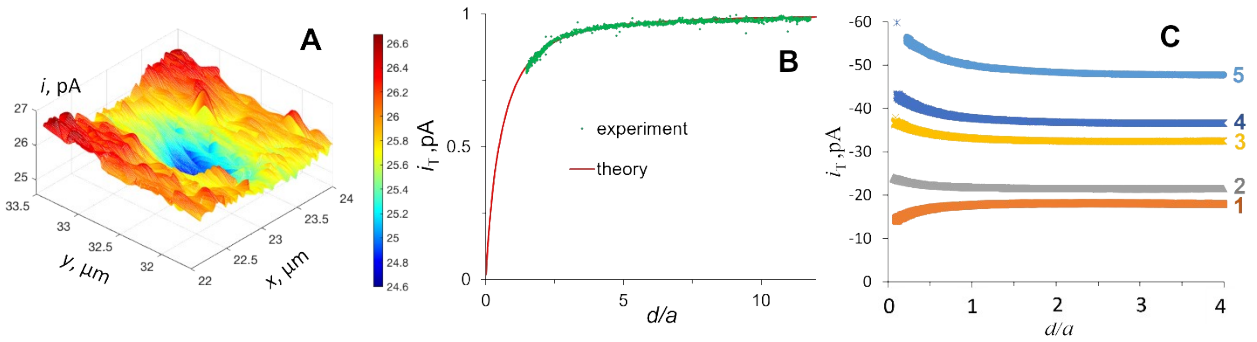


Fig. S8. SECM topography image of a ~0.5-µm-radius HEA particle on HOPG support (A) and current-distance curves obtained with the same 100 nm Pt tip approaching the same particle (B,C). (A,B) Solution contained 0.1 mM Fc in 0.1M KCl, E_T = 0.4V vs. Ag/AgCl. (C) SG/TC mode OER-based SECM current-distance curves. Solution contained 0.1 M NaOH. E_T = -0.7 V. E_s, V *vs.* Ag/AgCl = 0.2 (1), 0.3 (2), 0.4 (3), 0.5 (4) and 0.6 (5).

Table S1. The atomic percentages (at. %) of Cr, Mn, Ni, Fe, Co and Cu in a FeCoNiCuMnCr HEA measured by EDS analysis.

Elements	at. % in HEA(CrMnNiFeCoCu)/HOPG as prepared
Fe	16.6
Co	18.7
Ni	17.5
Cu	14.9
Mn	16.8
Cr	15.5

COMSOL MULTIPHYSICS®

SG/TC mode of SECM for OER Contents

1 Global Definitions

1.1 PARAMETERS

PARAMETERS 1

Name	Expression	Value	Description
a	355[nm]	3.55E−7 m	radius of electrode
RG	1.5	1.5	glass radius
D	2.4e-9[m*m/s]	2.4E−9 m²/s	diffusion coefficient
cbO2	0.25 [mmol/L]	0.25 mol/m³	concentration of O2
L	20	20	distance
id	4*F_const*D*cbO2*n*a	3.2882E−10 A	diffusion current at tip
n	4	4	number of electrons
id_e	id*1.162635233	3.823E−10 A	RG correction for 1.5
kt	0.27[cm/s]	0.0027 m/s	tip kinetics, ratio 0.3
f	0[nmol/(cm^2*s)]	0 mol/(m²·s)	flux of O2 from substrate
su	15[um]	1.5E−5 m	substrate

