รายงานวิจัยบับสมบูรณ์

โครงการ การพัฒนาแบบจำลองทางคณติศาสตร์ของระบบ Reforming/Fischer-Tropsch สำหรับกระบวนการเปลี่ยนก๊าซเป็นของเหลวจากก๊าซธรรมาชาติ

Project Development of an Integrative Modeling of Reforming/Fischer-Tropsch Process for Gas-to-Liquid production from natural gas and biogas)

ชื่อหัวหน้าโครงการวิจัยผู้รับทุน : ดร.พรรณนิภา ดอกไม้งาม

ชื่อนักวิจัยที่ปรึกษา : ศ.ดร. นวดล เหล่าศิริพจน์

สัญญาเลขที่ MRG5980139

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สนับสนุนโดยสำนักงานกองทุนสนับสนุนการวิจัยและสำนักงานคณะกรรมการการอุดมศึกษา

บทคัดย่อ

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ชื่อโครงการ: การพัฒนาแบบจำลองทางคณิตศาสตร์ของระบบ Reforming/Fischer-Tropsch สำหรับกระบวนการเปลี่ยนก๊าซเป็นของเหลวจากก๊าซธรรมาชาติ

ชื่อนักวิจัยและสถาบัน: ผู้ช่วยศาสตราจารย์ ดร. พรรณนิภา ดอกไม้งาม สำนักวิชาวิทยาศาสตร์สุขภาพ มหาวิทยาลัยแม่ฟ้าหลวง

อีเมลล์: pannipha.dok@mfu.ac.th หรือ dpannipha@gmail.com

ระยะเวลาโครงการ 2 ปี

บทคัดย่อ

งานวิจัยนี้ได้พัฒนาแบบจำลองทางคณิตศาสตร์สามมิติขึ้นเพื่อศึกษาพฤติกรรม ของระบบ internal reforming solid oxide fuel cell แบบท่อ โดยแบบจำลองทางคณิตศาสตร์จะประกอบด้วยสมการทางด้านพฤติกรรมของก๊าซที่ไหลภายในท่อ รวมกับการถ่ายเทความร้อนทั้งแบบการนำความร้อน การพาความร้อน และการแผ่รังสี ร่วมกับปฏิกิริยารีฟอร์มมิ่งและปฏิกิริยาไฟฟ้าเคมี ผลการคำนวณแสดงให้เห็นถึงการความไม่สอดคล้องกันในการถ่ายเทความร้อนระหว่างปฏิกิริยาดูดความร้อนและปฏิกิริยาคายความร้อนภายในระบบจึงเกิดการลดลงของอุณหภูมิภายในท่อ ซึ่งส่งผลกระทบโดยตรงต่อปริมาณไฟฟ้าที่เกิดขึ้นในเซลล์เชื้อเพลิง โดยเมื่ออุณหภูมิลดลงค่าความต้านทาน Ohmic จะเพิ่มมากขึ้นปริมาณไฟฟ้าที่ได้จะลดลงและการเปลียนแปลงอุณภูมิอย่างกระทันหันยังส่งผลต่อคุณสมบัติทางกลของวัสดุที่ใช้ทำเซลล์เชื้อเพลิง จากการปรับตัวแปรในการควบคุมการทำงานของเซลล์เชื้อเพลิงพบว่าหากลด อัตราส่วนของเชื้อเพลิงต่อไอน้ำลงจะทำให้อุณหภูมิที่ลดลงมากขึ้น

คำสำคัญ IR-SOFC, แบบจำลอง, สามมิติ

Abstract

Project code: MRG5980139

Project name: Development of an Integrative Modeling of Reforming/Fischer-Tropsch Process for Gas-to-Liquid production from natural gas and biogas

Investigater: Asst.Prof.Dr.Pannipha Dokmaingam, School of Health Science, Mae Fah Luang University

Project period: 2 years

Abstract

The 3D mathematical model was developed to predict the behavior of a tubular internal reforming solid oxide fuel cell. In this model is also integrated with momentum transfer, heat transfer, reforming reactions and electrochemical reactions. The simulation results shown that that cooling effect at the entrance of reformer could be reduced by adding small amount of oxygen into IIR-SOFC configuration; nevertheless, the system's efficiency decreased. It was also observed that the electrical efficiency could be increased when the system was operated at lower S/C ratio or higher operating temperature but the temperature distribution in the IIR-SOFC system became more undesirable.

Key words: IR-SOFC, Simulation, 3D

บทสรุปผู้บริหาร

งานวิจัยนี้เป็นการสร้างแบบจำลองคณิตศาสตร์สามมิติเพื่อทำนายพฤติกรรมของวัสดุที่ใช้ในการสร้างเซลล์เชื้อเพลิงควบรวมกับรีฟอร์มเมอร์เพื่อมีการแลกเปลี่ยนความร้อนระหว่างปฏิกิริยาคายความร้อนและปฏิกิริยาดูดความร้อนแต่เนื่องจากความไม่สอดคล้องกันระหว่างอัตราในการดูดและคายความร้อนส่งผลให้เกิดปัญหาเรื่องการแตกของวัสดุของเซลล์เชื้อเพลิงเพื่อป้องกันปัญหาดังกล่าวจึงมีการทำนายพฤติกรรมของเซลล์เชื้อเพลิงจากการปรับ ปัจจัยในการควบคุมการดำเนินงาน (Operating Parameter)เพื่อลดความแตกต่างของอัตราการคายความร้อนและดูดความร้อนของปฏิกิริยาข้างต้น หากสามารถลดปัญหาเรื่องการแตกของเซลล์เชื้อเพลิงได้จะสามารถยืดอายุการใช้งานของเซลล์เชื้อเพลิงและลดการใช้พลังงานในการดำเนินการของเซลล์เชื้อเพลิง ส่งผลต่อความเหมาะสมด้านราคาที่จะนำเซลล์เชื้อเพลิงมาใช้เป็นพลังงานทดแทนมากขึ้น

ในงานวิจัยฉบับนี้ ได้สร้างแบบจำลองสามมิติของเซลล์เชื้อเพลิงแบบ Solid Oxide โดยออกแบบให้มีการควบรวมปฏิกิริยาสตรีมรีฟอร์มมิ่งกับปฏิกิริยาไฟฟ้าเคมีไว้ด้วยกัน การแลกเปลี่ยนความร้อนของทั้งสองปฏิกิริยาทำนายโดยใช้สมดุลโมเมนตัม สมดุลมวล สมดุลพลังงาน ร่วมกับปฏิกิริยาข้างต้น ผลการคาดการณ์พฤติกรรมที่เกิดในท่อของเซลล์เชื้อเพลิงชี้ให้เห็นอุณหภูมิที่ลดลงอย่างรวดเร็วในช่วง 2 ส่วน 3 ของท่อ เพื่ออุณหภูมิลดลงค่าความต้านทาน Ohmic ของเซลล์เชื้อเพลิงจะเพิ่มขึ้น กระแสไฟฟ้าที่ผลิตได้จึงลดลงไปด้วย จะเห็นว่า ความไม่สอดคล้องของอัตราการคายความร้อนของปฏิกิริยาสตรีมรีฟอร์มมิ่งและการดูดความร้อนของปฏิกิริยาไฟฟ้าเคมี นอกจากจะส่งผลเสียต่อวัสดุของเซลล์เชื้อเพลิงแล้วยังส่งผลเสียปริมาณไฟฟ้าที่เซลล์เชื้อเพลิงจะผลิตได้ด้วย

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1. Introduction

1.1 Rational

Nowadays, the world economy is driven by using conventional fuels such as coal, oil, and natural gas. Among them, natural gas is known to be one of the most widely used energy resources, particularly Thailand (Department of Alternative Energy Development and Efficiency, 2020). This is the major source of greenhouse gases (GHGs) that cause climate change. In order to reduce this impact, there are a lot of renewable energy technologies had been proposed. One of the promising technologies is a fuel cell. The fuel cell is a technology that converts chemical energy directly to electricity without GHG emission. For stationary application, Solid Oxide Fuel Cell (SOFC) is interesting because it is operated without noise, flexible for hydrocarbon fuel, and high efficiency Since it is operated at high temperature, it could be applied for co-generation (Ramadhani, Hussain, Mokhlis, & Hajimolana, 2017; Santos et al., 2018). During operation, the hydrocarbon fuel is reformed to syngas via several reactions such as partial oxidation reaction, dry reforming reaction, and autothermal reforming reaction. After that, the syngas is feed to the anode side of SOFC where exothermic electrochemical takes place. The energy optimization between these two reactions is operated in an internal reforming SOFC (IR-SOFC) configuration where the waste heat from the reforming reaction can be applied for the cogeneration system. The coupling of reformer with SOFC would improve the overall process efficiency and leads the process to be more economic feasible. However, the main limitation of IR-SOFC is energy matching between both sections.

This work is proposed to study the thermal behavior of those integrated systems to determine the optimum operating condition. The kinetic equations of catalytic steam reforming and electrochemical reactions from the literature would be integrated with theoretical thermodynamic equations. Several operating conditions (i.e. temperature, pressure, reactant ration, and flow rate) will be studied.

1.2 Objectives

- Developed mathematical models with three-dimensional IIR-SOFC operations to predict the thermal behavior and system efficiency.
- Compared the system efficiency and performance with different reformer and FT reactor configurations (i.e. packed bed, coated wall).

  6.3 Proposed the alternative design of integrative reforming/FT system for GTL production.

1.3 Scope of research

- Review the kinetic rate equations for the methane reforming and FT reaction from the literatures.
- Set up experiment to validate the simulation results of selected kinetic rate expressions
- Develop mathematical models based on theory of momentum heat and mass transfer in the micro reactor.
- Fill the kinetic rate equations with parameters in the models to predict the behavior of a reformer integrated FT system.
- Simulate the characteristic of a reformer integrated FT system in different stack configurations.

2. Literature review

The main objective of this research study is to develop a model for investigating internal reforming SOFCs. Typically, mathematical models are used for predicting behaviors of gases temperature in the interested system. The gases are kinetically controlled by reforming and electrochemical reactions, whereas the thermal characteristic of the system can be predicted by thermodynamic considerations. The previous related literatures are reviewed on the following topics: steam reforming kinetic analysis and modeling of IIR-SOFCs.

2.1 Kinetic Analysis and Kinetic Model of Methane Steam Reforming

As the reforming aspect, there are several kinds of reactions that could converted the hydrocarbon gases to synthesis gases, as listed in Table 2.1. All advantages and disadvantages of each reaction are also compared. It could be seen that autothermal reforming is benefited optimized heat utilization in the reactor. This is also advantage for economic consideration. However, it is limited for commercial application since the mismatch of endothermic and exothermic reaction. So this work will predict the chemical electrical and heat behavior of IIR-SOFC.

During the past decade, Ni-based catalysts, such as Ni/Al2O3 and Ni/YSZ, were widely utilized in methane steam reforming (MSR) processes. In order to explore the impact parameters of these processes, much of the literatures presented kinetic models with different applications, as summarized in Table 2.2.

Table 2-1Comparison of syngas production technologies fueled by natural gas (Gangadharan, Kanchi, & Lou, 2012; Wilhelm, Simbeck, Karp, & Dickenson, 2001)

Technology	Advantages	Disadvantages
Steam Reforming	• Most extensive industrial experience Oxygen not required • Lowest process temperature requirement • Best H_2/CO ratio for hydrogen production applications	• H_2/CO ratio often higher than required when CO also is to be produced • Highest air emissions • Limited commercial experience
Partial Oxidation	• Feedstock desulfurization not required. • Absence of catalyst permits carbon formation and, therefore, operation without steam, significantly lowering syngas CO_2 content • Low methane slip • Low natural H_2/CO ratio is an advantage for applications requiring ratio<2.0	• Low natural H_2/CO ratio is a disadvantage for applications requiring ratio>2.0. • Very high process operating temperatures • Usually requires oxygen • High-temperature heat recovery and soot formation/handling adds process complexity • Syngas methane content is inherently low and not easily modified to meet downstream processing requirements
Dry Reforming	• Greenhouse gas CO_2 can be consumed instead of releasing in to atmosphere • Almost 100% of CO_2 conversion	• Formation of coke on catalyst
Autothermal Reforming	• Natural H_2/CO ratio often is favorable • Lower process temperature requirement than POX • Low methane slip • Syngas methane content can be tailored by adjusting reformer outlet temperature	• Limited commercial experience • Usually requires oxygen

Syngas generation processes

I) Steam reforming is the endothermic reaction for produce syngas from a hydrocarbon gas. The general steam reforming reactions are presented in Eqs. 1-3. It should be noted that water gas shift reaction, Eq. 2, is also occurred together with a steam reforming reaction. Actually, the steam reforming reaction is operated over nickel-based catalysts. During the past decade, Ni-based catalysts, such as Ni/Al_2O_3 and Ni/YSZ, were widely utilized in the methane steam reforming (MSR) processes. In order to explore the impact parameters of these processes, most of the literature presented kinetic models with different applications, as summarized in Table 2. One of the well-known MSR kinetic models was presented by Jianguo Xu and Gilbert F. Froment (1989). They

studied the rate of methane steam reforming reactions which accompanied by water-gas shift reactions over Ni/$MgAl_2O_4$ catalysts. The intrinsic rate equations were derived base on the three significant reactions in a MSR system, as shown below.

$$CH_4 + H_2O \leftrightarrow 3H_2 + CO \quad (1)$$

$$CO + H_2O \leftrightarrow H_2 + CO_2 \quad (2)$$

$$CH_4 + 2H_2O \leftrightarrow 4H_2 + CO_2 \quad (3)$$

These three reactions were also converted into the works of (Hou & Hughes, 2001a) and (Hoang, Chan, & Ding, 2005). Hou and Hughes (2001b) studied the MSR kinetics over commercial Ni/αAl_2O_3 catalysts at the temperature range of 748-823 K. They developed the kinetic models by using the Langmuir-Hinshelwood-Hougen-Watson assumption and the Freudlich's adsorption theory, which reasonably fix their experimental data.

