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**Permalink** https://escholarship.org/uc/item/4t26r6g9

**Journal** ECS Transactions, 42(1)

**ISSN** 1938-5862

**ISBN** 978-1-60768-339-1

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Publication Date 2012-04-20

## DOI

10.1149/1.4705501

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#### ECS Transactions, 42 (1) 247-263 (2012) 10.1149/1.4705501 ©The Electrochemical Society

## Life Cycle Analysis of Ceramic Anode-Supported SOFC System Manufacturing Processes

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The environmental impact of manufacturing an yttrium-doped strontium titanate (SYT) anode-supported planar solid oxide fuel cell (SOFC) system has been examined. Key components of an SOFC system including anode substrate, PEN (Positive electrode-Electrolyte-Negative electrode) structure, interconnects, and balance of plant (BoP) have been evaluated in terms of energy consumption and key air emissions using the Life Cycle Analysis (LCA) approach.

## Introduction

Current state-of-art SOFC systems most frequently use Ni/YSZ cermet anode materials, which possess excellent conductivity and catalytic activity. However, Ni/YSZ has many disadvantages, including nickel coarsening, sulfur poisoning and carbon deposition which can hinder the direct utilize of practical hydrocarbon fuels. What is more, nickel and nickel oxide may lead to allergies or cancer, adding difficulties in handling the material during manufacture (1).

Recently yttrium-doped strontium titanate (SYT) is considered to be a promising alternative SOFC anode material (1-6). Several studies demonstrated that SrTiO<sub>3</sub>-based materials satisfied the anode requirements well, being thermodynamically stable in anodic conditions, electronically and ionically conductive, chemically compatible with the electrolyte and interconnect, and has similar thermal expansion coefficient as other cell components (7-10). One of the most attractive properties of SYT compared to the nickel-based anode is the intrinsic sulfur tolerance and coking resistance, which indicate that practical fuels could be directly utilized in SOFC without addition of steam and extra balance of plant (BoP). These properties further enable SOFC use in stationary power generation to utilize fuels that are currently used such as natural gas and coal syngas.

For anode-supported SOFC manufacturing, tape casting is often perceived to be the most cost-effective process to manufacture the anode substrate. In this study, SYT based anode substrates were manufactured via solvent-based tape casting, non-toxic solvent-based tape casting and water-based tape casting processes. The development of the tape casting industry clearly shown that solvent-based tape casting outweigh water-based tape casting in terms of tape performance, because of it has higher tolerance to variations in influencing parameters (11). And solvent-based tape casting generally requires less energy to dry to form the green tape. However, the environmental impact of some of the organic solvents should be considered and evaluated. Compared to solvent-based, water-based tape casting is generally less expensive and more environmentally benign.

Analyses of the manufacture of the SOFC anode support should be carried out to carefully evaluate the overall merits of water-based system and compare to the traditional organic solvent-based system. As it was identified in other's study, more energy was required in the drying process in aqueous tape casting, and in some cases, the emissions and environmental impact were equally balanced for both solvent-based and water-based systems. It is important to find out whether the extra emissions associated with generating the energy required drying the aqueous systems is less equivalent to the environmental burden associated with the emissions introduced by the organic solvent.

To evaluate the overall environmental impact of ceramic anode-supported SOFC system manufacturing, SYT-based anode-supported SOFC were manufactured and evaluated using Life cycle analysis in this study. Life cycle analysis (LCA) is an analysis tool that accounts for the total emissions and impacts of a product or item throughout its life cycle (from cradle to grave). In this study, the LCA approach is used to assess 1) SYT-based anode substrates manufacturing via three different types of tape casting processes and 2) two types of SOFC systems which are an SYT ceramic based anode-supported planar SOFC system and a traditional Ni-YSZ based planar SOFC system. This study is focused on the stage of anode production and fuel cell system fabrication, which includes raw materials production and manufacture of the stack and the balance of plant.