2 Component 1

2.1 DEFINITIONS

2.1.1 Coordinate Systems

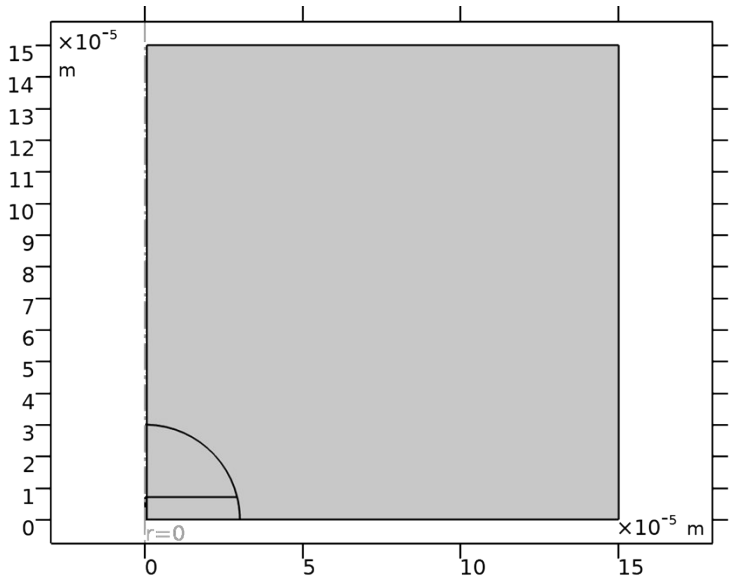
Boundary System 1

Coordinate system type	Boundary system
Tag	sys1

COORDINATE NAMES

First	Second	Third
t1	to	n

2.2 GEOMETRY 1



Geometry 1

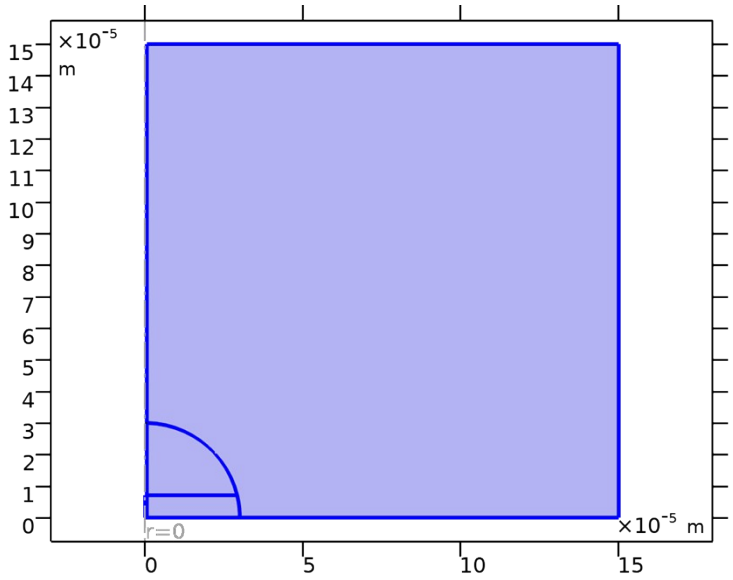
UNITS

Length unit	m
Angular unit	deg

2.3 TRANSPORT OF DILUTED SPECIES

USED PRODUCTS

COMSOL Multiphysics
Chemical Reaction Engineering Module

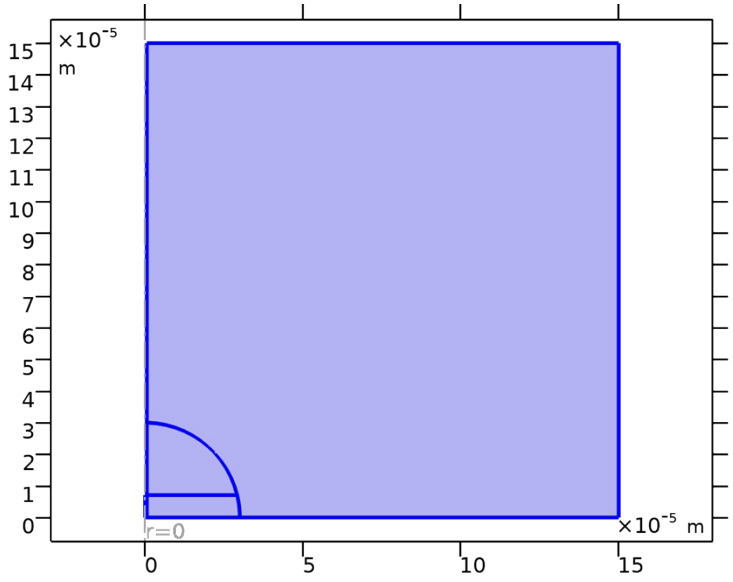


Transport of Diluted Species

EQUATIONS

$$\nabla \cdot \mathbf{J}_i = R_i$$
$$\mathbf{J}_i = -D_i \nabla c_i$$

2.3.1 Transport Properties 1



Transport Properties 1

EQUATIONS

$$\nabla \cdot \mathbf{J}_i = R_i$$
$$\mathbf{J}_i = -D_i \nabla c_i$$

Diffusion

SETTINGS

Description	Value	Unit
Source	Material	
Material	None	
Diffusion coefficient	User defined	
Diffusion coefficient	D	m²/s
Diffusion coefficient	User defined	
Diffusion coefficient	5E-9	m²/s

Coordinate System Selection

SETTINGS

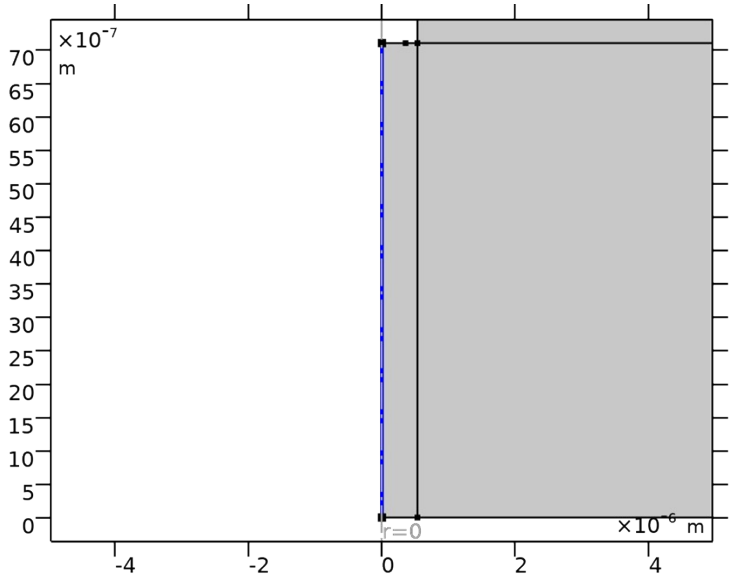
Description	Value
Coordinate system	Global coordinate system