Table 2-2 Summary of methane steam reforming rate expressions over Ni base-catalyst

Rate Equation	Reference
$R_{CH_4} = \dfrac{k \Big/ p_{H_2}^{2.5}\left(p_{CH_4}p_{H_2O} - \dfrac{p_{H_2}^{3}p_{CO}}{k_{eq}}\right)}{(1 + k_{CO}p_{CO} + k_{H_2}p_{H_2} + k_{CH_4}p_{CH_4} + \dfrac{k_{H_2O}p_{H_2O}}{p_{H_2}})^2}$	Jianguo Xu and Gilbert F Froment (1989)
$R_{CH_4} = \dfrac{k\ (T)p_{CH_4}}{\left(1 + K_H(T)p_{H_2}^{0.5} + K_s(T)\,p_{H_2O}/p_{H_2}\right)^n}$	Dicks, Pointon, and Siddle (2000)
$R_{CH_4} = kp_{CH_4}^{\ 0.85}p_{H_2O}^{\ -0.35}$	Ahmed and Foger (2000)
$R_{CH_4,1} = k_1\left(\dfrac{p_{CH_4}p_{H2O}^{0.5}}{p_{H_2}^{1.25}}\right)\left(1 - \dfrac{p_{H_2}^3 p_{CO}}{k_{eq1}p_{CH_4}p_{H_2O}}\right)\times\dfrac{1}{DEN^2}$ $R_{WGS} = k_{WGS}\dfrac{p_{CO}p_{H_2O}^{0.5}}{p_{H_2}^{0.5}}\left(1 - \dfrac{p_{H_2}p_{CO_2}}{k_{eq,WGS}p_{H_2O}p_{CO}}\right)\times\dfrac{1}{DEN^2}$ $R_{CH_4,2} = k_2\left(\dfrac{p_{CH_4}p_{H2O}}{p_{H_2}^{1.75}}\right)\left(1 - \dfrac{p_{H_2}^4 p_{CO_2}}{k_{eq2}p_{CH_4}p_{H_2O}^2}\right)\times\dfrac{1}{DEN^2}$	Hou and Hughes (2001b)

Rate Equation	Reference
Where $DEN = 1 + K_{CO} p_{CO} + K_{H_2} p_{H_2}^{0.5} + \frac{K_{H_2O} p_{H_2O}}{p_{H_2}}$	
$R_{CH_4,1} = \frac{k_1}{p_{H_2}^{2.5}} \left(p_{CH_4} p_{H_2O} - \frac{p_{H_2} p_{CO}}{k_{eq1}} \right) \times \frac{1}{Q_r^2}$ $R_{WGS} = \frac{k_{WGS}}{p_{H_2}} \left(p_{CO} p_{H_2O} - \frac{p_{H_2} p_{CO_2}}{k_{eq,WGS}} \right) \times \frac{1}{Q_r^2}$ $R_{CH_4,2} = \frac{k_2}{p_{H_2}^{3..5}} \left(p_{CH_4} p_{H_2O}^2 - \frac{p_{H_2}^4 p_{CO_2}}{k_{eq2}} \right) \times \frac{1}{Q_r^2}$ where $Q_r = 1 + K_{CO} p_{CO} + K_{H_2} p_{H_2} + K_{CH_4} p_{CH_4} + \frac{K_{H_2O} p_{H_2O}}{p_{H_2}}$	Hoang et al. (2005)

Currently, several commercial catalysts are being developed. For instance, Hoang et al. (2005) analyzed the MSR kinetics over a commercial catalyst, sulfide Ni/γAl2O3, over 773-1073K. Their model presented the dependency of reforming performance on temperature and steam-to-carbon ratio. It is noted that their MSR kinetic models do not only apply to the reformer, but also the internal reforming approach for solid oxide fuel cells (SOFC). Dicks et al. (2000) and Ahmed and Foger (2000) presented the MSR kinetic models which were investigated over Ni/YSZ anode at 1073 – 1173 K to optimize important system parameters in the DIR-SOFC. According to Dick's model (Table 1) the hydrogen partial pressure presents both positive effect and negative effect of the steam partial pressure on the reaction rates. Ahmed and Foger (2000) presented further simplified rate equations.

In Table 1.1, k, k_1, and k(T) are the rate constants of reaction 1.1. k_{WGS} and k_2 are the rate constants of reactions 2 and 3, respectively. k_{eq} is the equilibrium constant. K_{CH4}, K_{CO}, K_{H2} and K_{H2O} are the equilibrium constants for CH_4, CO, H_2, and H_2O adsorption respectively. p_i is the partial pressure of gas component *i*.

2.1 Modeling of IR-SOFC

The integrated system had been study for designing, controlling and management (Ramadhani et al., 2017). Regarding the designing aspect, the right synthesis design and sizing are the most important part minimizing the cost of investment. Optimization of system design and its size depends on the load, weather conditions and government regulations. Now a day, the 3D model is concerned for predict the momentum, mass and thermal behavior since it could explain the behavior of gaseous working fluid and electronic charges flow fields in general are 3D entities comprising not only plain channels but also solid ribs(Conti et al., 2019).

As the benefit of 3D model, though it consumes the resource of computation for calculation, there are several 3D simulation of SOFC that had been developed since the last decade. Choudhary (2016) used the 3D mathematical model to predict the behavior of a planar anode supported high temperature direct internal reforming SOFC fueled by syn-gas. The model was fabricated under the configuration of Colpan, Dincer, and Hamdullahpur (2007). This mode simulates gases and temperature characteristic along the cell channels. The mismatch temperature between fuel channel and the SOFC had been found. This effect is also found in the work of Mohammad Ebrahimi (2017). They developed the 3D-modeling of planar SOFC to explore the transport phenomena inside a single SOFC and assess it overall performance. They found that incensing of cell temperature is advantage for current density because higher temperature results in lower activation and ohmic loss. Jiang et al. (2020) established 3D modelling of a double-sided cathodes SOFC. They found that around 47%~54% of maximum 1st principal stress in SOFC is caused by the mismatch of coefficients of thermal expansion (CTEs) among materials, while the other part of the maximum 1st principal stress is mainly caused by temperature gradient.

3. Methodology

Since this research mainly focuses on the temperature distribution of autothermal GTL (reformer integrated with SOFC) fueled by methane, it is necessary to investigate both kinetic and thermodynamic behaviors of the interested systems. The rate expressions of steam reforming and the thermodynamic model of the integrated system are investigated in order to predict the temperature profile of the reformer and SOFC. It has to be noted that internal configurations of a reformer and SOFC are also different. These mathematical models were developed using COMSOL. In order to ensure that the simulation results are accurate, an experiment will be set up. The predicted conversion of reactants and products in methane reforming and electrochemical reactions is also validated with the experimental results.

3.1 Mathematic model of IIR-SOFC study

In order to develop a model to predict the temperature distribution of the system, it is first necessary to establish the investigated geometry. In this study, SOFC with the tubular design was selected to be the investigated configuration. Two couples of reformer-SOFC reactor applications were simulated, as shown in Fig. 5. It could be seen that the orders of the components in each system configurations are different to optimize the heat unitization and system efficiency. Secondly, the relationship between mass and energy transport behavior in each channel will be derived to develop the model.

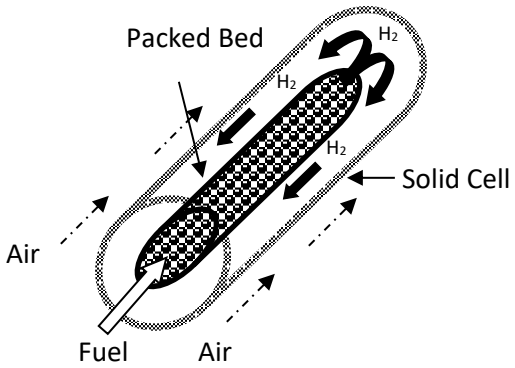

Figure 3.1.

Figure 3-1 Configurations of packed-bed reforming catalyst integrated with FT

3.2 Governing Equations

According to its structure, each component can be separated into porous media (packed-bed) and non-porous media (bulk gas channel). For porous structures, its porosity, tortuosity, and

permeable properties of the material that normally affects the transport behaviors of gases. In this study, there are two main sets of equations: kinetic models and thermal models.

Kinetic Models

The transformation of each reactant in the system is kinetically controlled by the reforming reaction and the electrochemical reaction. Firstly, the methane is reformed to hydrogen and carbon compound gases via the reforming reaction. However, kinetic behavior depends on the type of reforming reaction, as reviewed above. The hydrogen and the carbon monoxide gases are then consumed via the electrochemical reaction. Finally, electricity is generated.

Thermal Model

Heat transfer phenomena in the system involve conduction along with the stack materials, convection from the heat flow through the system, and radiation between gray bodies. It has to be noted that the heat transfer mechanism relies on the system geometry.

- **Conduction**

This phenomenon was investigated by using Fourier's law, Eq. 1

$$\vec{q}_{cond} = -k_{cond} \nabla T \qquad (1)$$

where q_{cond} is the heat flux (W/m^2) from conduction, and k_{cond} is thermal conductivity (W/mK)

- **Convection**

The effect of convective heat transfer is obtained from Newton's law, Eq.2

$$\vec{q}_{conv} = h_{conv}(T_1 - T_2) \qquad (2).$$

where q_{conv} is the heat flux (W/m^2) from convection, and h_{conv} is the convection heat transfer coefficient (W/m^2.K).

- **Radiation**

The net heat flux from heat radiation between two gray bodies is defined on Eq. 3

$$Q_{rad} = \frac{\sigma(T_1^4 - T_2^4)}{\left(\frac{1-\varepsilon}{\varepsilon A}\right)_1 + \frac{1}{A_1 F_{12}} + \left(\frac{1-\varepsilon}{\varepsilon A}\right)_2} \qquad (3)$$

where Q_{rad} is the heat flux (W) from convection, σ is the Stefan-Boltzmann coefficient ε is emittance, A is the area (m^2), and F_{12} is the gray-body transfer factor from surface 1 to surface 2.