### Methodology

The life cycle analyses are conducted to investigate the interaction between the manufacturing stage of the life cycle and the environment. In this study, three different tape casting formulas have been developed in our center for SYT-based anode-supported SOFCs, and the environmental impacts of these formulas applied in the manufacturing are evaluated and compared using LCA approach. The material input results are calculated based upon the anode geometry and porosity assumption combined with the tape casting formulas developed, while the energy input data are obtained from various sources (12-14). To further evaluate the environment impact of an SYT-based anode-supported SOFC system, an SOFC system including interconnect and BoP have been evaluated and compared to a traditional Ni-YSZ-based anode-supported planar SOFC system. To calculate the emissions associated with the process energy, emission factors are derived from EPA's eGRID database and other sources for both California and U.S. average electric power generation emission factors.

#### Boundaries of the Study

Life cycle analysis is used to assess the environmental burden of a product, process or activity over its entire life cycle starting with raw materials extraction and ending with the final waste disposal (13, 14). The four steps of an LCA are (13, 14): 1) Goal definition and scoping to define the purpose of the study. 2) Inventory analysis: Quantify the energy and raw material requirements, air emissions, waterborne effluents, solid waste and other environmental releases of each stage of the life cycle of the product, process or activity. 3) Impact assessment: Data resulting from the inventory are translated into their corresponding environmental impacts for various impact categories. 4) Improvement assessment to identify and evaluate different options to reduce the environmental impact of the system based upon the study. In this study only the first two steps of the LCA have been carried out for the anode fabrication and the fuel cell system production, followed by discussions on environmental burdens based upon inventory data. A complete life cycle of a fuel cell system consists of raw materials extraction, manufacture and assembly of the stack and balance of plant, installation, operation and eventual decommissioning (13, 14). This study is only focused on the stage of anode production and fuel cell system fabrication, which includes raw materials production and manufacture of the stack and the balance of plant. The primary data on material and energy inputs includes information on the emissions associated with materials production are largely included. Data for the inventory was gathered from variety of sources (12-14). Where data was unavailable, assumptions were made and are described in the next section.

## Key Assumptions

The basic assumptions used in the analysis are as follows:

• The functional unit for the analysis is a  $100 \text{ cm}^2$  anode-supported single cell with 0.5 mm thick anode substrate in the anode production analysis, and a PEN (Positive electrode-Electrolyte-Negative electrode) structure with 0.08 mm thick anode substrate in the fuel cell system analysis. The overall porosity of the anode substrate is 30%.

• The material inputs are calculated under zero process losses, assuming 100% utilization and no rejection of faulty products. Neither recycling nor reuse was taken into consideration.

• The energy and the materials consumed in manufacturing the equipments used in the fabrication processes are not considered in this study. The land use associated with the production of the anode substrate and the fuel cell system is also not considered.

• Energy required for ventilation associated with the anode production is considered, to fully evaluate the difference between solvent-based tape casting and water-based tape casting.

• The emissions and the energy required associated with the transportation of materials to the anode/fuel cell system manufacturing infrastructure are not.

• The energy inputs is assumed to be 100% electricity when calculating the energy related emissions.

• To compare the emissions associated with the source of the electricity production, two sets of emission factors are utilized.

• Only key air pollutant emissions are considered and evaluated. Other than the air emissions, waterborne effluents, solid waste and other environmental releases are not considered in this study.

• PEN structures, geometries and the power density of unit cell to be identical.

## Ceramic Anode Manufacturing

Yttrium-doped strontium titanate based anode substrates (SYT-YSZ) were manufactured via solvent-based tape casting, non-toxic solvent-based tape casting and water-based tape casting processes.  $Sr_{0.86}Y_{0.08}TiO_3$  powders were prepared by a modified Pechini method. A detailed description of the synthesis is published elsewhere (15). The YSZ used in the study is commercial YSZ powder (TZ-8Y, Tosoh. Corp., Japan). The tape casting was performed on a laboratory scale tape casting equipment with a stationary doctor-blade and moving polyethylene film. The cast tapes were allowed to dry in the

drying chamber for several hours with controllable under-bed temperature and surface air flow temperature. After the solvent in the tapes was completely evaporated, the composite anode green tape was obtained. Two non-aqueous tape casting formulas for SYT-based ceramics anodes are developed using toluene/ethanol and ethanol as a solvent respectively, using poly vinyl butyral as a binder, poly propylene glycol and Santicizer 160 as plasticizers and Menhaden fish oil as a dispersant. The aqueous tape casting formula for SYT-based ceramic anodes is developed using de-ionized water as a solvent and poly vinyl alcohol as a binder, glycerol as plasticizer and Menhaden fish oil as a dispersant.

