

PERFORMANCE ANALYSIS OF A PROTON EXCHANGE MEMBRANE

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ABSTRACT

Proton Exchange Membrane Fuel Cells (PEMFC) are emerging on the market as a promising technology for powering cars and combined heat and power applications in buildings. The thermodynamic performance of these cells is limited at the moment by membrane technology and power use of the peripheral equipment. The interaction of the fuel cell with its peripheral equipment is often not optimized at the moment. Therefore, in this paper a thermodynamic model for Proton Exchange Membrane (PEM) Fuel Cells is developed under ASPEN PLUS. This model has to be sufficiently accurate while consuming little computational time. This condition is necessary to integrate the fuel cell model with its peripheral equipment.

The model is based on a 3 control volume approach including the complete reaction modelling. The PEM model not only calculates the cell voltage, it also includes a detailed heat transfer model that predicts outlet temperatures and cooling rates. The PEM model can predict the steady-state performance of a stand-alone fuel cell stack. The model is validated by comparing simulated working characteristics with experimental data out of the open literature and with the measured performance of the bench-scale PEM fuel cells at the VITO.

Finally the performance and temperature distribution is calculated and compared to know measurement data and other modelling results. The model predictions for temperature are slightly differing from the measurements, but the new model is more accurate than previously formulated models.

INTRODUCTION

A proton exchange membrane (PEM) fuel cell is an electrochemical device where the energy of a chemical reaction is converted directly into electricity, by combining hydrogen fuel with oxygen from air [1]. Water and heat are the only byproducts if pure hydrogen is used as fuel source. A fuel cell consist of a negatively charged anode, a positively

charged cathode and an electrolyte which transports protons from the anode to the cathode but blocks electrons, forcing them to move through the external load, as shown in Fig. 1.

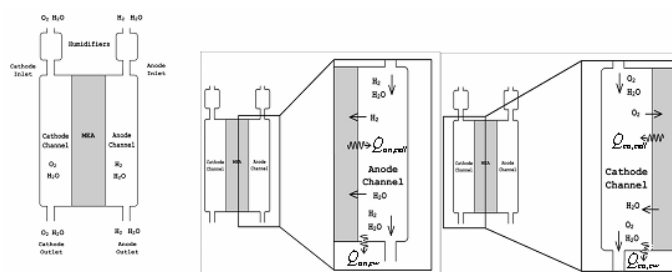


Figure 1 PEM Fuel Cell and control volumes

In addition to the common advantages of fuel cell systems such as high efficiency and clean emissions, PEMFCs are capable of high power density and they operate at a relatively low temperature. The low operating temperature yields that PEMFCs can be housed safely on various carriers, have a quick start-up time compared to other types of fuel cells, can be easily maintained and have higher reliability and longer expected lifetime. On the other hand, this low operating temperature generally leads to the two-phase change of water liquid-vapor inside the PEMFC [2].

Water plays a critical role in PEMFC operations. In order to efficiently conduct the hydrogen protons and prevent the localized hot spots, the membrane needs to be appropriately hydrated [3]. To achieve sufficient hydration, water is introduced into the fuel cell by passing the reactants through humidifiers before entering the cell [4,5]. Suitable heat management is also one of the fundamental requirements to maintain the fuel cell performance [4]. Overheating the fuel cell might lead to severe dehydration of the membrane and results in a larger internal resistance and thus fuel cell performance loss. If the temperature is too low, the cathode gas channel will be flooded, the liquid water blocking the

oxygen transport path. This would mean that the reaction at the cathode is starved of oxygen and that the fuel cell performance will drop. This implies that the fuel cell temperature must be controlled by means of a coolant.

Over the years, PEMFCs have been modelled at various levels of complexity with different focuses. The present work aims at developing an accurate and easy-to-use model, based upon well-known electrochemical and thermodynamic equations. The model is developed using the control volume approach, dividing the fuel cell into three control volumes: the anode channel, the cathode channel and the fuel cell body. When a fuel cell stack is considered, the anode channel, cathode channel and body control volumes will be the summation of corresponding volumes of each single cell. The model is created and simulated under the Aspen Custom Modeler environment. The model is validated with a range of experimental results, released by the VITO (Vlaamse Instelling voor Technologisch Onderzoek). This work presents a comprehensive one-dimensional system-level steady-state model for PEMFCs that can be used for design and control studies.