Model Input

SETTINGS

Description	Value	Unit
Temperature	User defined	
Temperature	293.15	K

2.3.2 Axial Symmetry 1

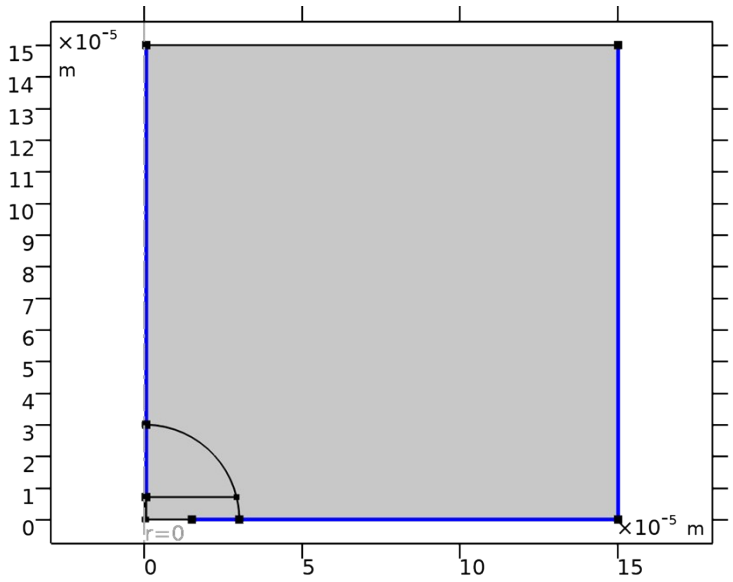


Axial Symmetry 1

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: All boundaries

2.3.3 No Flux 1



No Flux 1

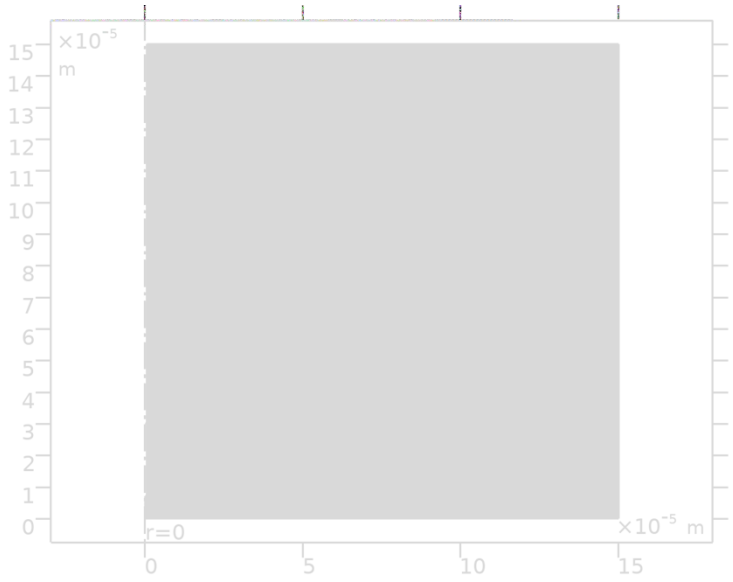
SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: All boundaries

EQUATIONS

$$-\mathbf{n} \cdot \mathbf{J}_i = 0$$

2.3.4 Initial Values 1



Initial Values 1

SELECTION

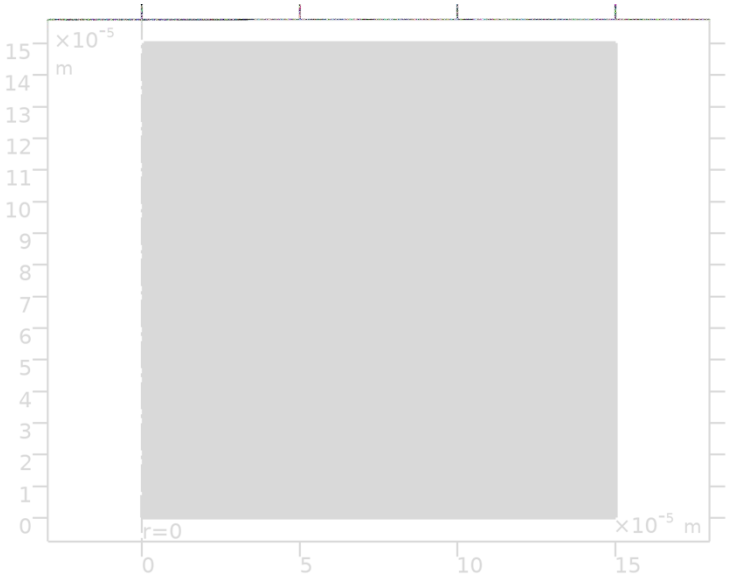
Geometric entity level	Domain
Selection	Geometry geom1: Dimension 2: All domains