### **Results and Discussion**

#### Ceramic Anode Substrates

With optimized formulas, porous and homogeneous SYT ceramic-based anode substrates manufactured via solvent-based tape casting, non-toxic solvent-based tape casting and water-based tape casting processes were successfully fabricated and shown in Figure 1.

## SYT Ceramic Anode Manufacturing Energy Inputs and Emissions

Material inputs and energy inputs for each tape casting formula developed in this study are presented in Table I to Table III. The energy inputs consist of both energy consumed for the production of raw materials and energy consumed during the manufacturing process. The material input results are calculated based upon the anode geometry and porosity assumption combined with the tape casting formulas developed, while the energy inputs data are obtained from various sources (12-14). The results indicate that the anode fabricated via water-based tape casting has the highest material input per cell while it requires the lowest energy input for materials production as shown in Figure 2. Solvent-based formulas require more than twice the energy input for materials production compared to the water-based formula. The major difference in the energy input profile for materials production is the solvent used in the tape casting process, de-ionized water production requires much less energy compared to the production of organic solvent (toluene and/or ethanol).

Figure 3 presents the process energy inputs for the three tape casting formulas developed. For all formulas, it is noted that the energy inputs for manufacturing processes are dominated by the thermal processes such as sintering and drying. Since water has a lower drying rate than organic solvents, water-based tape casting requires a longer drying process and therefore higher energy consumption. With the consideration of health effect of toluene, extra ventilation process is required in the solvent-based tape casting process. The results indicate that in terms of manufacturing processes, water-based tape casting has the highest energy input demand. The energy input associated with the additional drying process for solvent-based tape casting outweighs the energy inputs in the extra ventilation process for solvent-based tape casting. It is also noted that the non-toxic solvent-based tape casting formula has the lowest energy input for manufacturing mainly because the formula removed toxic solvent (toluene) and the solvent (ethanol) used still have a relative higher volatile rate than water, as a result, energy consumed in the

ventilation process and drying process can be reduced. For all anode tape casting formulas developed in this study, the energy consumed in anode manufacturing (process energy) presented in Figure 3 are negligible when compared to the energy input for materials production that is shown in Figure 2.

Key air emissions associated with the SYT-based anode production are evaluated and shown in Figure 4 and Figure 5, which include carbon dioxide, carbon oxide, hydrocarbons, nitrate oxide, sulfur oxide and particulate matter. Total emissions from the SYT-based anode production are calculated using two sets of emission factors presented in Table IV. As shown in the figures, in terms of some key air emissions, anode substrates made by the water-based tape casting has the lowest total emissions under both CA and US emission factors. In terms of key air emissions, SYT-based anode substrates manufactured via the water-based tape casting method have the lowest total emissions. Using water as the solvent can significantly lower the total emissions and offset the additional emissions burden that is introduced by the extra drying required in water-based tape casting. Under both emission factors, anode fabricated using non-toxic solvent-based casting formula has the highest CO<sub>2</sub>, CO and PM emissions, while solvent-based tape casting formula has the highest SOx emissions. If the water-based tape casting is not an option for anode production under some circumstances, the solvent-based tape casting could be a better choice since it has relatively lower overall emissions than the non-toxic based tape casting. The total emissions results of each formula are demonstrated to be sensitive to the emission factors selected. Using CA emission factors, the total emissions calculated for each formula are much lower than using US emission factors. The results imply that comprehensive consideration is needed for locating a fuel cell manufacturing factory, since the emissions generated from a fuel cell stack at the manufacturing stage may be largely varied depend upon the electricity source. Moreover, due to the sensitivity of the emission factors, comparisons between these analyses and other fuel cell manufacturing life cycle analysis should be made with caution.