NOMENCLATURE

A	effective area of the membrane (m ²)
c	molar concentration (mol/m ³)
E	energy (W)
E _{nernst}	thermodynamic potential (V)
F	Faraday's constant (C/mol)
\dot{m}	species mass flow rate in the channels (kg/s)
G	Gibbs free energy (J/kg)
h	total enthalpy (J/kg)
αA	effective convection heat transfer coefficient (W/K)
I	load current (A)
M	species mole mass (kg/kmol)
MEA	membrane electrode assembly
\dot{n}	molar flow rate (mol/s)
N	number of cells in the stack
p	species partial pressure (bar)
P	power (W)
EM	proton exchange membrane
\dot{Q}	heat (W)
R	universal gas constant (J/kmol/K)
T	temperature (K)
V	voltage (V)
\dot{W}	power output (W)
η	overpotential (V)
ξ	empirical coefficient of overpotential
ϕ	relative humidity
χ	empirical coefficient of overpotential

Subscripts

an	anode
act	activation
ca	cathode
cell	fuel cell body

cw	cooling water
in	inlet
liq	liquid
ohmic	ohmic
out	outlet
surr	surroundings
vap	vapour
w	water

MODEL DEVELOPMENT

Modelling assumptions

In order to quantify the complicated mass and energy interactions within the fuel cell, the control volume approach is used and the following three control volumes are defined: the anode channel, the cathode channel and the fuel cell body, which includes the membrane electrode assembly (MEA) and all other metal structures involved in the fuel cell [6].

Throughout this paper, the following assumptions are made:

1. The electrochemical reaction occurs in the MEA, and generates heat to the fuel cell body control volume. The generated water is in the liquid phase.
2. Hydrogen and oxygen diffuse to the membrane in the form of vapor. Water leaves the membrane partially as vapor, partially as liquid. The vapor fraction is determined by the phase equilibrium of the water, in the presence of oxygen and hydrogen in the membrane.
3. Ideal gas law is employed for gaseous species.
4. The pressure drop over the gas channels can be neglected. The pressure in the membrane is the average of the pressure in the gas channels.
5. Liquid water exists at the surface of the channels in the form of small droplets, whose volume is negligible. The liquid water droplets are forced out of the channels through regular purging.
6. Heat transfer by conduction in the gas phase is negligible.
7. The fuel cell body temperature is considered uniform due to its high thermal conductivity.
8. The temperature within the anode channel and cathode channel control volume is uniform.
9. The model will be used for steady state operations.
10. The membrane is sufficiently hydrated to perform a good proton transport.
11. Vapour transport across the MEA includes electro-osmotic drag and back diffusion. The effect of hydraulic permeation is neglected.
12. The cooling water is flowing behind a separator and heat transfer is considered among the anode gas, the cathode gas, the MEA and the cooling water [9].

Anode channel

To obtain the anode gas consumption and the temperature in the anode channel the mass and energy conservation principles is used.

Mass conservation

$$\dot{m}_{H_2,out} = \dot{m}_{H_2,in} - M_{H_2} \frac{NI}{2F} \quad (1)$$

$$\dot{m}_{H_2O,an,out} = \dot{m}_{H_2O,an,in} - \dot{m}_{H_2O,MEA} \quad (2)$$

Eq. (1) defines the mass balance for hydrogen and indicates that the amount of hydrogen used is dependent on the load current. The amount of water introduced into the anode gas channel through humidification does not remain the same throughout the channel because of water transport across the membrane which includes the combined effect of electro-osmotic drag and back diffusion, Eq. (2). Water transport across the MEA is modelled as in [2].

Energy conservation

$$\dot{Q}_{an} = (\alpha A)_{an,cell} (T_{cell} - T_{an}) + (\alpha A)_{an,cw} (T_{cw} - T_{an}) \quad (3)$$

$$\begin{aligned} \dot{Q}_{an} = & \dot{m}_{H_2,out} h_{H_2,out} + \dot{m}_{H_2O,an,out} h_{H_2O,an,out} + M_{H_2} \frac{NI}{2F} h_{H_2,an} \\ & + \dot{m}_{H_2O,MEA} h_{H_2O,an} - \dot{m}_{H_2,in} h_{H_2,in} - \dot{m}_{H_2O,an,in} h_{H_2O,in} \end{aligned} \quad (4)$$