Finally, all developed models in this study are calculated based on the algorithm in Fig. 6

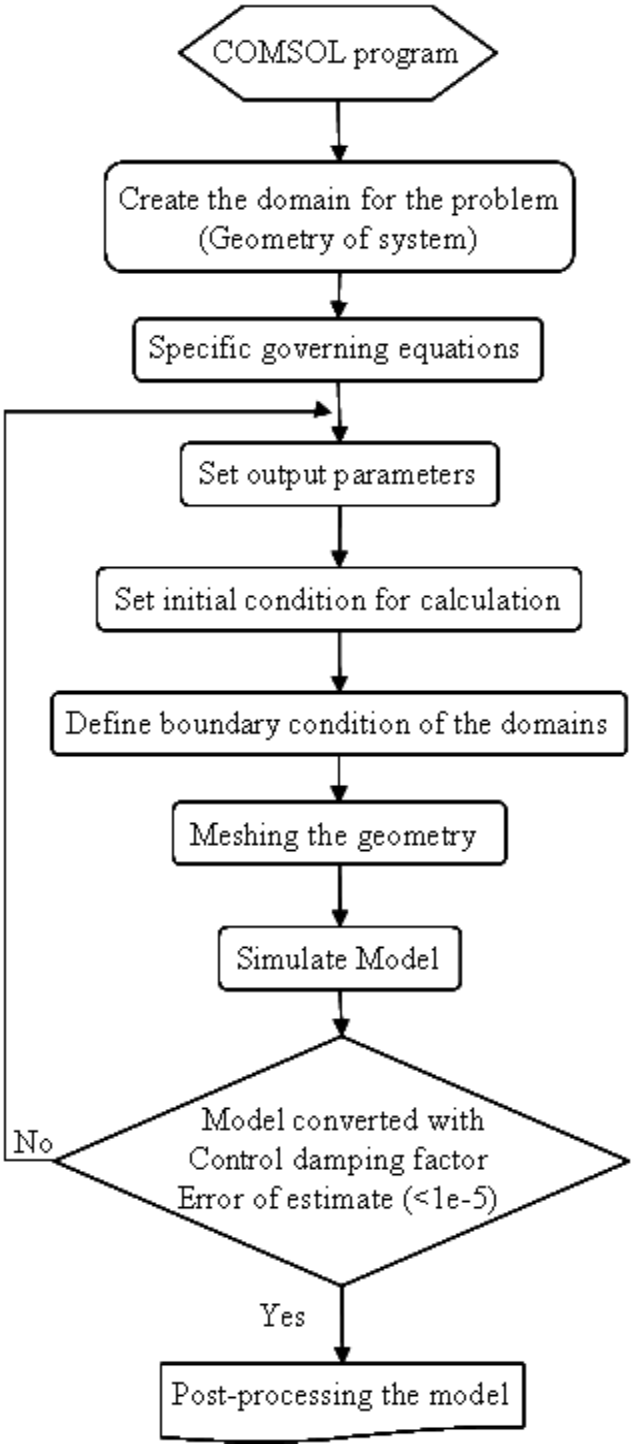

Figure 3-2 Flow Diagram of a coded procedure for COMSOL®

3.3 Framework of Study

According to the aims of this study, mathematical models were developed for several investigated configurations. Many sets of equations were investigated in order to simulate the case studies. The path ways of this study were summarized in the Fig. 3.3. Firstly, kinetic equations of selected primary fuels were review from previous publications. The momentum, mass, and heat transfer equations suitable for the investigated configurations were also theoretically developed. Then, sets of kinetic and mathematic equations were integrated to form the model of IR-SOFC systems. Finally, the reliability of the developed model was validated with experimental results from pervious literatures

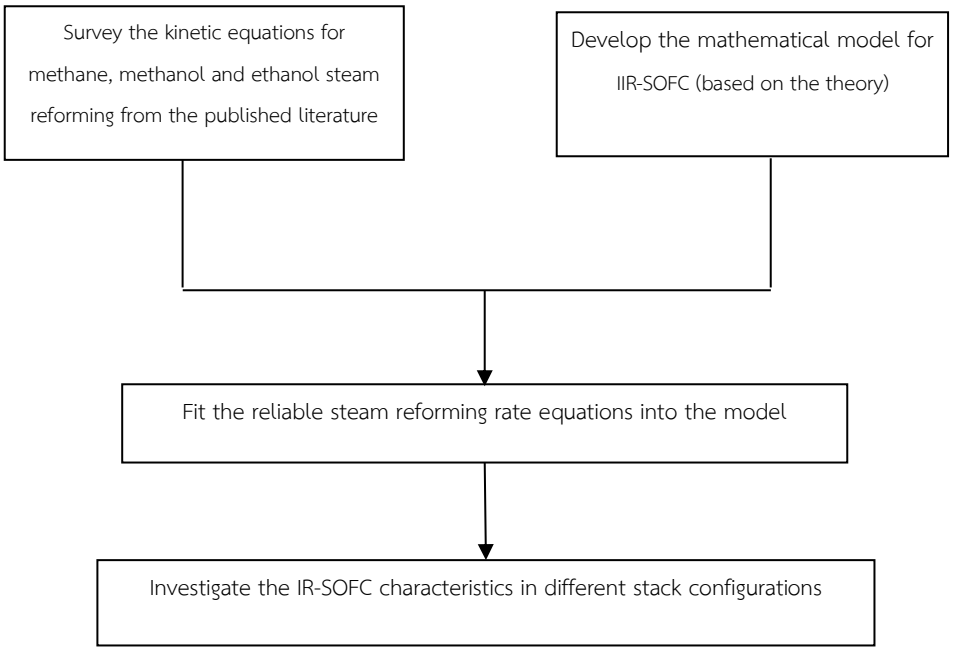

Figure 3-3 Framework of this study

4. Results and Discussion

4.1 Base case

The 3D model of a planar solid oxide fuel cell (SOFC) was established based on the work of (Colpan et al., 2007). As the base case, the model was simulated at 1173 K with normal pressure. In addition, the fuel utilization was always kept at 80%. Based on the same rate of electrochemical reactions of H2 and CO at the anode, the obtained electrical power can be directly used to compare the electrical efficiency among different operations. The methane was directly fed to the anode side of the cell. Meanwhile, the O_2 stream was fed counter to the methane stream. The simulation result of the base case as shown below. It could be seen in Figs. 4.1&4.2, methane is converted to hydrogen nearly complete. At the same time, the hydrogen was consumed at the anode/electrolyte interface to generate electricity, as seen in Fig. 4.3. As the configuration of SOFC was fabricated as a microchannel reactor. The mismatch of the rate of exothermic steam reforming reaction and endothermic electrochemical reactions is not extreme. Thus, the temperature distribution is varied around 20 K. During the hydrogen is consumed, the electricity is also generated. The behavior of power generation along the cell is shown in Fig 4.4. Noted that the carbon formation from reforming reaction is also perdicted in Fig. 4.5. It could be seen that more carbon deposition where the higher reforming reaction took pleaced.

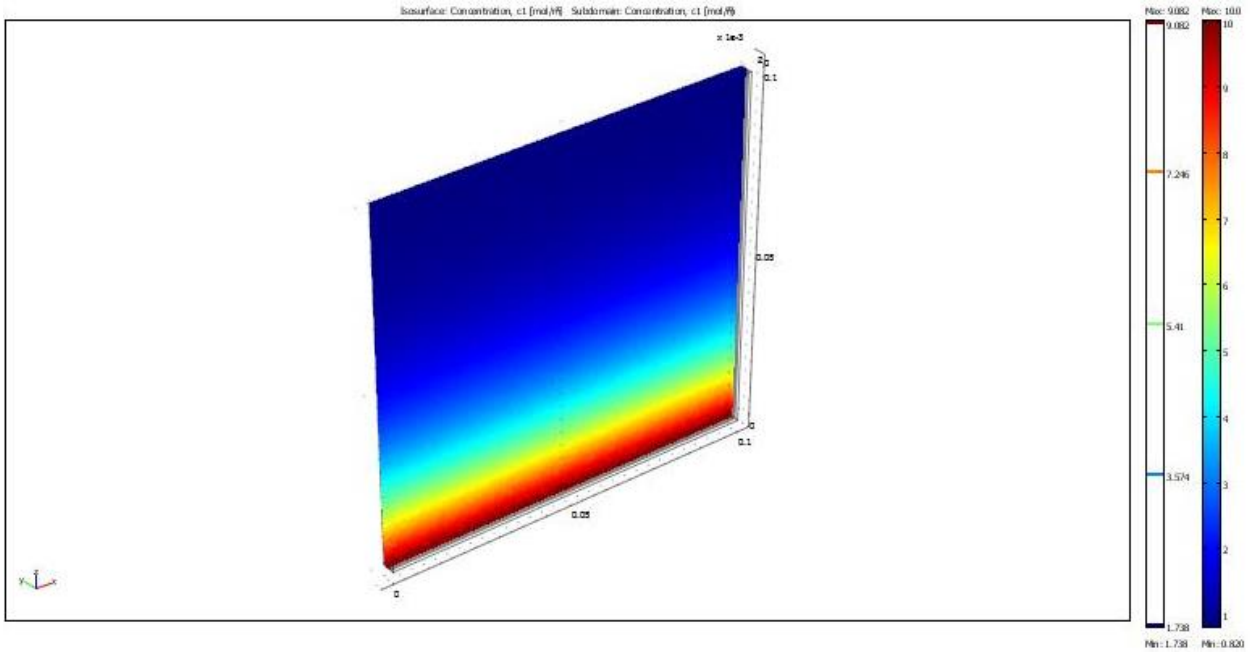

Figure 4-1 Methane Conversion at the based case.

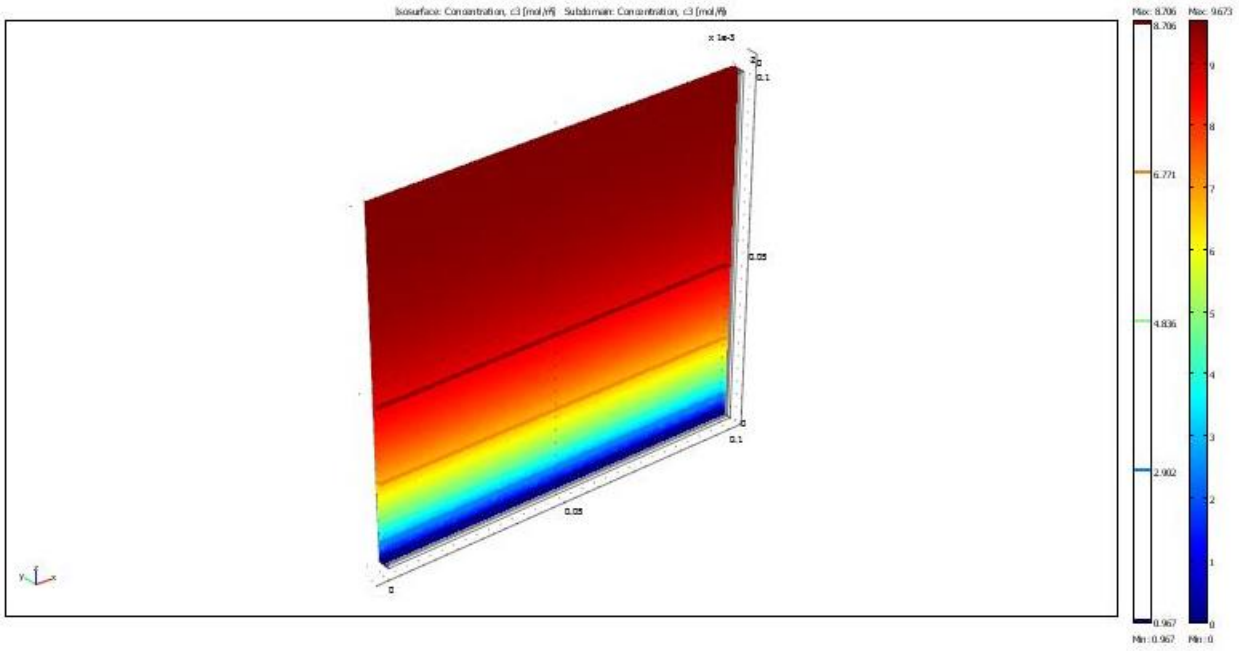

Figure 4-2 Hydrogen Conversion at the based case.