## PEN Structure Manufacturing Energy Inputs

Material input and energy inputs for the electrolyte and cathode manufacturing are presented in Table V and Table VI, respectively. In both the electrolyte and cathode manufacturing processes, the material inputs are calculated assuming zero process losses and 100% utilization. The energy inputs consist of the energy used for the production of raw materials and the energy consumed in the manufacturing processes. The electrolyte material inputs data are calculated based upon a 20  $\mu$ m thick YSZ electrolyte layer assumption and the electrolyte slurry formula developed in (10), while the process energy input data are obtained from the literature (12). The cathode material input data are evaluated based upon a 50  $\mu$ m thick LSM-YSZ composite cathode (50%/50% vol. %) with porosity of 0.4 and the cathode slurry formula utilized in (10). After the energy and material inputs of a PEN structure can be calculated. Table VII presents the energy and material inputs of an SYT-based anode-supported planar PEN structures using 3 different anode fabrication methods developed, denoted as Ceramic PEN route I, II and III.

Results presented in Table VII and Figure 6 indicate that in all PEN structures compared, energy consumed in fabrication processes are similar. The main reason is that all PEN structures assumed to utilize two co-sintering steps (electrolyte co-sintering and

cathode co-sintering) which have been demonstrated to be the predominant process in the manufacturing processes in terms of energy consumption. It can be also seen that the SYT-based anode-supported SOFC PEN structure using water-based tape casting for anode fabrication requires the lowest material and energy inputs for manufacturing and thus the lowest emissions generated associated with PEN fabrication.

## SOFC Systems Energy Inputs and Emissions

Environmental impacts of SYT-based anode-supported planar SOFC system manufacturing is also compared to a traditional Ni-YSZ based planar SOFC system (13) and presented in Figure 7 and Figure 8. The material and energy input data of interconnect presented in Table VIII is based upon a quantitative analysis of primary data obtained from the literature for a traditional planar SOFC (13). Table IX presents the main components of the balance of plant (BoP) assumed for the SYT-based anode-supported planar SOFC system and a traditional Ni-YSZ-based anode-supported planar SOFC system. In this study, it is also assumed that the BoP components listed in Table IX are relatively standard and based upon established technology (13). For the SYT-based anode-supported SOFC system, both de-sulphurizer and pre-reformer are excluded from the system to evaluate the merits of the ceramic based SOFC. Material and energy inputs of the BoP manufacturing for both systems are presented in Table X.

With the same PEN dimensions, the SYT-based anode-supported SOFC system has less energy input requirements for manufacturing and lower emissions than a traditional Ni-YSZ anode-supported SOFC system. This finding is mainly due to the significantly lower amount of energy that is required in the BoP manufacturing for an SYT-based SOFC system, assuming that using SYT-based ceramic anode could utilize natural gas fuel more directly and thus eliminate some of the fuel processing equipment. Interconnect material (Cr alloy) production has the highest percentage in both systems and the total energy input for materials production is not very sensitive to the energy consumed in PEN materials production.

For the SOFC systems investigated, the energy consumption and emissions are dominated by interconnects and the BoP manufacturing, while PEN manufacturing only accounts for 2% of the total energy inputs and emissions. The major energy consumption and emissions contributor in the SOFC system manufacturing has also been pinpointed to be the materials productions for PEN, interconnects and BoP, which made up to more than 97% of total energy consumption for both SOFC systems, compared to 3% energy consumed as process energy. In the future, it is recommended that optimization should be focused upon the interconnects and BoP manufacturing processes to reduce the energy consumption and the emissions.

## Conclusions

Yttrium-doped strontium titanate (SYT) based anode substrates were manufactured via solvent-based tape casting and water-based tape casting processes. Life cycle analyses of SYT-based anode manufacturing are carried out and the results of energy consumption and emissions indicate that the anode fabricated via water-based tape casting has the highest material input per cell while it requires the lowest energy input for materials

production. For all anode tape casting formulas developed in this study, the energy consumed in anode manufacturing (process energy) are negligible when compared to the energy input for materials production. The energy inputs for manufacturing processes are dominated by thermal processes. In terms of key air emissions, SYT-based anode substrates manufactured via the water-based tape casting method have the lowest total emissions.

With the same PEN dimensions, the SYT-based anode-supported planar SOFC system has less energy input requirements for manufacturing and lower emissions than a traditional Ni-YSZ anode-supported planar SOFC system. For the SOFC systems investigated, the energy consumption and emissions are dominated by interconnects and the BoP manufacturing, while PEN manufacturing only accounts for 2% of the total energy inputs and emissions.