By neglecting gravitational and kinetic effects, Eq. (4) states the first law of thermodynamics for the anode control volume. The parameter h_i represents the total enthalpy, including the enthalpy of formation. This means the energy balance also accounts for the condensation/evaporation of the water and the heat released or consumed by it. Being able to work with the total enthalpy and to have access to an enormous database of thermodynamic data is one of the great advantages of working in the Aspen Custom Modeler environment. The terms $(\alpha A)_{an,cell}$ and $(\alpha A)_{an,cw}$ represent the convective heat transfer coefficients between the anode channel control volume and respectively the fuel cell body and the cooling water. It is worth noting that the detailed heat transfer mechanism is very complicated and is beyond the scope of the current study. For this model, the thickness of the channel is taken to be of moderate scale and thus the overall effect can be equivalently represented by the convection coefficient, which is consistent with previous relevant studies [7]. Similar representations are used for the heat transfer between the fuel cell body, the cathode channel control volume, the coolant water and the surroundings. The gas flow in the anode channel could be either pure hydrogen or a gas mixture, consisting of the reforming reaction products hydrogen and carbon dioxide. In this pure hydrogen is used. However, the case of reforming products can be similarly derived by additionally considering the mass conservation of carbon dioxide and its contribution to the energy balance.

Cathode channel

Similar to those obtained for the anode channel control volume, the governing equations of the cathode channel control volume can be derived by using mass and energy conservation principles.

Mass conservation

$$\dot{m}_{O_2,out} = \dot{m}_{O_2,in} - M_{O_2} \frac{NI}{4F} \quad (5)$$

$$\dot{m}_{H_2O,ca,out} = \dot{m}_{H_2O,ca,in} + M_{H_2O} \frac{NI}{2F} + \dot{m}_{H_2O,MEA} \quad (6)$$

The consumed oxygen diffuses through the cathode to the fuel cell body and reacts with the hydrogen, migrated through the membrane as protons, Eq. (5). The reaction water then diffuses back into the cathode channel, partially as liquid, partially as vapour, Eq (6). The water transport across the membrane is also considered.

Energy conservation

$$\dot{Q}_{ca} = (\alpha A)_{ca,cell} (T_{cell} - T_{ca}) + (\alpha A)_{ca,cw} (T_{cw} - T_{ca}) \quad (7)$$

$$\begin{aligned} \dot{Q}_{ca} = & \dot{m}_{O_2,out} h_{O_2,out} + \dot{m}_{H_2O,ca,out} h_{H_2O,ca,out} + M_{O_2} \frac{NI}{4F} h_{O_2,ca} - \dot{m}_{O_2,in} h_{O_2,in} \\ & - \dot{m}_{H_2O,ca,in} h_{H_2O,ca,in} - M_{H_2O} \frac{NI}{2F} h_{H_2O,cell} - \dot{m}_{H_2O,MEA} h_{H_2O,cell} \end{aligned} \quad (8)$$

Eq. (8) states the first law of thermodynamics for the cathode channel control volume. The gas flow in the cathode channel could be either pure oxygen or air. In this study the use of pure oxygen is assumed. However, the case of air can be similarly derived by additionally considering the mass conservation of nitrogen and its contribution to the energy balance.

Fuel cell body

Energy conservation

One of the most important parameters for a satisfactory performance of a PEM fuel cell is the fuel cell body temperature. This temperature can be obtained, using the energy conservation principle on the fuel cell body control volume.

$$\begin{aligned} \dot{Q}_{cell} = & (\alpha A)_{an,cell} (T_{an} - T_{cell}) + (\alpha A)_{ca,cell} (T_{ca} - T_{cell}) \\ & + (\alpha A)_{surr} (T_{surr} - T_{cell}) + (\alpha A)_{cell,cw} (T_{cw} - T_{cell}) \end{aligned} \quad (9)$$

$$\dot{W}_{cell} = NV_{cell} I \quad (10)$$

$$\begin{aligned} \dot{Q}_{cell} - \dot{W}_{cell} = & M_{H_2O,vap} \frac{NI}{2F} h_{H_2O,cell,vap} + M_{H_2O,liq} \frac{NI}{2F} h_{H_2O,cell,liq} \\ & - M_{H_2} \frac{NI}{2F} h_{H_2,an} - M_{O_2} \frac{NI}{4F} h_{O_2,ca} + \dot{m}_{H_2O,MEA} (h_{H_2O,cell} - h_{H_2O,an}) \end{aligned} \quad (11)$$