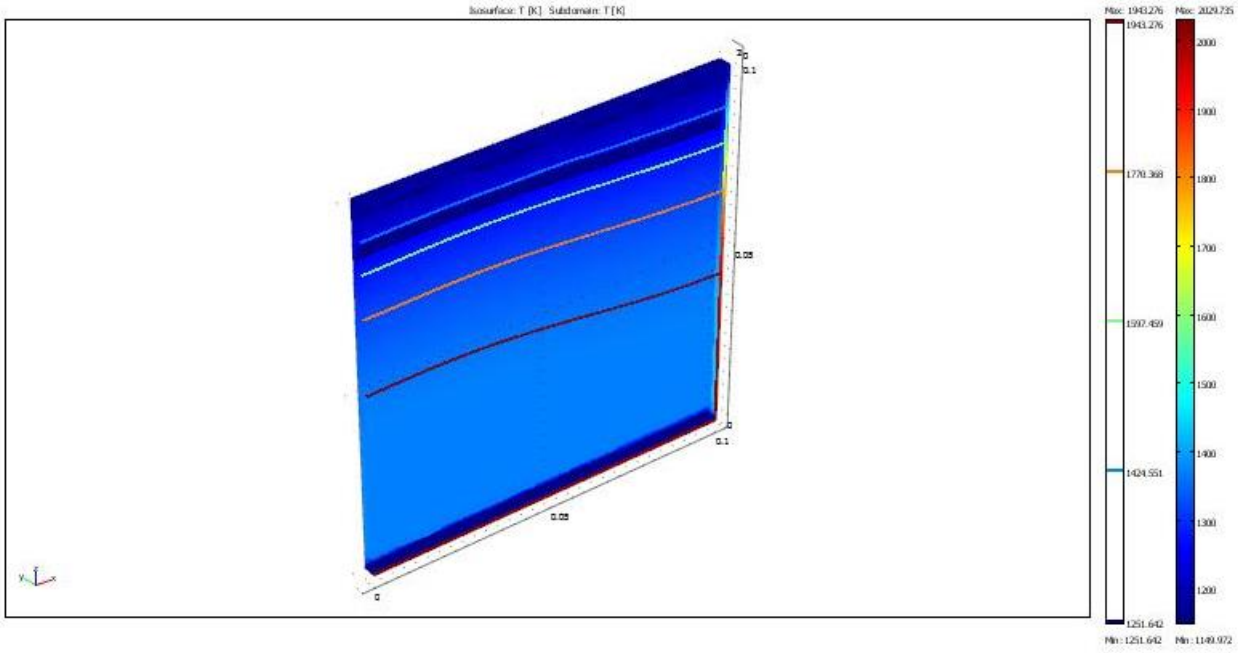

Figure 4-3 Temperature distribution at the based case

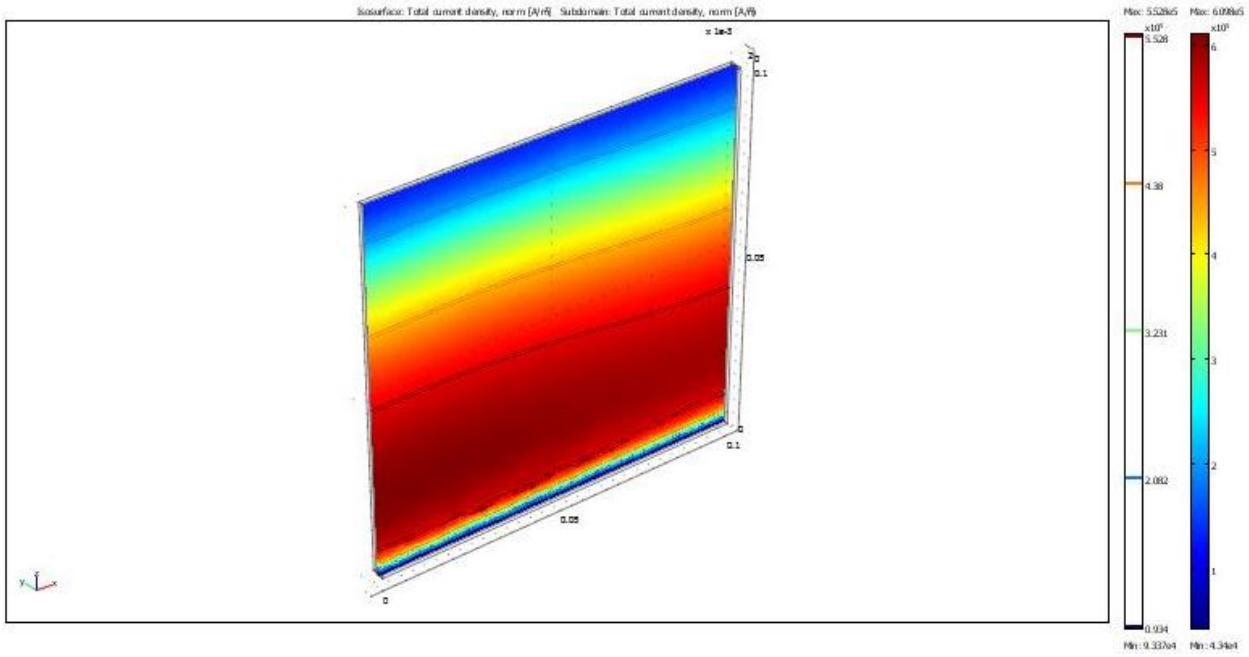

Figure 4-4 Generated electricity along the SOGFC at the based case.

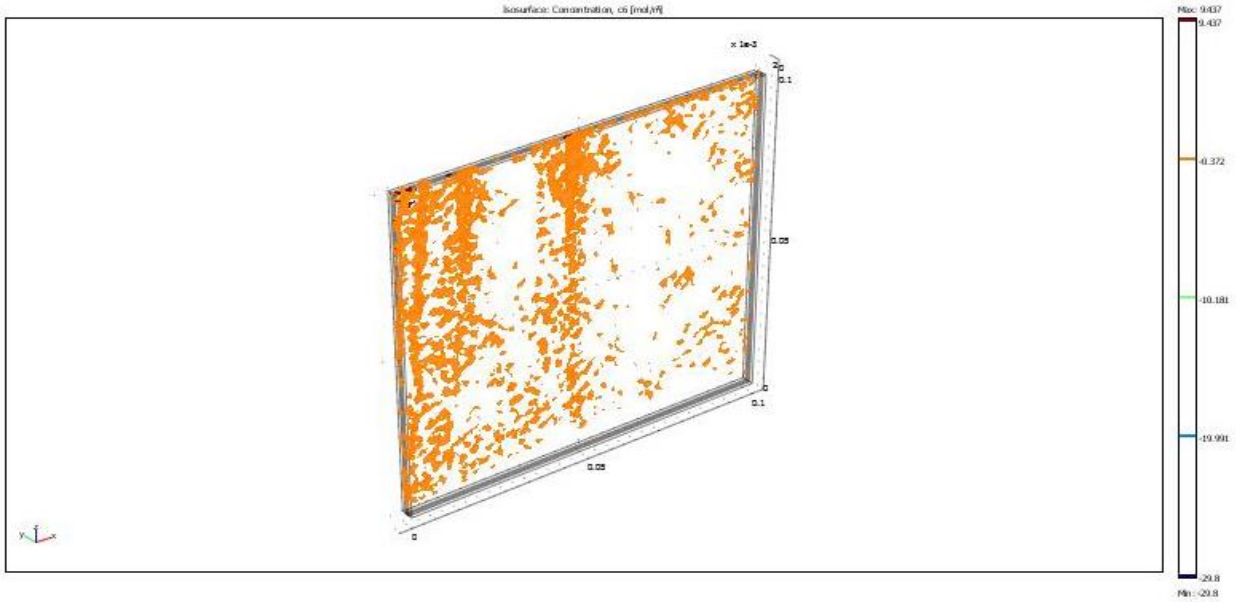

Figure 4-5 Carbon formation along the SOFC at the based case.

4.2 Effect of Flow Direction

For typical autothermal application e.g. heat exchanging system, flow direction of exchanged fluids strongly affects the heat transfer and reaction behavior in the fluid stream, thus the effect of fuel and oxidant flow direction on the IIR-SOFC performance was also considered here. In the previous sections, air flow is counter-flow to fuel flow in the fuel channel. As an alternative option, the system behavior was analyzed as co-flow pattern by changing mass and energy balances in air channel along with their corresponding boundary conditions while keeping all other operating conditions identical to those of counter-flow pattern. Figs. 4.6 -4.9 show the effect of co-flow pattern on the temperature profiles along the fuel channels. It can be seen that the flow directions of fuel gases and air strongly affect the temperature gradient along the system.

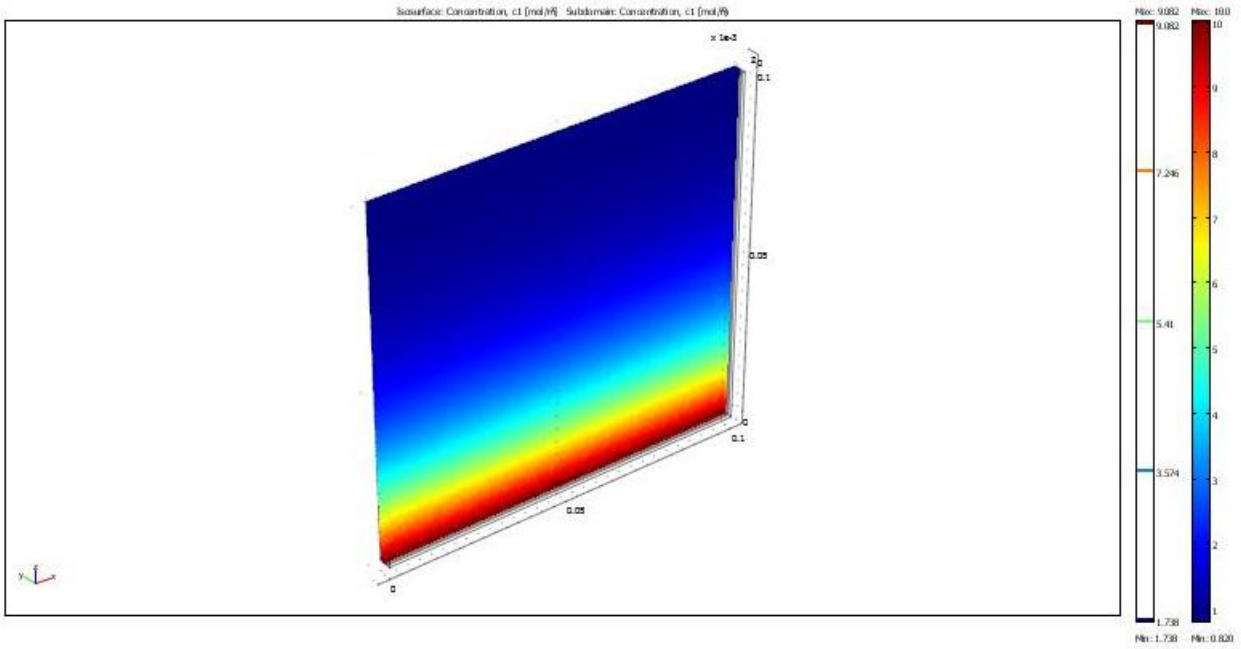

Figure 4-6 Methane conversion under co-flow pattern.

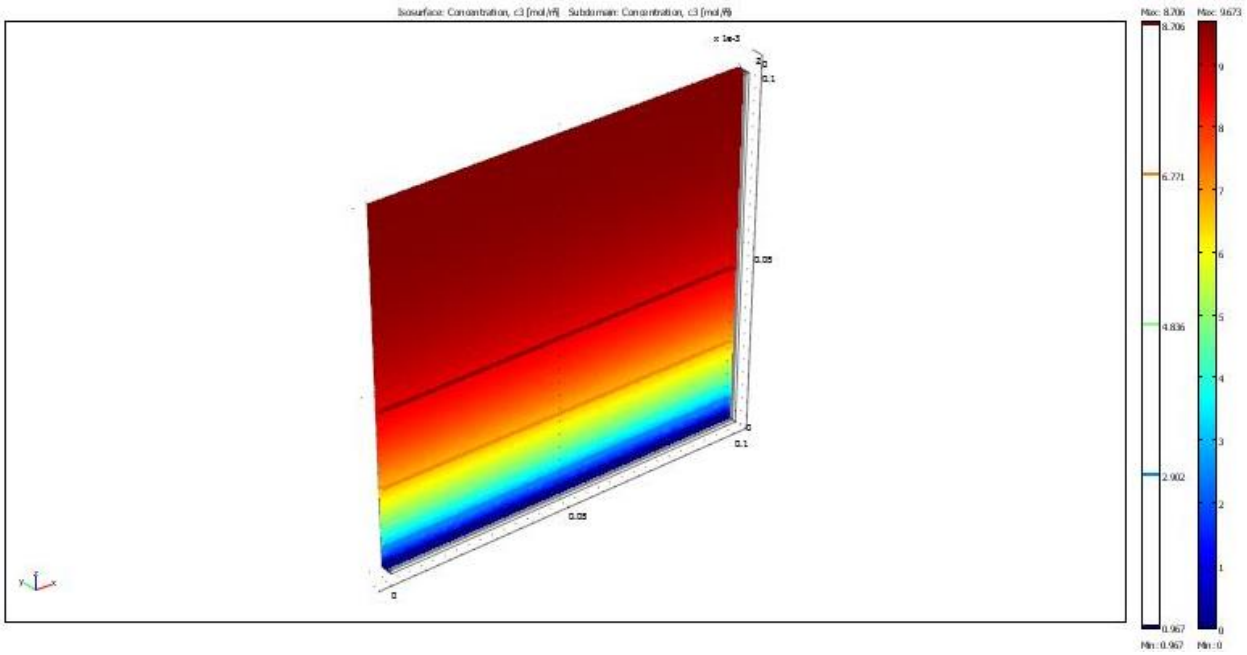

Figure 4-7 Hydrogen conversion under co-flow pattern.