## Acknowledgments

We gratefully acknowledge the support of Edison Materials Technology Center and Dr. Michael Martin. We also acknowledge the support of the Department of Defense (DoD) Fuel Cell Research Program, with our project managed by Mr. Franklin Holcomb.

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Material	Material input (kg/cell)	Energy inputs for materials production (MJ/cell)	Process	Energy inputs for manufacturing processes (MJ/cell)
SYT	0.082	1.144	Ball Milling	0.018
YSZ	0.095	1.322	Tape Casting	0.001
Graphite	0.074	7.310	Drying	0.034
Ethanol	0.061	3.136	Ventilation	0.018
Toluene	0.151	10.213	Sintering	0.169
PVB	0.030	1.509		
PEG	0.055	3.323		
Total	0.550	27.957		0.239

TABLE I. Energy and Material Inputs for Manufacturing Anode via Solvent-based Tape Casting.

**TABLE II.** Energy and Material Inputs for Manufacturing Anode via Non-toxic Solvent-based Tape Casting.

Material	Material input (kg/cell)	Energy inputs for materials production (MJ/cell)	Process	Energy inputs for manufacturing processes (MJ/cell)
SYT	0.082	1.144	Ball Milling	0.018
YSZ	0.095	1.322	Tape Casting	0.001
Graphite	0.040	3.936	Drying	0.034
Ethanol	0.362	18.592	Sintering	0.169
PVB	0.074	3.703		
PEG	0.059	3.557		
Total	0.713	32.253		0.222

TABLE III. Energy and Material Inputs for Manufacturing Anode via Water-based Tape Casting.

	0,	1	5	1 0
Material	Material input (kg/cell)	Energy inputs for materials production (MJ/cell)	Process	Energy inputs for manufacturing processes (MJ/cell)
SYT	0.082	1.144	Ball Milling	0.018
YSZ	0.095	1.322	Tape Casting	0.001
Graphite	0.040	3.936	Drying	0.068
$H_2O$	0.494	0.010	Sintering	0.169
PVA	0.024	1.176		
PEG	0.033	1.952		
Total	0.769	9.54		0.255

TABLE IV.	Emission	Factors	for Key 1	Air	Emissions.

TABLE IV. Emission Factors for Key An Emissions.							
Electric power generation emission factors (g/MJ)							
	PM 2.5*	PM 10*	PM*	CO*	CO2**	Sox**	NOx**
California (CA)	0.0033	0.0035	0.0069	0.0333	91.2224	0.0668	0.0781
U.S. Average (US) 0.0353 0.0429 0.0782 0.0439 167.4510 0.6627 (				0.2444			
* Data sources: http://www.epa.gov/air/emissions/index.htm							
http://www.eia.doe.gov/cneaf/electricity/epa/epates.html							
http://www.eia.doe.gov/cneaf/electricity/epa/epa_sprdshts.html							
** Data source: http:	//oaspub.epa	gov/powpro	o/ept_pack.c	harts			

	RAW material	kg/cell	Energy input for materials production (MJ/kg)	Energy input for materials production (MJ/cell)	Total energy input for materials production (MJ/cell)	
	YSZ	0.00118	13.8844	0.0164		
	Toluene	0.00236	67.6042	0.1595		
	Ethanol	0.00236	51.3115	0.1211	0.3295	
	PVB	0.000295	50	0.0148		
Electrolyte Fabrication	PEG	0.000295	60	0.0177		
radication	Processes		Energy input for manufacturing (MJ/cell)		Total energy input for manufacturing (MJ/cell)	
	Ball milling		0.0190			
	Pressurized spraying		0.0004			
	Drying		0.0342		0.2281	
	Vent	ilating	0.0024			
	Co-Si	ntering	0.1720			

<b>TABLE V.</b> Energy and Material Inputs for Manufacturing the Electrolyte.
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TABLE VI. Energy and Material Inputs for Manufacturing the Cathode.