Eq. (11) represents the power and heat generated by the electrochemical reaction and the heat consumed by the evaporation of liquid water. The water is in equilibrium with the hydrogen and oxygen in the membrane. A fraction will be transported from the cathode catalyst layer to the cathode channel in the vapour phase, the rest in the liquid phase. Again water transport across the membrane is considered. Because the term h_i represents the total enthalpy, as mentioned before, it includes the enthalpy of formation and the enthalpy of evaporation, which are the negative of the higher heating value (HHV) of the chemical reaction of hydrogen and oxygen, combining to water. This HHV is the driving energy source for the fuel cell performance. Part of this energy is converted into electrical power, Eq. (10), while the rest is converted into heat, Eq. (9). This thermal energy is transported, by means of convective heat transfer, to the various components of the fuel cell system including the

anode channel, the cathode channel, the cooling water and the surrounding air respectively.

Cooling water

In order to control the fuel cell temperature water cooling through micro cooling channels was incorporated into the PEMFC model. Since the liquid cooling water is incompressible, the mass conservation is trivial. Convective heat transfer between the cooling water and the anode gas, cathode gas and the fuel cell body is considered. The energy balance for the cooling water states

$$\begin{aligned} (\alpha A)_{an,cw}(T_{an} - T_{cw}) + (\alpha A)_{ca,cw}(T_{ca} - T_{cw}) + \\ (\alpha A)_{cell,cw}(T_{cell} - T_{cw}) = m_{cw}(h_{cw,out} - h_{cw,in}) \end{aligned} \quad (12)$$

Electrochemical model

Fuel cells are energy conversion devices that convert chemical energy into electrical energy through electrochemical reactions. Typically, the fuel cell output voltage is the summation of three effects, the Nernst potential, the cathode and anode activation overvoltage and the ohmic overvoltage [8].

$$V_{cell} = E_{nernst} + \eta_{act} + \eta_{ohmic} \quad (13)$$

The Nernst potential equals the Gibb's free energy and is calculated by the following equation

$$E_{nernst} = -\frac{\Delta G^0}{2F} - \frac{RT_{cell}}{2F} \left[\ln(p_{H_2}) + 0.5 \ln(p_{O_2}) \right] \quad (14)$$

In this equation, the channel hydrogen/oxygen partial pressure instead of the effective partial pressure at the electrode, as considered in [8] is used, in order to simplify the PEMFC system modelling. The use of Aspen Custom Modeler® makes it possible to calculate the Gibbs-free energy out of the enthalpy and entropy of the gases and the water. This method gives a good accuracy. According to assumption (1) the potential based on the Gibbs-free energy will be closely to 1,23 V.

The model that is used to determine the overvoltages is based upon the linear regression of experimental data under various cell temperatures and load currents [8]. This approach has been employed in various system-level fuel cell models, leading to accurate values in correlation with experimental results [6,7]. The total activation overvoltage can be written as

$$\eta_{act} = \xi_1 + \xi_2 T_{cell} + \xi_3 T_{cell} \ln(c_{O_2}) + \xi_4 T_{cell} \ln(I) \quad (15)$$

The parametric coefficients ξ_1 to ξ_4 in the activation overvoltage term are based on theoretical equations from kinetic, thermodynamic and electrochemistry fundamentals. The parametric coefficients ξ_5 to ξ_7 in the ohmic overvoltage term are purely empirical based on temperature and current experimental data [6]. The ohmic overvoltage is caused by electron flow resistance, proton flow resistance through the membrane and other contact resistances.

$$\eta_{ohmic} = -I(\xi_5 + \xi_6 T_{cell} + \xi_7 I) \quad (16)$$

It should be noted that the equations expressing the overvoltages apply only to the steady-state case. Values for ξ_1 to ξ_7 can be found in literature [6].

MODEL VALIDATION

The VITO provided experimental results of a transient analysis of a PEMFC. However, the data contain different quasi steady-state set points that are suitable for validation.