Initial Values

SETTINGS

Description	Value	Unit
Concentration	{cbO2, 100}	mol/m³

2.3.5 Initial Values 2



Initial Values 2

SELECTION

Geometric entity level	Domain
Selection	Geometry geom1: Dimension 2: All domains

Initial Values

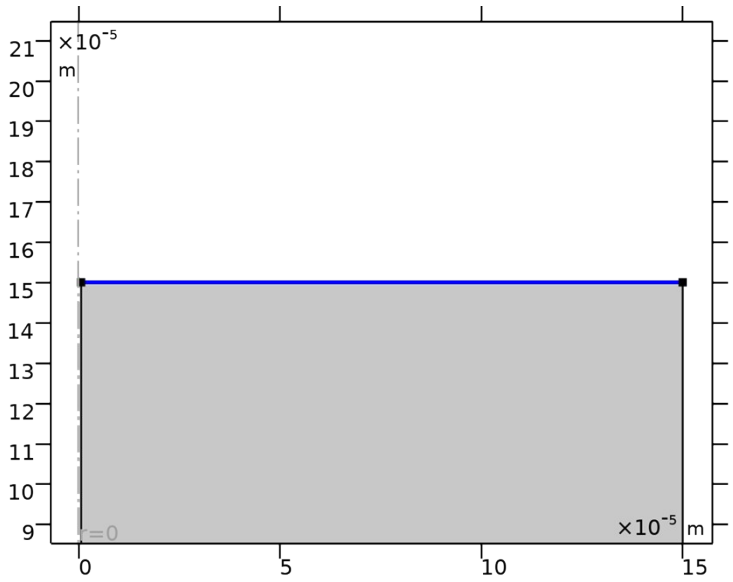
SETTINGS

Description	Value	Unit
Concentration	{cbO2, 100}	mol/m³

Variables

Name	Expression	Unit	Description	Selection	Details
tds.c0_cO2	cbO2	mol/m³	Concentration	Domains 1–4	+ operation
tds.c0_cOH	100	mol/m³	Concentration	Domains 1–4	+ operation

2.3.6 Concentration 1



Concentration 1

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: Boundary 10

EQUATIONS

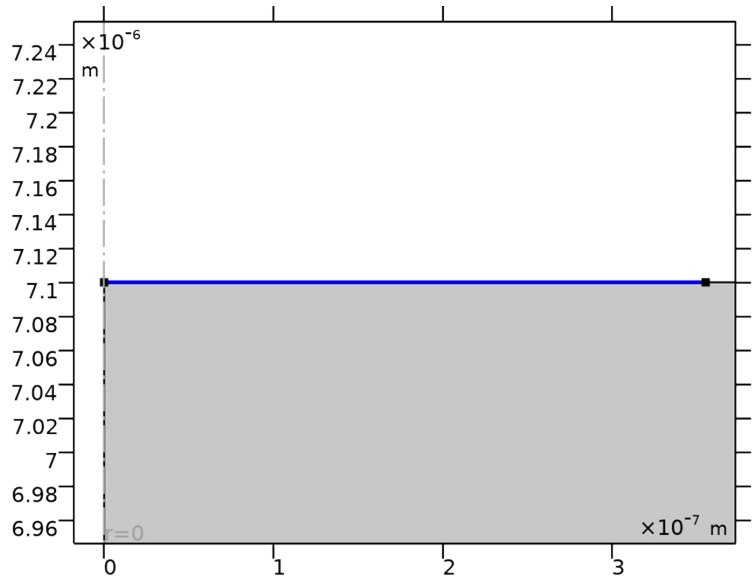
$$c_i = c_{0,i}$$

Concentration

SETTINGS

Description	Value	Unit
Species cO2	On	
Species cOH	On	
Concentration	{cbO2, 100}	mol/m³

2.3.7 Flux 2



Flux 2

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: Boundary 3

EQUATIONS

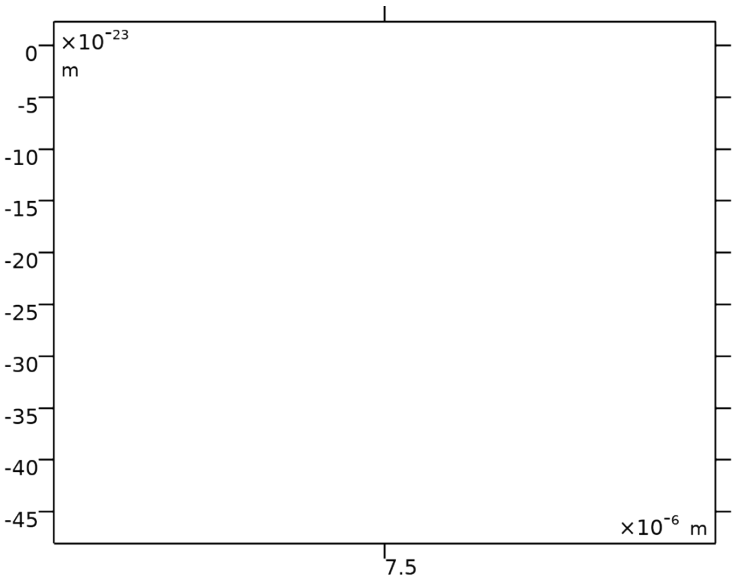
$$-\mathbf{n}\cdot\mathbf{J}_i = J_{0,i}$$

Inward Flux

SETTINGS

Description	Value	Unit
Flux type	General inward flux	
Species cO2	On	
Species cOH	On	
	{0.25*(-ktr*cO2 + kto*cOH), ktr*cO2 - kto*cOH}	mol/(m²·s)