	RAW material	kg/cell	Energy input for materials production (MJ/kg)	Energy input for materials production (MJ/cell)	Total energy input for materials production (MJ/cell)	
	YSZ	0.000885	13.8844	0.0123		
	LSM	0.00099	20.9797	0.0208	0.0799	
Cathode	Binder	0.0009375	50.0000	0.0469		
Fabrication	Processes		Energy input for manufacturing (MJ/cell)		Total energy input for manufacturing (MJ/cell)	
	Mixir	ng (ink)	0.0	028		
	Screen	printing	0.0026		0.2116	
	Dr	ying	0.0342			
	Co-Si	intering	0.1720			

			Energy input for materials production	Energy input for manufacturing processes	Total en inpu	0.
		Process	(MJ/cell)	(MJ/cell)	(MJ/cell)	%
DEM	Anode	Solvent-based tape casting	4.473	0.239	4.712	84.7%
PEN Bauta I	Electrolyte	Spraying	0.329	0.228	0.558	10.0%
Route I	Cathode	Screen printing	0.080	0.212	0.292	5.2%
		PEN Total	4.883	0.679	5.561	
	Anode	Non toxic solvent- based tape casting	5.160	0.222	5.382	86.4%
PEN Desete H	Electrolyte	Spraying	0.329	0.228	0.558	8.9%
Route II	Cathode	Screen printing	0.080	0.212	0.292	4.7%
		PEN Total	5.570	0.662	6.232	
PEN	Anode	Water-based tape casting	1.498	0.255	1.753	67.4%
Route	Electrolyte	Spraying	0.329	0.228	0.558	21.4%
III	Cathode	Screen printing	0.080	0.212	0.292	11.2%
		PEN Total	1.907	0.695	2.602	

## TABLE VII. Energy and Material Inputs for Manufacturing the PEN Structures.

#### TABLE VIII. Material and Energy Inputs for Interconnect.

Interconnect	Material input (kg/cell)	Energy input for materials production (MJ/cell)	Energy input for manufacturing process (MJ/cell)	Total energy input (MJ/cell)
Cr alloy	0.2682	70.8648	0.0086	70.8734

**TABLE IX.** Main Components of the BoP for Ni-YSZ-Based and SYT-based SOFC Systems.

Component	Ni-YSZ based SOFC system	SYT-based SOFC system
Casing		
Air and fuel supply systems	$\checkmark$	
De-sulphurizer		
Pre-reformer	$\checkmark$	
Heat exchanger	$\checkmark$	
Power conditioning system	$\checkmark$	$\checkmark$
Conventional gas heating unit	$\checkmark$	$\checkmark$

## TABLE X. Material and Energy inputs for Manufacturing the BoP for both SOFC Systems.

SOFC system	Energy input for materials production (MJ/cell)	Energy input for manufacturing process (MJ/cell)	Total energy input (MJ/cell)
Ni-YSZ based	49.426	2.246	51.672
SYT-based	43.948	1.972	45.920

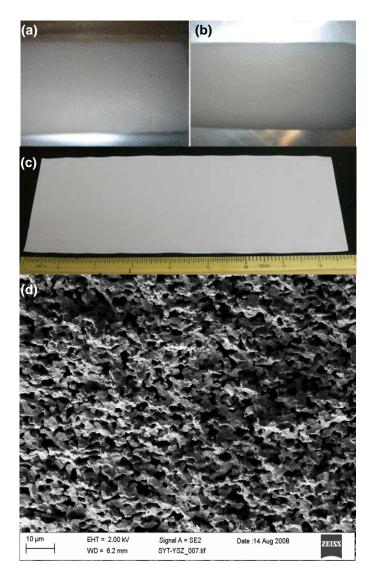


Figure 1. (a) SYT-based anode tape made by solvent-based tape casting; (b) SYT-based anode tape made by non-toxic solvent-based tape casting; (c) SYT-based anode tape made by water-based tape casting; (d) Microstructure of an SYT-YSZ composite anode substrate after sintering to 1400°C in air for 4 hours.