The VITO PEMFC

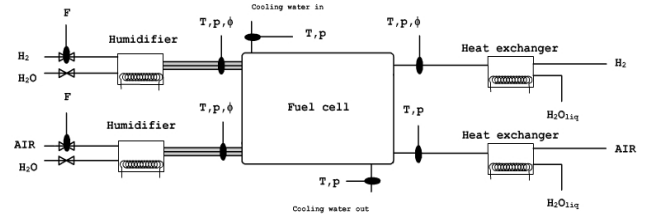


Figure 2 : VITO fuel cell configuration

The fuel cell created at the VITO is a PEM fuel cell of 2 kW. The stack contains 50 cells that consist of an anode channel, a cathode channel and the Nafion® membrane between both channels. All cells are separated from each other with water-cooled flow plates. The stack is fed with hydrogen and air at approximately 60 °C. The cooling water is also added at the same temperature. Humidifiers add vapor to the inlet gas streams to control the water management of the membrane. The configuration of the fuel cell is shown in Figure 2.

Adaptations to the model

To match the model with the new experimental data the cell specific parameters need to be adjusted. New values for the parameters ξ_i have to be calculated, based on the experimental polarization curve provided by the VITO. This is done because the fuel cells at the VITO have a different configuration and consist of different materials as the Ballard cells, used in [7] to determine the parameters ξ_i . As the ohmic and activation losses are not known separately, combining equations (15) and (16) to a new correlation and thus expressing the overall voltage loss η_{loss} , gives:

$$\eta_{loss} = \chi_1 + \chi_2 I + \chi_3 I^2 + \chi_4 \ln(I) \quad (17)$$

Because the cell temperature was not measured, the temperature was integrated into the χ parameters. The same was done for the concentration of O_2 at the cathode. The χ parameters are then calculated through regression analysis on the measured values. This method provided for a good electrochemical model. Figure 3 shows the good agreement of

the presented electrochemical model with the experimental results.

With this electrochemical model, the model that will return the control volume temperatures is further developed. The next step is to estimate the heat transfer coefficients in order to fit the experimental results. They should be calculated for each individual load conditions, but in this first attempt, only mean heat transfer coefficients will be considered, averaged over a whole range of working conditions. These coefficients can be seen as parameters which depend on the geometry of the fuel cell and the flow rates in the channels. The coefficients used in the model for the PEMFC of the VITO are given in Table 1.

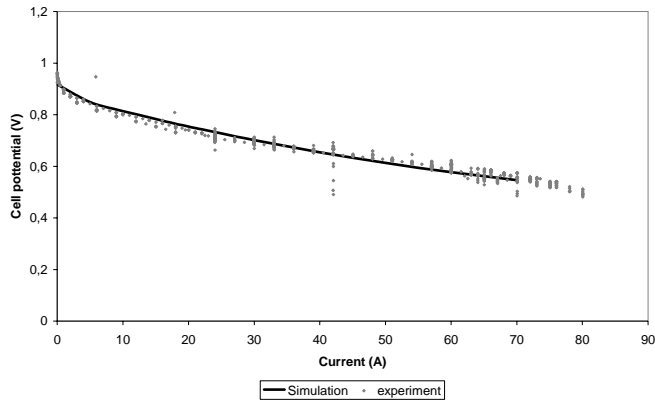


Figure 3 : -Polarization curve of the VITO fuel cell, simulation and experimental results

Simulation results

The input values of five set points, measured by the VITO, are listed in Table 2. The load current is the independent variable. The other input parameters remain close

to constant. With the adjustments made to the model, as described in the previous section, the performance of the fuel cell was simulated as a function of the load current. The comparison between the simulated and the experimental values is given in Table 3. The only parameter that was not registered at the VITO is the fuel cell body temperature.

Because the electrochemical model was created through linear regression, based on the electrochemical model presented in [7], an almost perfect match of cell voltages was obtained. Looking at the temperatures of the different control volumes, an excellent agreement between simulation and experiment is found, except for the anode temperature. The maximum relative error equals only 1%, i.e. for the cathode absolute temperature of set point 2.

The only anomaly that occurs is the anode channel temperature. The model overestimates this value. It can be noticed that for all the set points the outlet temperature of the anode channel is smaller than the inlet temperature. By some means, the anode channel is cooled. A possible explanation is the purging of the channels every 30 seconds. However, all models in the literature suffer from bad predictions of the anode gas temperature. This highlights the need to further examine the physical phenomena occurring at the anode.