2.3.8 Flux 3



Flux 3

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: Boundaries 2, 6

EQUATIONS

$$-\mathbf{n}\cdot\mathbf{j}_i = j_{0,i}$$

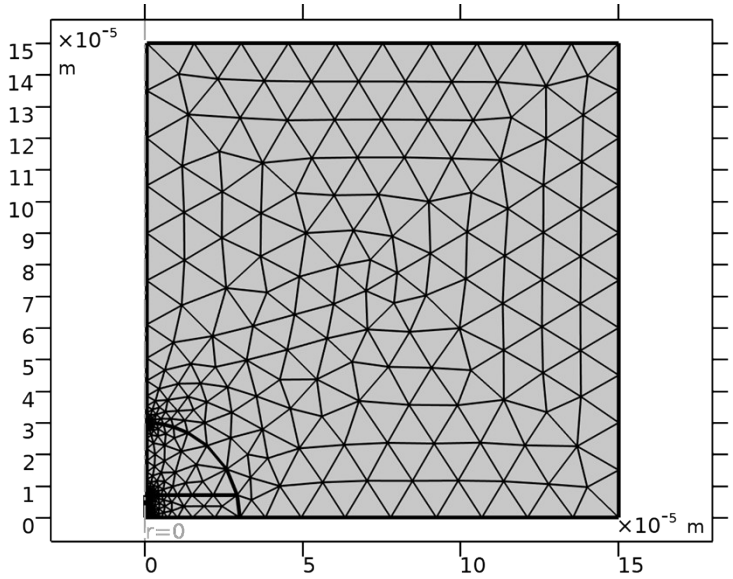
Inward Flux

SETTINGS

Description	Value	Unit
Flux type	General inward flux	
Species cO2	On	
Species cOH	On	
	{0.25*(-ksr*cO2 + kso*cOH) + f, ksr*cO2 - kso*cOH}	mol/(m²·s)

2.4 MESHES

2.4.1 Mesh 1



Mesh 1

MESH STATISTICS

Description	Value
Status	Complete mesh
Mesh vertices	312877
Triangles	23364
Quads	300000
Number of elements	323364
Minimum element quality	0.4195
Average element quality	0.9883
Element area ratio	2.9236E-8
Mesh area	2.242E-8 m²

Size (size)

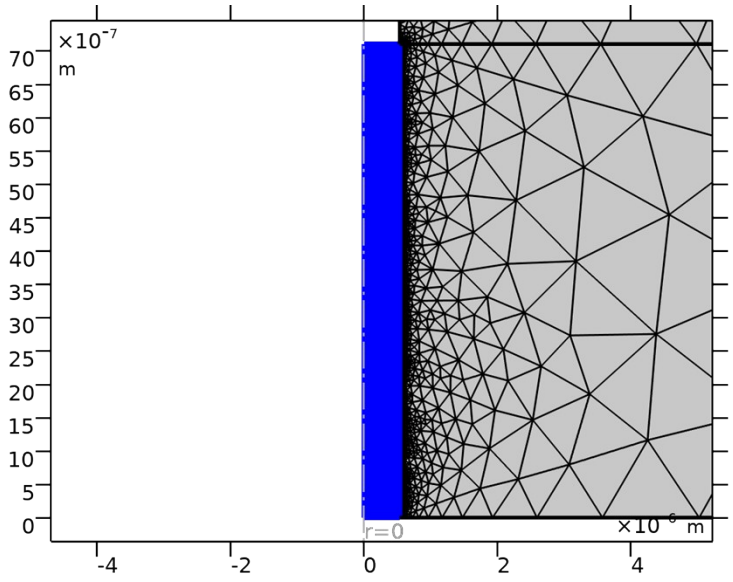
SETTINGS

Description	Value
Maximum element size	1.5E-5
Minimum element size	3E-7
Curvature factor	0.4
Maximum element growth rate	1.4
Predefined size	Coarse

Mapped 1 (map1)

SELECTION

Geometric entity level	Domain
Selection	Geometry geom1: Dimension 2: Domain 1



Mapped 1

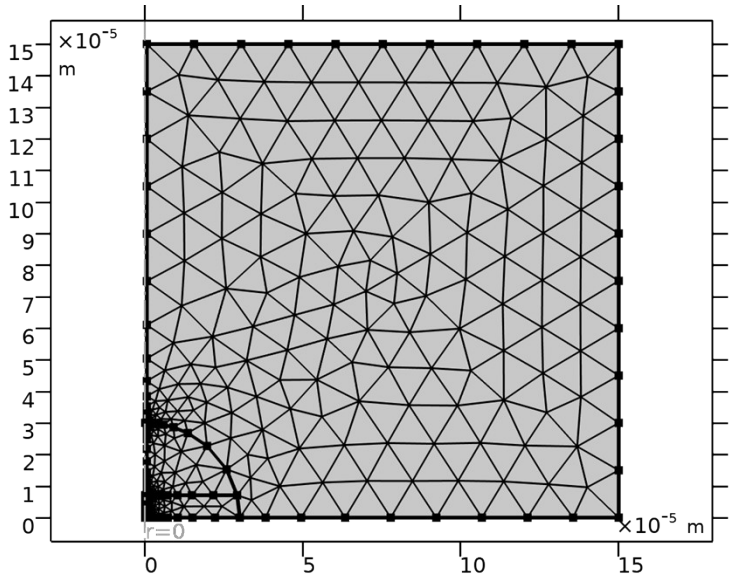
SETTINGS

Description	Value
Number of iterations	4
Maximum element depth to process	4
Last build time	0
Built with	COMSOL 6.1.0.282 (win64)|2024 - 11 - 25T14:06:52.178213900