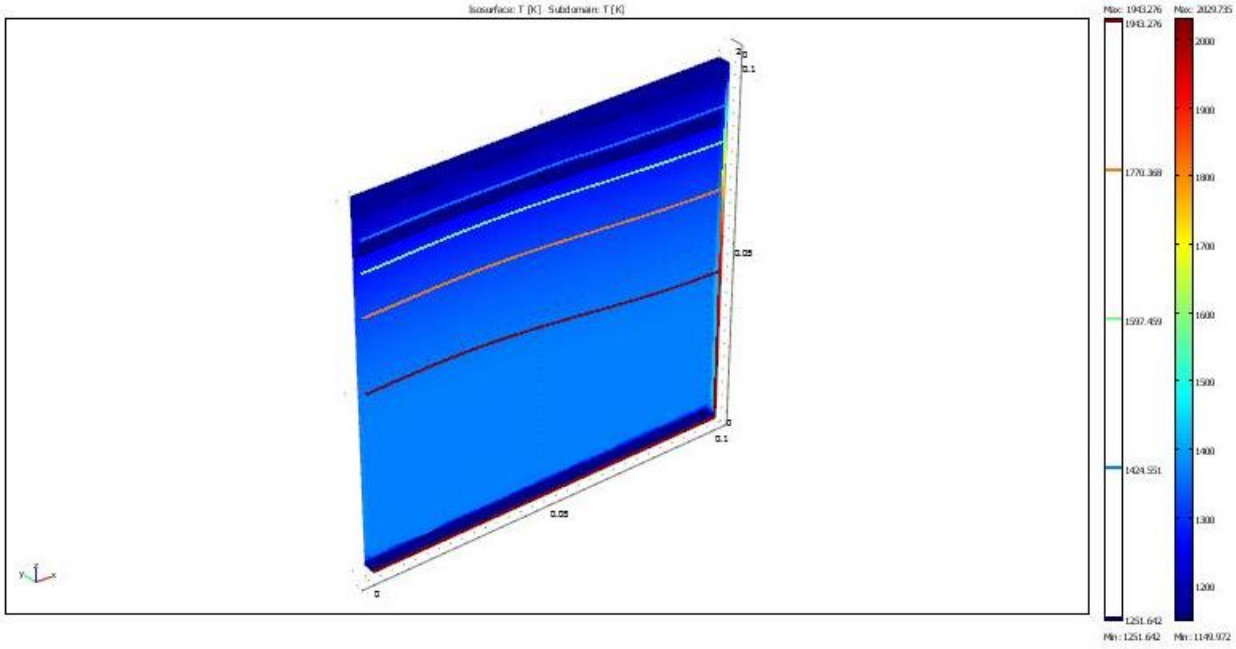

Figure 4-8 Temperature distribution under co-flow pattern.

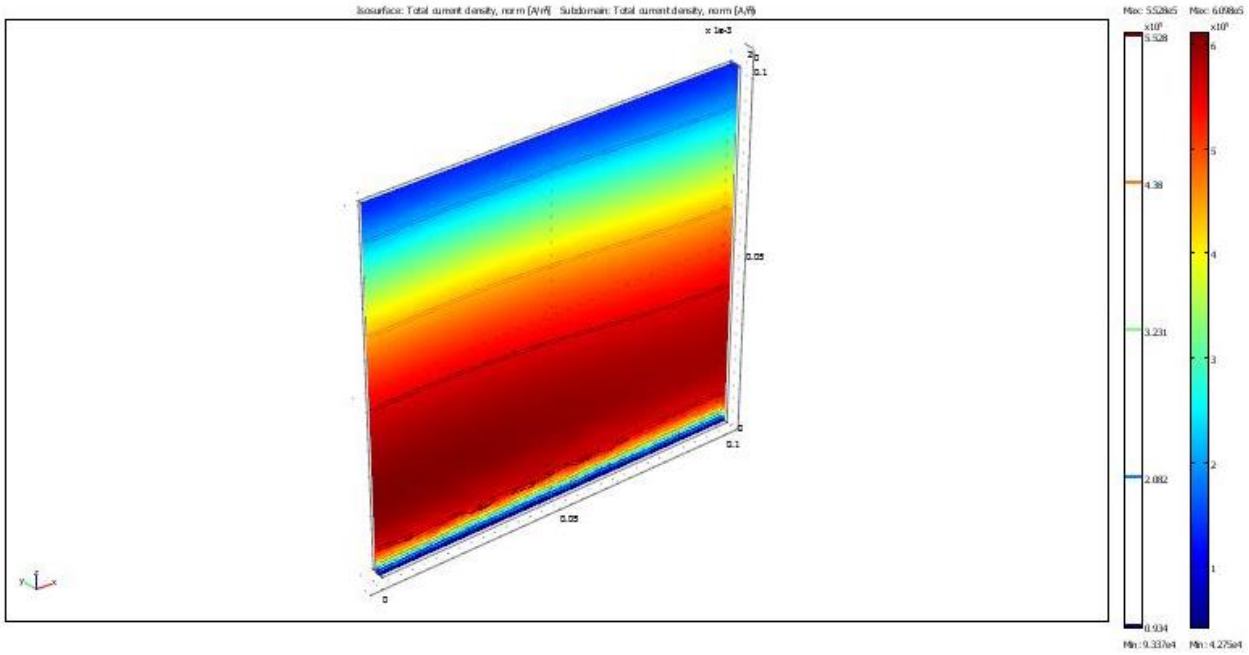

Figure 4-9 Generated electricity along the SOFC under Co-flow pattern.

4.3 Effect of Operating Temperature

Since the methane oxidation is an exothermic reaction, it is thermodynamically favored at lower operating temperature. This is in contrast with the electrical properties of SOFC which prefers higher operating temperature in order to reduce the cell polarizations. Therefore, the effect of operating temperature was considered in this present work. Figs. 4.10-4.13 presents the effect of operating temperature equal to 1073 on the chemical electrical and temperature profiles along the IIR-SOFC. Meanwhile, Figs. 4.14-4.17 show the effect of operating temperature equal to 973 on the chemical electrical and temperature profiles along the fuel channels. It can be seen that, at lower operating temperature, temperature distribution is smoother than that of high operating temperature. However, the electrical efficiency is relatively low.

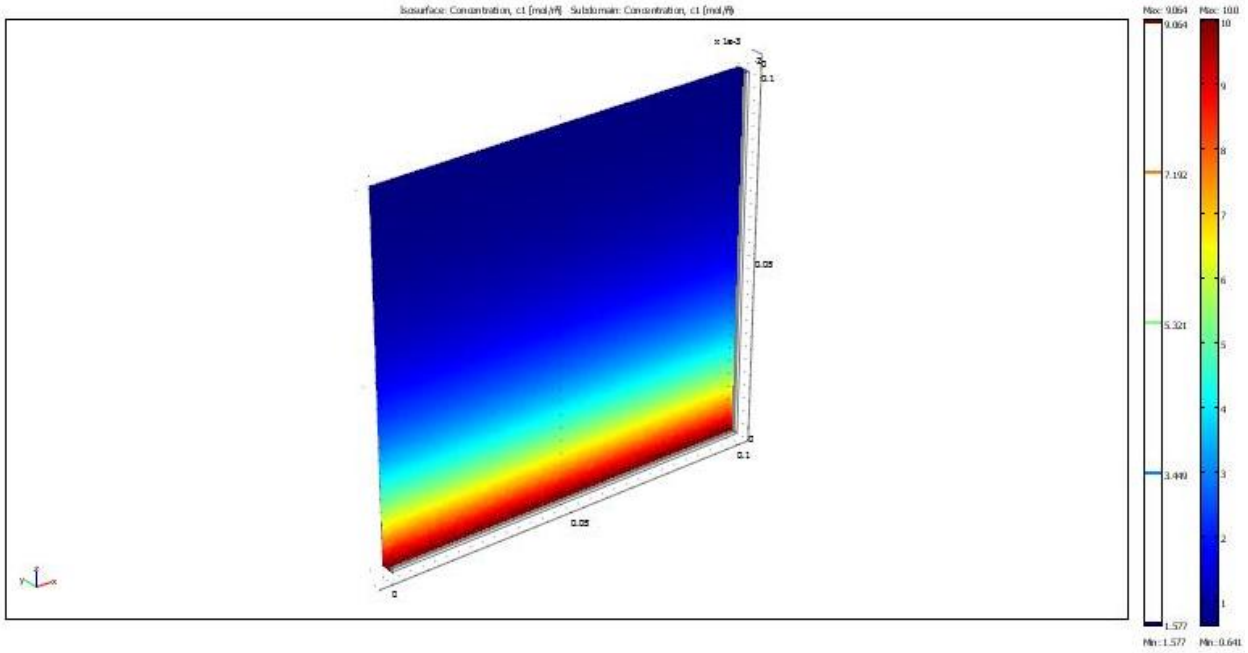

Figure 4-10 Methane conversion at operating temperate = 1073 K.

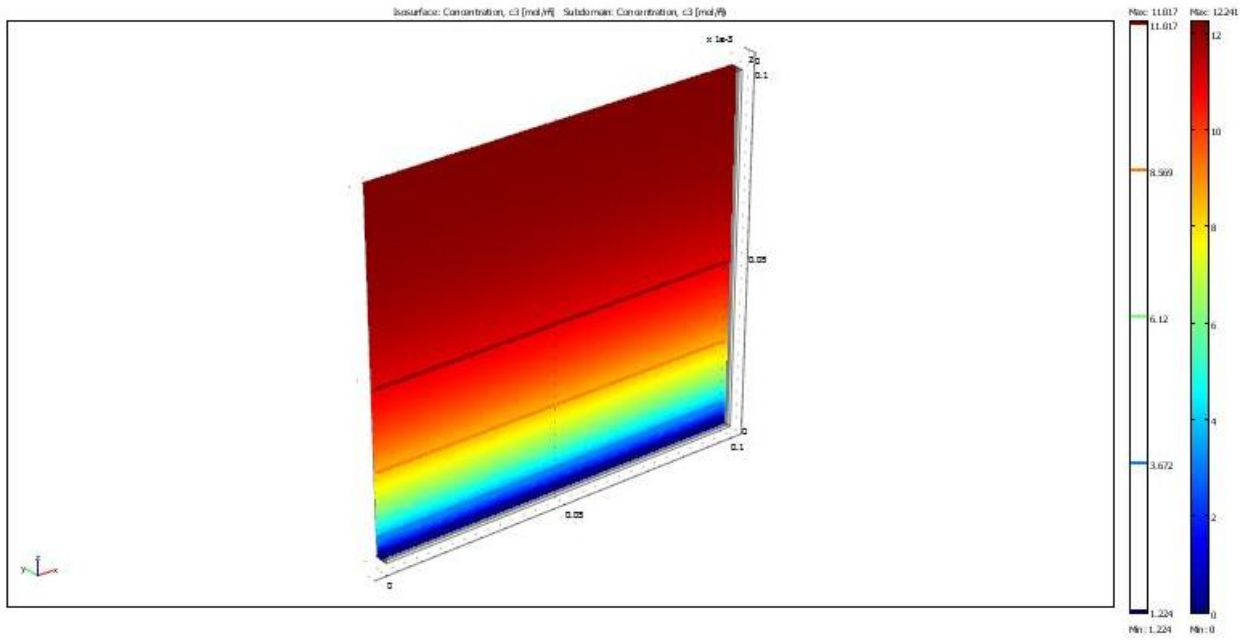

Figure 4-11 Hydrogen concentration at operating temperate = 1073 K.

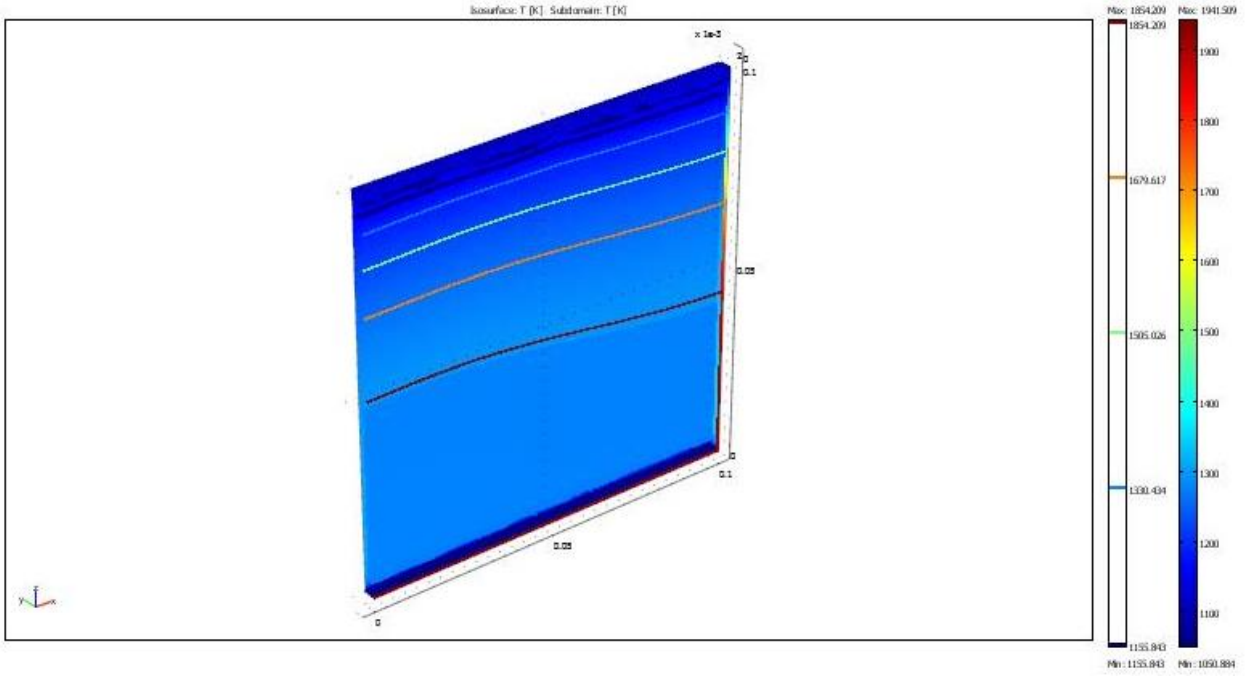

Figure 4-12 Temperature distribution at operating temperate = 1073 K.