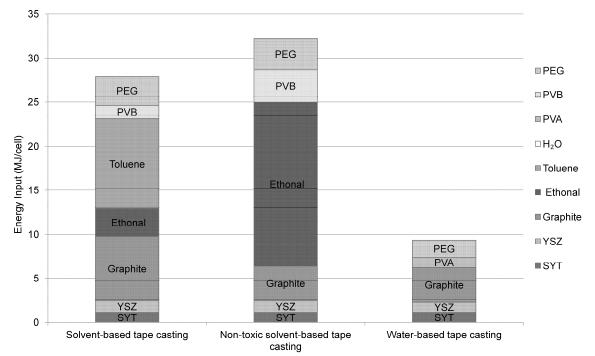
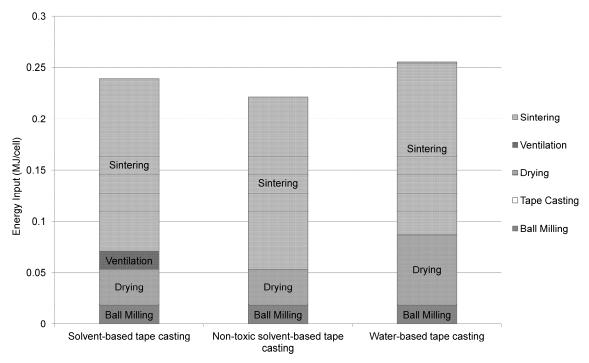
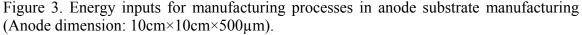


Figure 2. Energy inputs for materials production in the anode substrate manufacturing (Anode dimension:  $10 \text{cm} \times 10 \text{cm} \times 500 \mu \text{m}$ ).





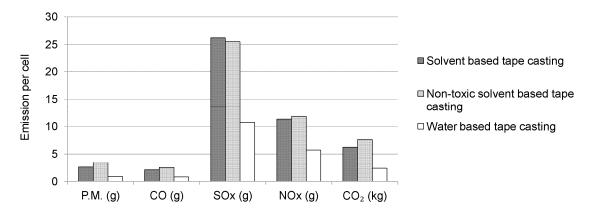


Figure 4. Key air emissions from anode production using U.S. average emission factors.

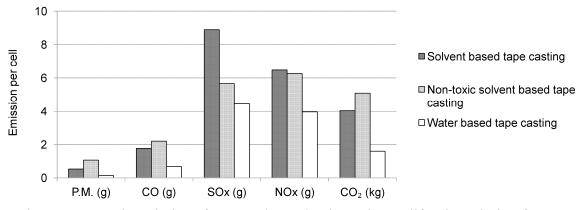


Figure 5. Key air emissions from anode production using California emission factors.

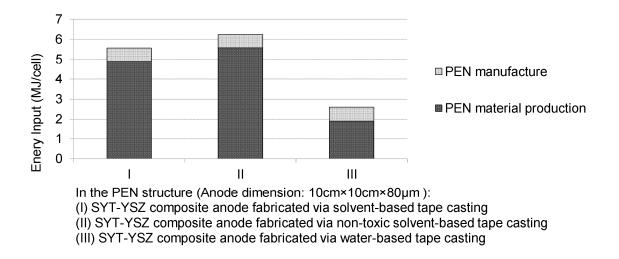


Figure 6. Energy inputs for PEN structure manufacturing.

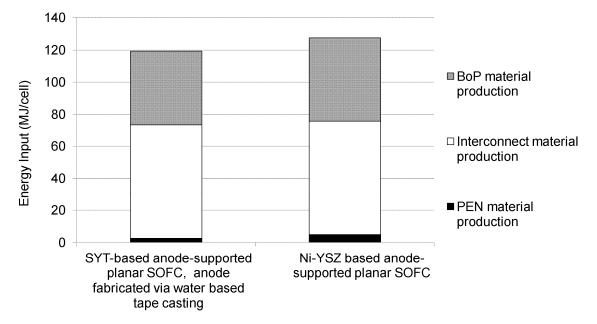


Figure 7. Energy inputs for materials production in each component.

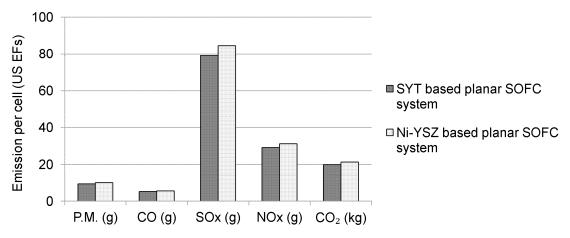


Figure 8. Emissions associated with manufacturing of two SOFC systems using U.S. average emission factors.