Distribution 1 (dis1)

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: No boundaries



Distribution 1

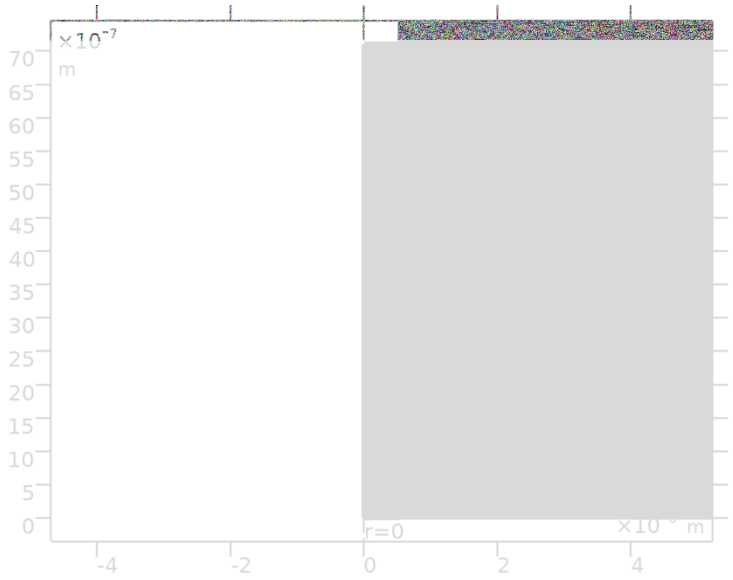
SETTINGS

Description	**Value**
Distribution type	Predefined
Number of elements	50
Element ratio	10
Reverse direction	On

Size 1 (size1)

SELECTION

Geometric entity level	Domain
Selection	Geometry geom1: Dimension 2: Domain 1



Size 1

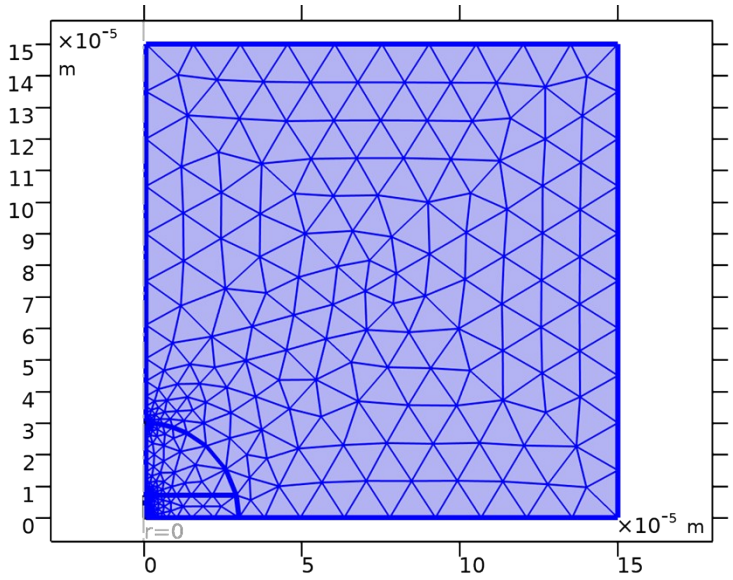
SETTINGS

Description	Value
Maximum element size	a*1e-2
Minimum element size	3E-6
Minimum element size	Off
Curvature factor	0.3
Curvature factor	Off
Resolution of narrow regions	Off
Maximum element growth rate	1.3
Maximum element growth rate	Off
Custom element size	Custom

Free Triangular 1 (ftri1)

SELECTION

Geometric entity level	Domain
Selection	Remaining



Free Triangular 1

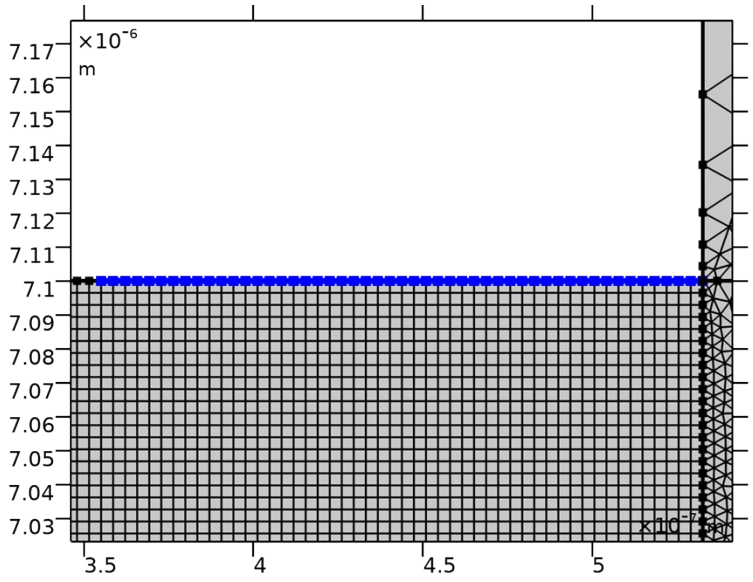
SETTINGS

Description	Value
Number of iterations	4
Maximum element depth to process	4
Last build time	1
Built with	COMSOL 6.1.0.282 (win64)|2024 - 11 - 25T14:06:53.877662100