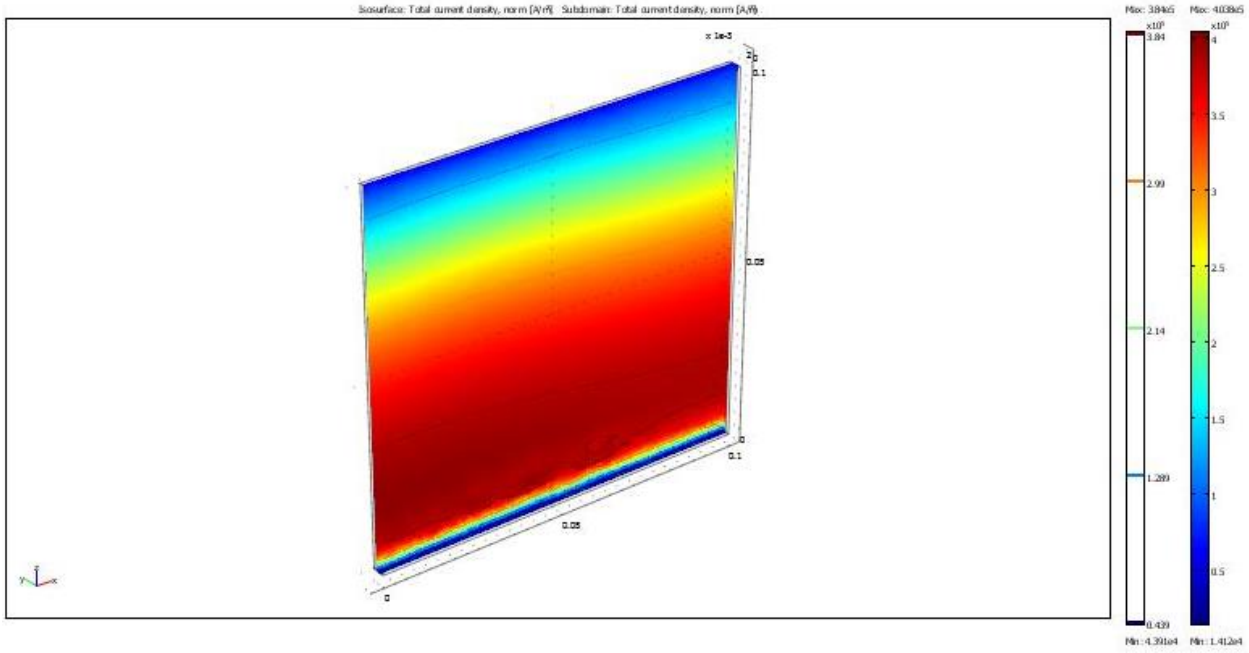

Figure 4-13 Generated electricity along the SOFC at operating temperate = 1073 K.

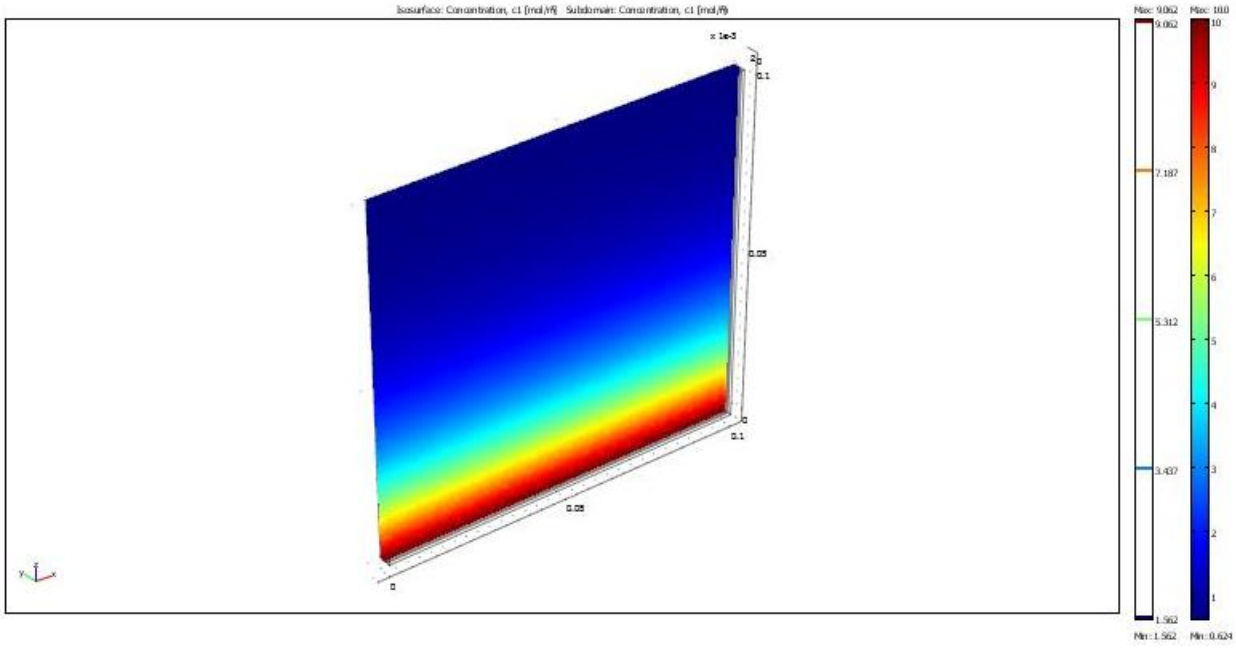

Figure 4-14 Methane conversion at operating temperate = 973 K.

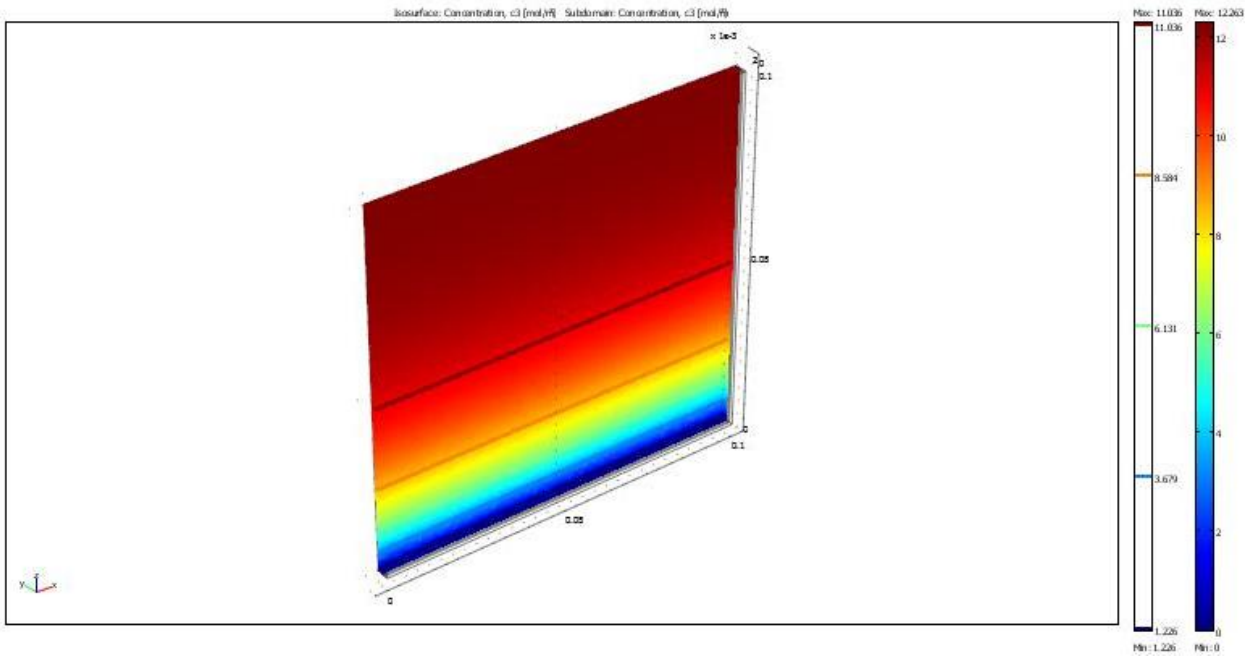

Figure 4-15 Hydrogen conversion at operating temperate = 973 K.

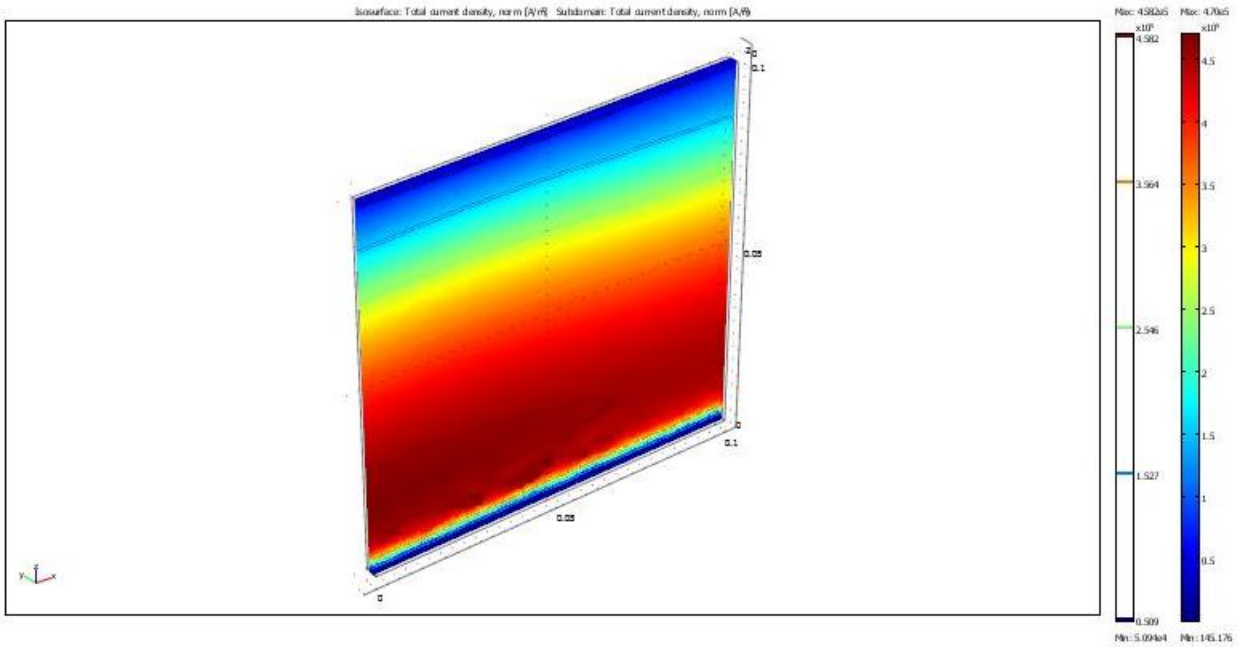

Figure 4-16 Generated electricity along the SOFC at operating temperate = 973 K.