$hA_{an,cell}$ (W/K)	0.02
$hA_{an,cw}$ (W/K)	10.0
$hA_{ca,cell}$ (W/K)	12.0
$hA_{ca,cw}$ (W/K)	80.0
$hA_{cell,cw}$ (W/K)	110.0
hA_{surr} (W/K)	10.0

Table 1 : Convection coefficients

Anode						
p_{an}	1,10	1,10	1,10	1,10	1,10	Bar
$T_{an,in}$	62,1	66,3	65,8	63,9	67,4	°C
Ψ_{H_2}	1,2	1,2	1,2	1,2	1,2	
ϕ_{an}	76,18	64,73	72,58	74,68	59,72	%
Cathode						
p_{ca}	1,10	1,28	1,21	1,09	1,15	Bar
$T_{ca,in}$	60,0	64,1	64,7	64,2	63,3	°C
Ψ_{O_2}	2,09	2,09	2,09	2,09	2,09	
ϕ_{ca}	98,27	88,89	94,91	99,00	98,04	%
Surroundings						
p_{ref}	1	1	1	1	1	Atm
T_{surr}	18	18	18	18	18	°C
Cooling water						
F_{cw}	0,10	0,13	0,13	0,08	0,11	mol/s
$T_{cw,in}$	65,5	65,8	65,8	65,3	65,4	°C
Number of Cells						
I	39,04	75,05	64,05	33,02	51,04	A

Table 2 : Input data for the VITO fuel cell

Set point 1	Exp. value	Calc. value
V_{cell} (V)	0.66	0.66
V_{stack} (V)	33.00	33.03
P (kW)	1.29	1.29
$T_{an,out}$ (°C)	58.3	67.3
T_{cell} (°C)		73.5
$T_{ca,out}$ (°C)	69.5	69.5
$T_{cw,out}$ (°C)	68.0	67.5
Set point 2	Exp. value	Calc. value
V_{cell} (V)	0.54	0.54
V_{stack} (V)	26.84	26.79
P (kW)	2.01	2.01
$T_{an,out}$ (°C)	65.3	69.7
T_{cell} (°C)		86.8
$T_{ca,out}$ (°C)	71.8	74.7
$T_{cw,out}$ (°C)	70.8	69.9
Set point 3	Exp. value	Calc. value
V_{cell} (V)	0.57	0.57
V_{stack} (V)	28.38	28.35
P (kW)	1.82	1.82
$T_{an,out}$ (°C)	65.5	69.0
T_{cell} (°C)		82.4
$T_{ca,out}$ (°C)	71.3	74.1
$T_{cw,out}$ (°C)	70.2	69.2
Set point 4	Exp. value	Calc. value
V_{cell} (V)	0.69	0.69
V_{stack} (V)	34.48	34.35
P (kW)	1.14	1.13
$T_{an,out}$ (°C)	60.6	67.3
T_{cell} (°C)		71.9
$T_{ca,out}$ (°C)	69.9	69.9
$T_{cw,out}$ (°C)	68.1	67.4
Set point 5	Exp. value	Calc. value
V_{cell} (V)	0.62	0.61
V_{stack} (V)	31.10	30.60
P (kW)	1.59	1.56
$T_{an,out}$ (°C)	65.8	68.3
T_{cell} (°C)		77.6
$T_{ca,out}$ (°C)	69.9	72.1
$T_{cw,out}$ (°C)	68.7	68.3

Table 3 : Simulated results of 5 set points

CONCLUSION

In this paper an easy-to-use steady state PEMFC model was developed, based on the control volume approach, as consistent as possible with the on-going physical phenomena inside the fuel cell. The considered consistency was translated in the formulated assumptions, which constitute the solid basis for a realistic and useful behaviour of the model. The model was validated using experimental results from a 2kW PEMFC stack provided by the VITO. The new model shows similar deviations from reality as the existing models in literature, e.g. considering the anode channel temperature, highlighting the importance of further investigation.

The validated model can be used to run simulations, predicting fuel cell performance with an emphasis on the thermal behaviour and the heat released by the fuel cell. The present model can also be used for fuel cell design and control purposes.

ACKNOWLEDGEMENTS

The authors would like to thank the VITO for their willingness to offer the highly important and both rare experimental data.

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