Distribution 1 (dis1)

SELECTION

Geometric entity level	Boundary
Selection	Geometry geom1: Dimension 1: Boundary 4



Distribution 1

SETTINGS

Description	Value
Number of elements	10

3 Study 2

3.1 PARAMETRIC SWEEP

Parameter name	Parameter value list
L	0.01,0.2,0.4,0.5,0.8,0.98,1.3,1.7,2,2.5,3,4,6,8,10,20

STUDY SETTINGS

Description	Value
Sweep type	All combinations
Parameter name	L
Unit	

PARAMETERS

Parameter name	Parameter value list	Parameter unit
L (distance)	0.01,0.2,0.4,0.5,0.8,0.98,1.3,1.7,2,2.5,3,4,6,8,10,20	

ADVANCED SETTINGS

Description	Value
Default solver sequence generation	Using first parameter tuple

3.2 STATIONARY

STUDY SETTINGS

Description	Value
Include geometric nonlinearity	Off

PHYSICS AND VARIABLES SELECTION

Physics interface	Solve for	Equation form
Transport of Diluted Species (tds)	On	Automatic (Stationary)

MESH SELECTION

Component	Mesh
Component 1	Mesh 1

4 Results

4.1 DERIVED VALUES

4.1.1 Line Integration 1

OUTPUT

Evaluated in	Table 49

DATA

Description	Value
Dataset	Study 2/Parametric Solutions 5

EXPRESSIONS

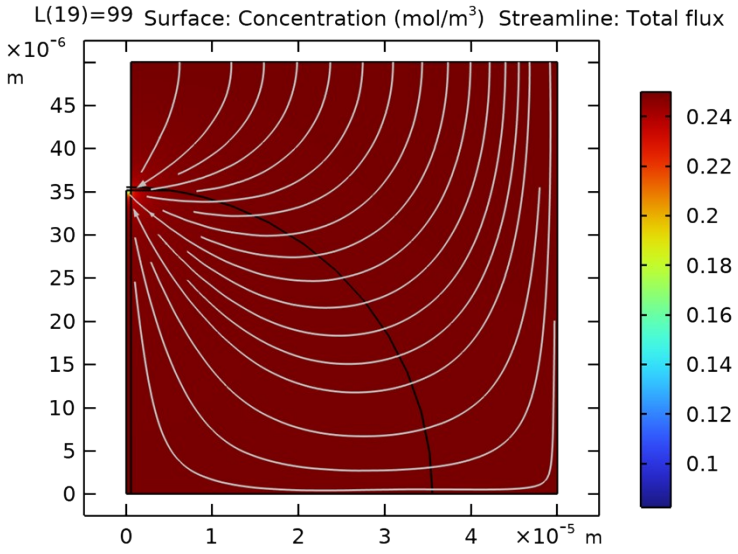
Expression	Unit	Description
tds.ntflux_cO2*F_const*4	pA	