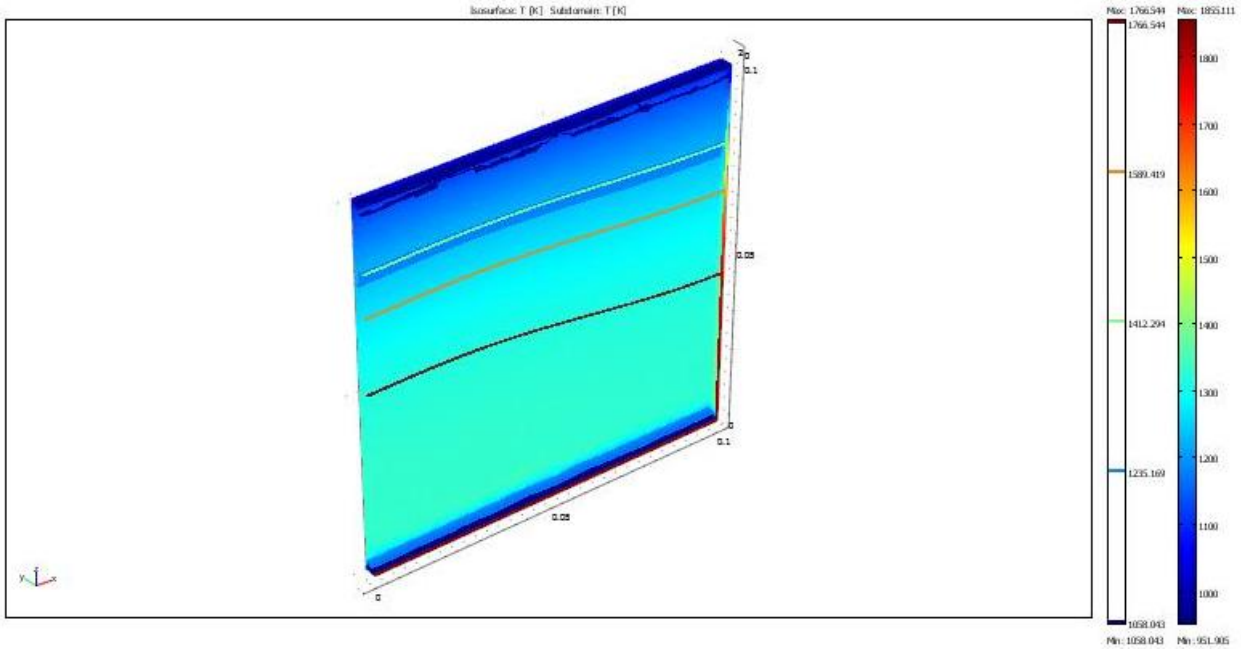

Figure 4-17 Temperature distribution at operating temperate = 973 K.

4.5 Effect of inlet S/C ratio

To minimize the potential for carbon deposition in the cell and maximize the performance of the system, the suitable inlet *S/C* ratio is also validated in Figs. 4.18- 4.21. It can be seen that the amount of carbon formation decreased with increasing inlet S/C ratio since the excess steam can prevent the formation of carbon species from the methane cracking reaction. Nevertheless, considering the temperature gradient and electrical efficiency, it was found that the cooling spot increased with increasing the inlet S/C ratio.

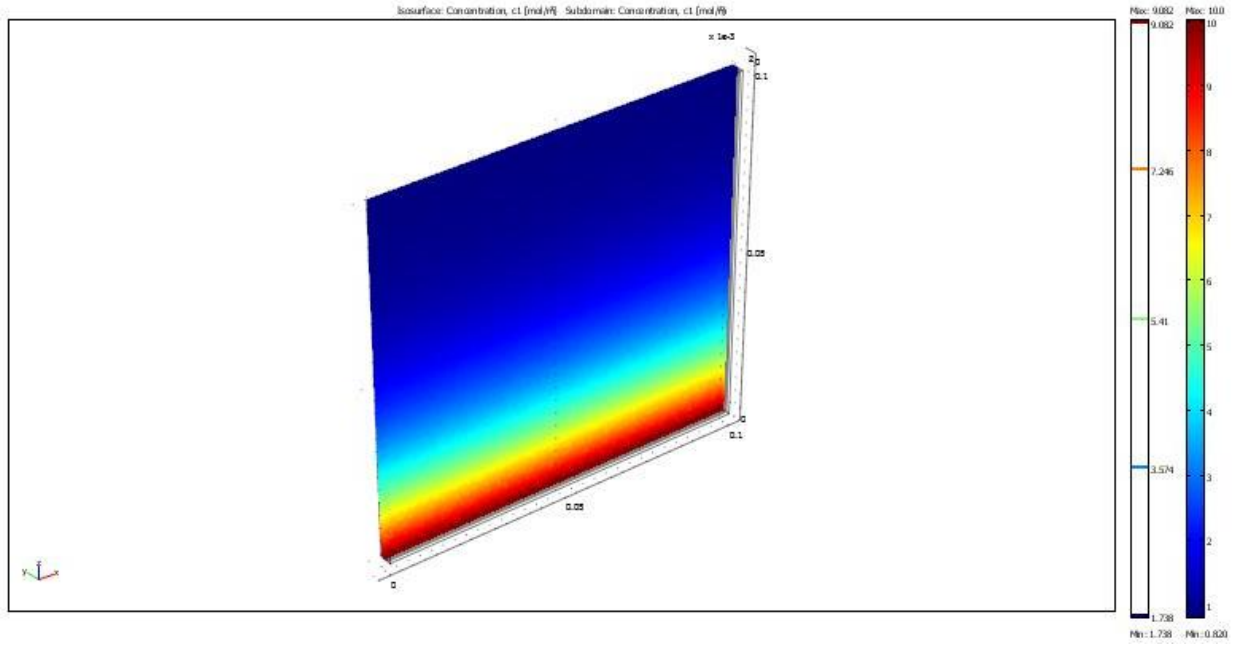

Figure 4-18 Methane conversion at S:C ratio =1:1.

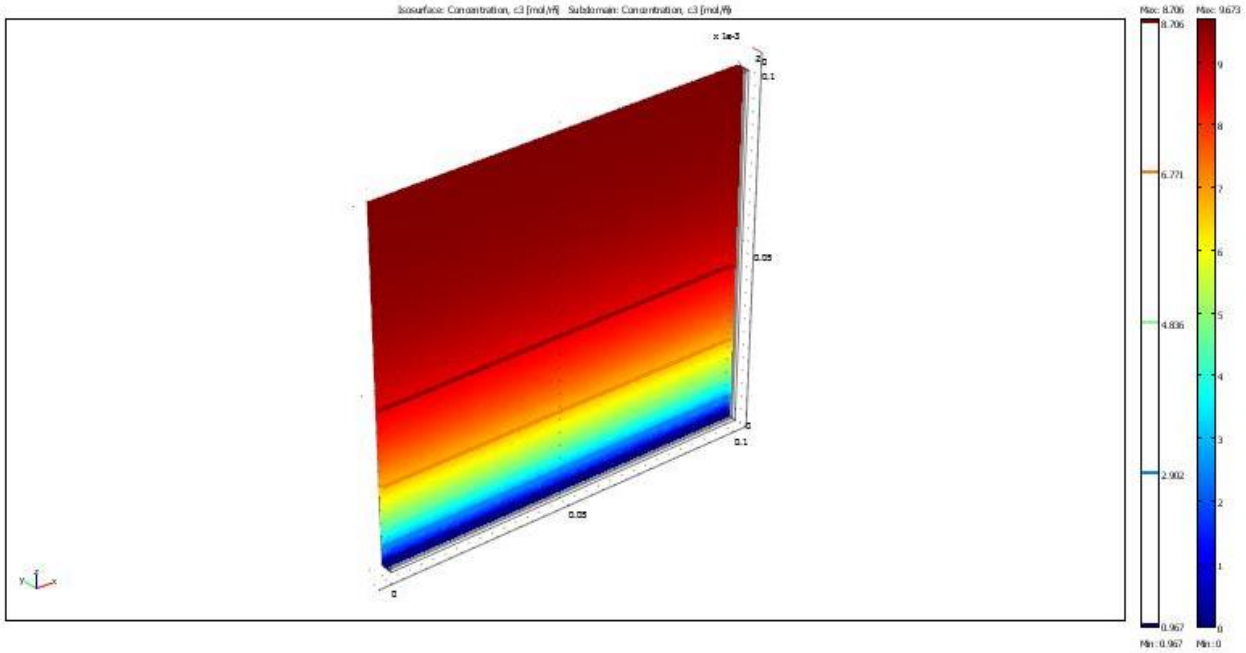

Figure 4-19 Hydrogen conversion at S:C ratio =1:1.

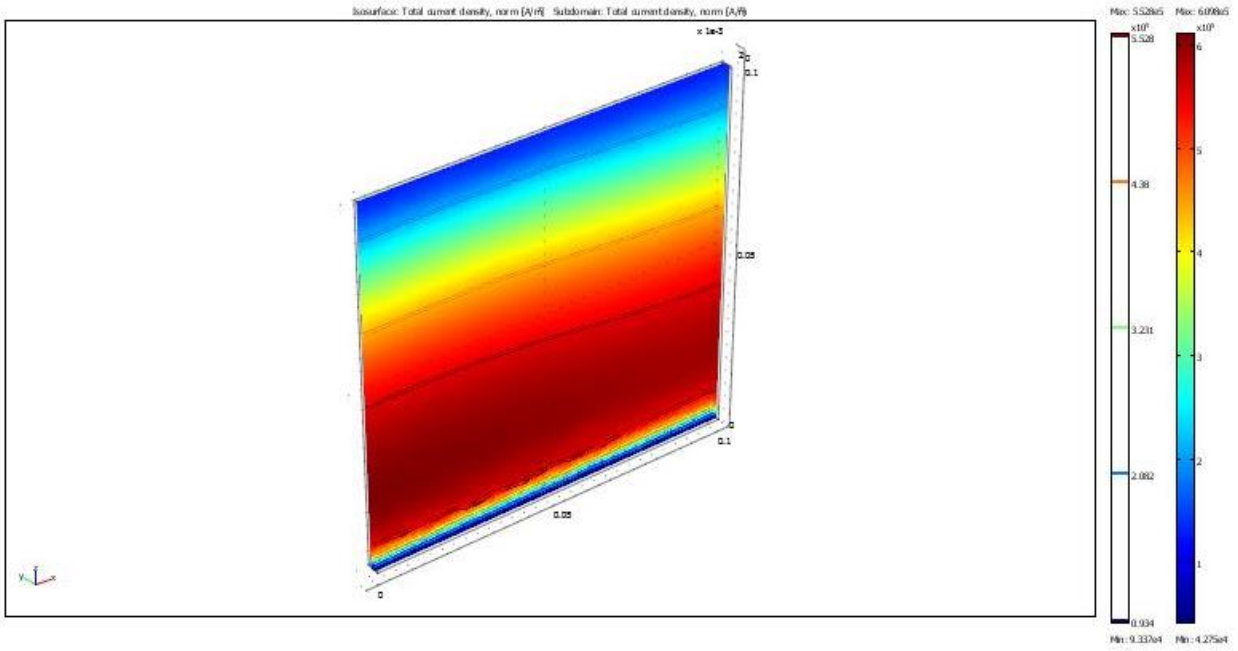

Figure 4-20 Generated electricity along the SOFC at S:C ratio =1:1.

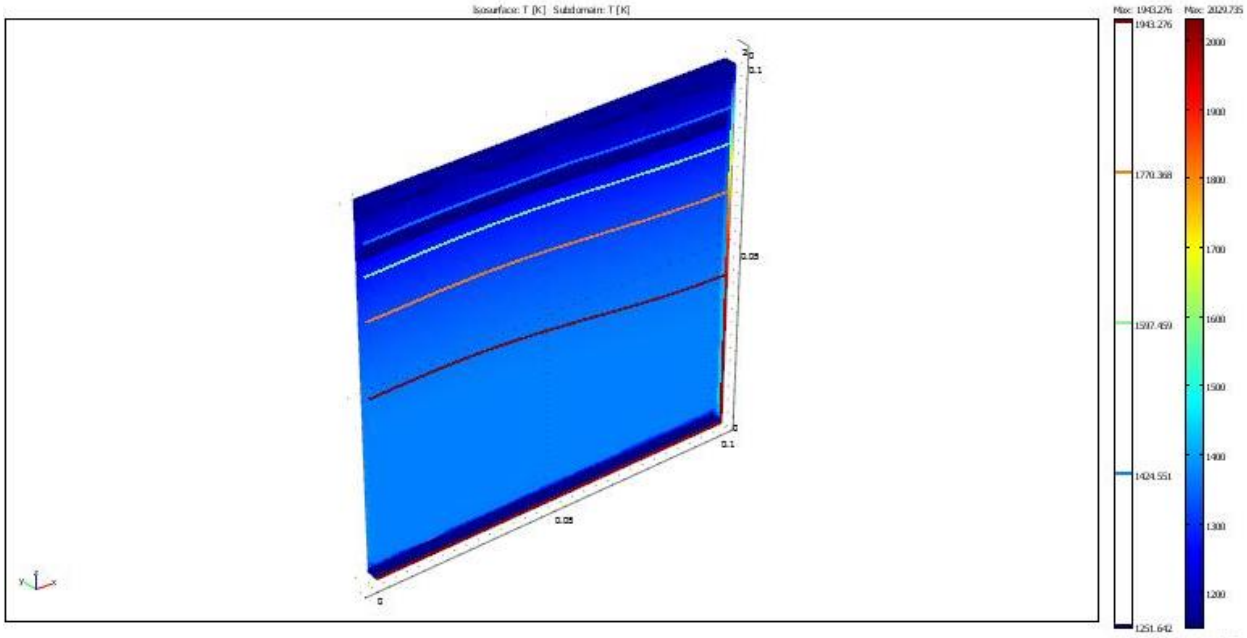

Figure 4-21 Temperature distribution at S:C ratio =1:1.

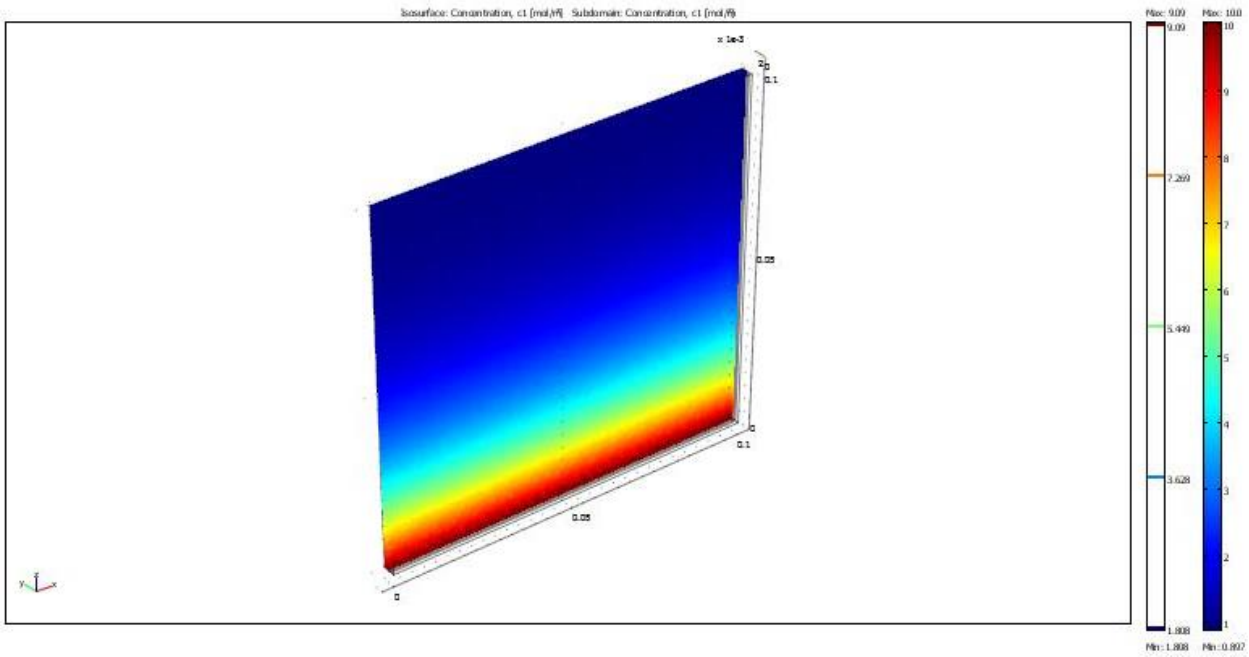

Figure 4-22 Methane conversion at S:C ratio =1:3.

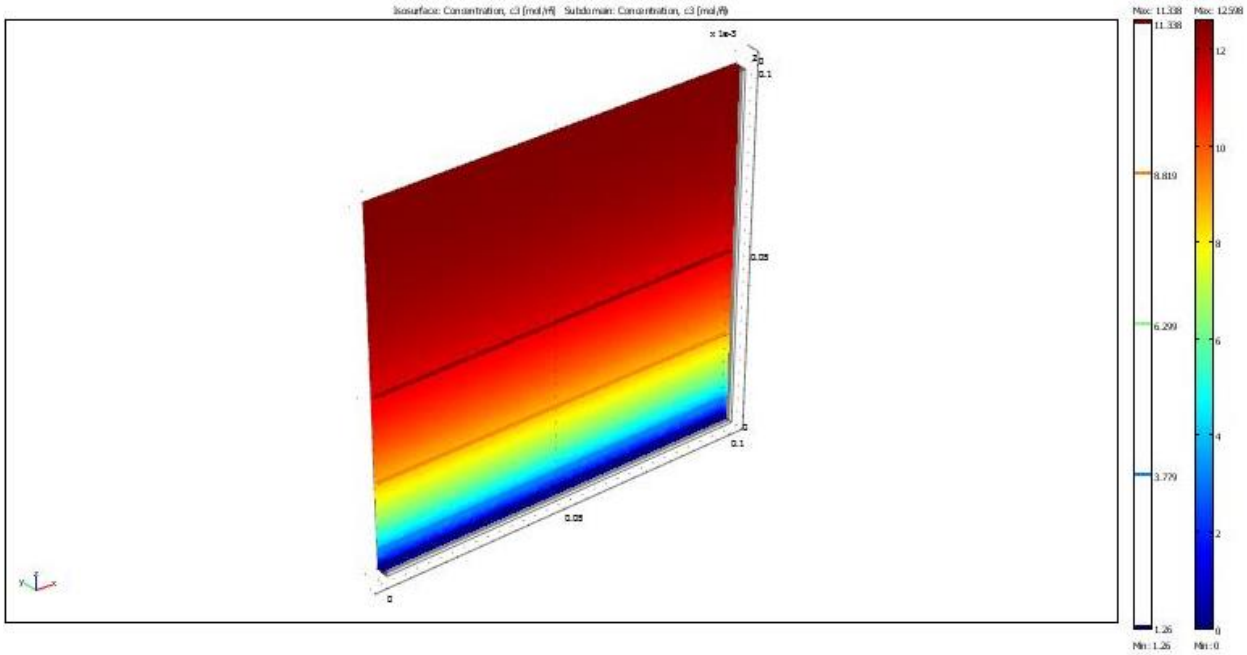

Figure 4-23 Hydrogen conversion at S:C ratio =1:3.

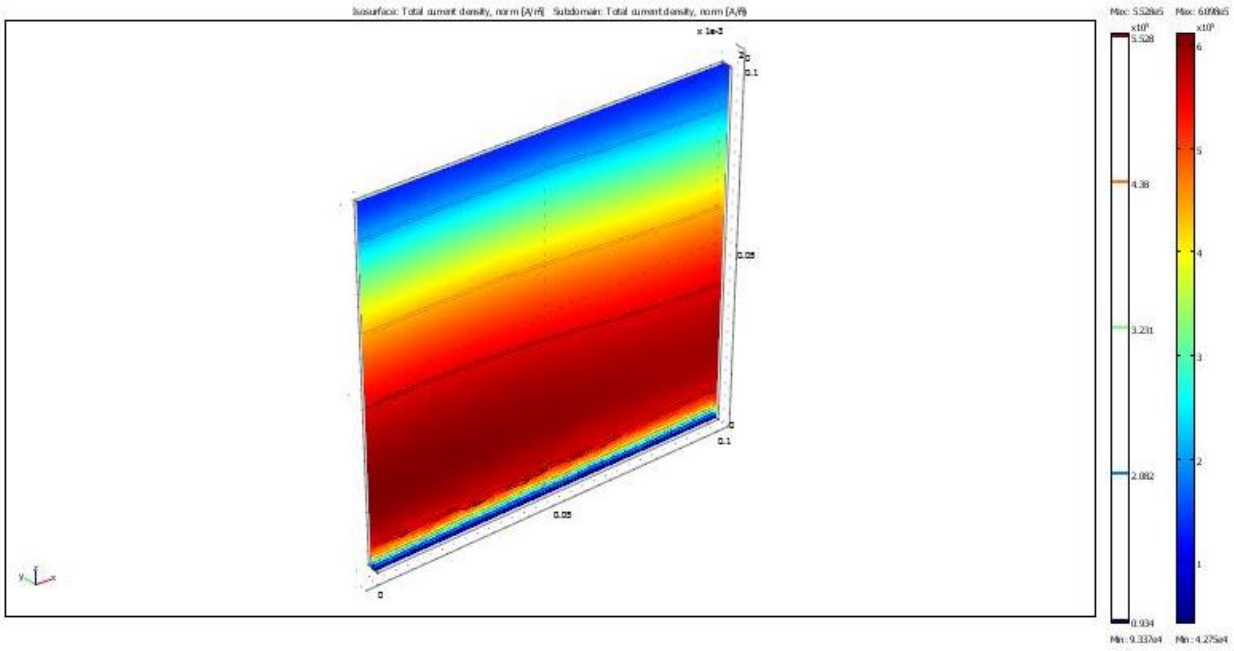

Figure 4-24 Generated electricity along the SOFC at S:C ratio =1:3.

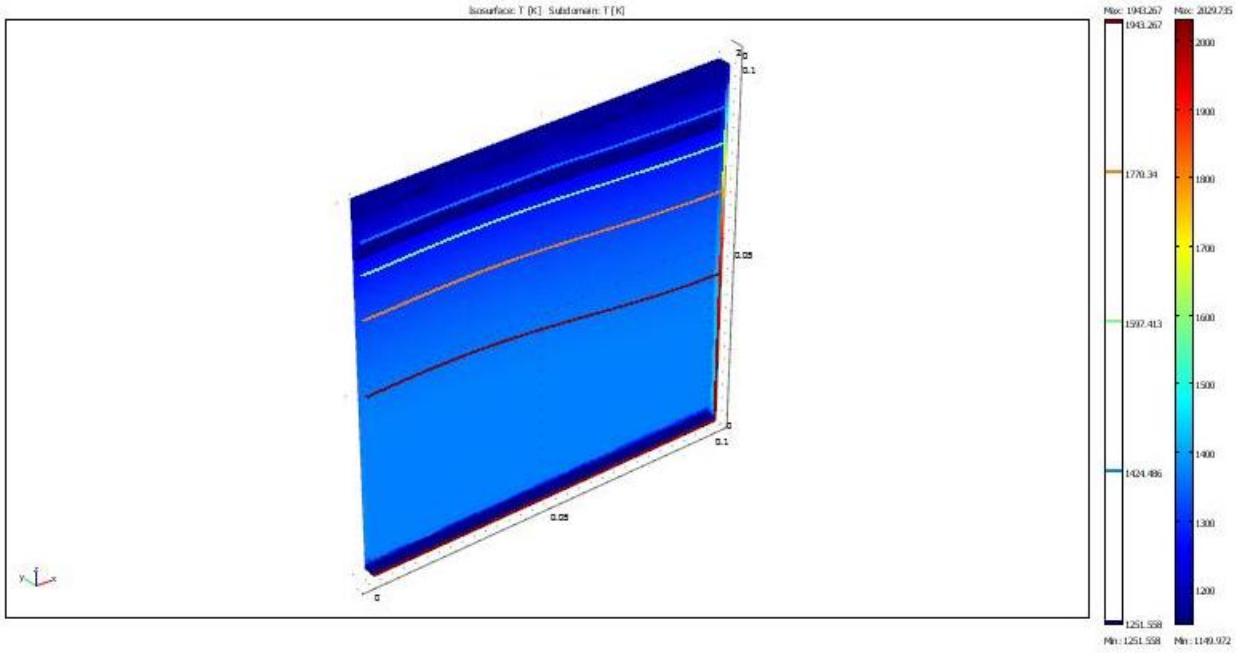

Figure 4-25 Temperature distribution at S:C ratio =1:3.

5. Conclusion

In this present work, a three-dimensional model of an IIR-SOFC fueled by methanol was first developed. The simulations indicated that temperature gradient still occurred at the entrance of the reformer chamber. Greater temperature profile along the system can be obtained by applying the catalyst with low reforming reactivity and increasing the inlet steam-to-carbon ratio. We also found from this work that an IIR-SOFC with a co-flow pattern (co-flow of air and fuel streams through the fuel cell) provides higher voltage and smoother temperature gradient along the system than that with a counter-flow pattern due to better matching between the heat exothermically supplied from the electrochemical reaction and the heat required for the endothermic steam reforming reaction along the fuel cell system. Therefore, we concluded here that the performance of an IIR-SOFC fueled by methanol can be maximized by applying a co-flow pattern and operated with high operating pressure.

Based on using methane as a primary fuel, physical configurations of tubular IIR-SOFC were studied. First, the performance of the IIR-SOFC with a conventional packed-bed internal reformer. Furthermore, it was found that the performance of IIR-SOFC fueled by methane can be maximized by applying catalytic coated-wall internal reformer with a co-flow pattern and operated at high operating pressure.

Finally, the benefits of the autothermal reforming reaction (coupling of exothermic methane oxidation and endothermic methane steam reforming reactions) were simulated based on the IIR-SOFC design. This SOFC system was operated at intermediate temperature (973 K). The simulation reveals that cooling effect at the entrance of reformer could be reduced by adding small amount of oxygen into IIR-SOFC configuration; nevertheless, the system's efficiency decreased. It was also observed that the electrical efficiency could be increased when the system was operated at lower S/C ratio or higher operating temperature but the temperature distribution in the IIR-SOFC system became more undesirable.

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