INTEGRATION SETTINGS

Description	Value
Integration order	4
Compute surface integral	On

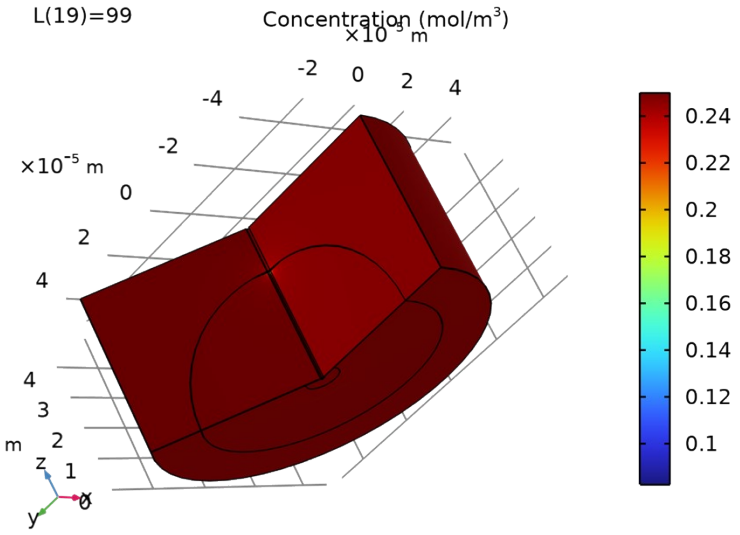
4.2 PLOT GROUPS

4.2.1 Concentration (tds)



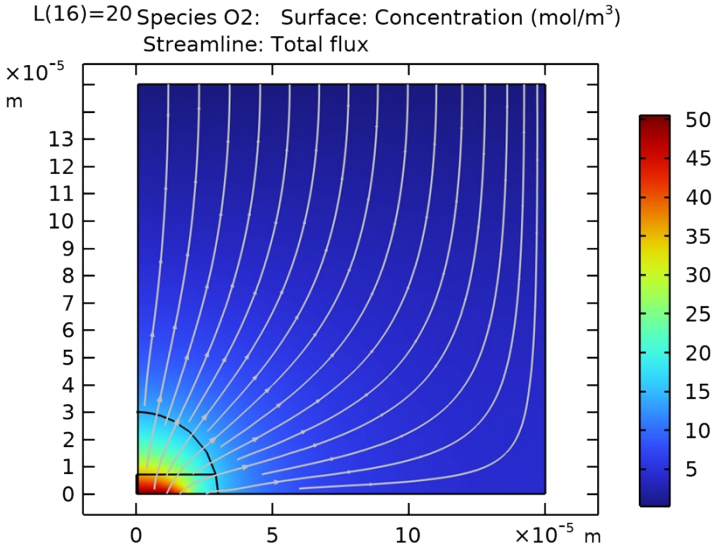
Surface: Concentration (mol/m³) Streamline: Total flux

4.2.2 Concentration, 3D (tds)



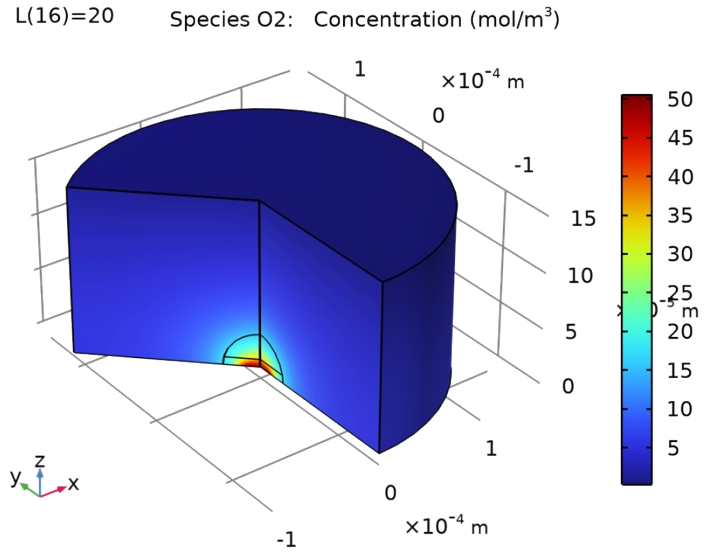
Concentration (mol/m³)

4.2.3 Concentration, O2 (tds)



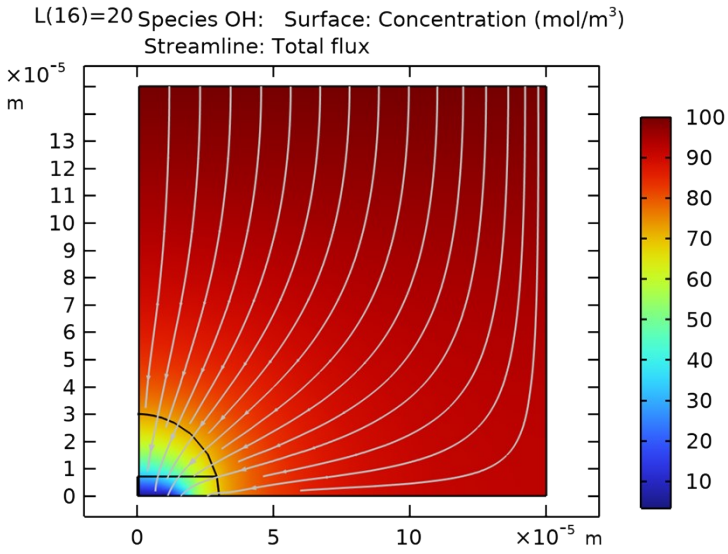
Species O2: Surface: Concentration (mol/m³) Streamline: Total flux

4.2.4 Concentration, O2, 3D (tds)



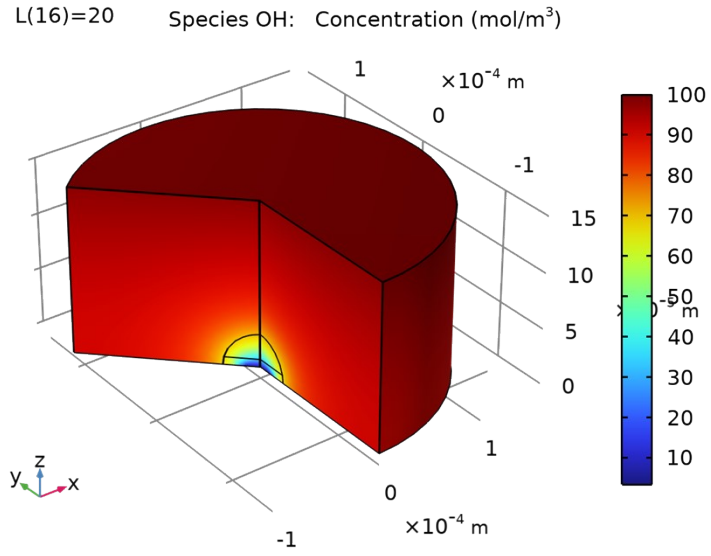
Species O2: Concentration (mol/m³)

4.2.5 Concentration, OH (tds)



Species OH: Surface: Concentration (mol/m³) Streamline: Total flux

4.2.6 Concentration, OH, 3D (tds)



Species OH: Concentration (mol/m³)