Review of necessary thermophysical properties and their sensivities with temperature and electrolyte mass fractions for alkaline water electrolysis Multiphysics modelling

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Abstract

This article presents the transport input properties necessary for alkaline water electrolyzer multiphysic modeling (CFD). This article provides experimental data and the needed correlations of the parameter (electrical conductivity, density, viscosity, heat capacity, heat and mass transfer diffusion coefficients used in multiphysical modeling depending on temperature and mass fraction for two classical alkaline electrolytes (KOH, NaOH) over a wide range of temperature and mass fraction. Thus, the different involved electrodes boundary layers can be calculated with precision. First of all, 6 usual inputs liquid electrolyte parameters (density, specific heat, electric and thermal conductivity, viscosity, mass diffusivity) are given as a function of temperature and electrolyte mass fraction (for KOH and NaOH). Different interpolation models from various authors and also original are compared to experimental rough data. The goal of this article is to give to the modeler the needed correlations to allow the simulation of the alkaline water electrolysis.

Keywords

Alkalin electrolysis, multi physic modeling, electrolyte, thermophysical properties

1 Introduction

The challenge of the 21st century is to decrease the CO_2 emission in order to manage and decrease the global warming. The main solution of this problem is to produce energy from the renewables energy and if possible without the help of hydrocarbon and carbon molecules. Due to the fact that the renewables energies are intermittent in space and time the current production can not be integrated properly on the electrical network. From this statement, it has been deduced that the energy must be stored in small or medium scale smart grids. The national electrical network is not calculated to receive renewable production and it is not always possible (isolated locations, islands, mountains, full sea…). In these cases the storage of produced electricity is necessary.

he present study takes place in this context. Using hydrogen as an energy vectors could solve this problematic because its production only needs H_2O and electricity. Nevertheless, hydrogen production processes and electrolysers (alkaline, acid, thermochemical combined cycles or high temperature) leads to a hydrogen cost too expensive comparatively to fuel, gasoil, or electrical energy produced with nuclear or thermal plants. Presently electrolysis processes are not enough efficient or cheap and they must be optimized, improved and become cheaper. The alkaline water electrolysis is the oldest process, the more robust and the cheapest technology. It has been early theoretically and experimentally studied but most of the theoretical works are primary or secondary charge transfer modeling (charge transfer modeling with activation overpotentials considered). These works do not take into account the local temperature and hydroxide; also the electro-active species hydroxide ions (OH^-) are not calculated. Modern technologies allow us to investigate and improve this electrolysis by using CFD modeling, under ternary assumptions. It will allow us to access to current density distribution at electrodes for example. This modelling goal needs at least six liquid electrolyte parameters:

1/ electrical conductivity σ ($S\ m^{-1}$),

2/ density ρ(T,Y) ($kg\ m^{-3}$) ,

3/ viscosity μ(T,Y) ($kg\ m^{-1}\ s^{-1}$),

4/ specific heat Cp(T,Y) ($J\ kg^{-1}\ K^{-1}$),

5/ thermal conductivity λ(T,Y) ($W\ m^{-1}\ K^{-1}$),

6/ mass transfer diffusion coefficient D(T,Y) ($m^2\ s^{-1}$).

All of them might be modelled as dependent of the local electrolyte mass fraction Y (-) and temperature T (K). First step is to obtained all the input properties and their sensitivity with temperature and electrolyte mass fraction. The present work will give a complete and exhaustive review of those parameters with their temperature and concentration sensivities and will give easy and accurate correlations ready to program for modelers scientists. It exists few works that give one or two parameters but never the 6 parameters cited and their sensitivities. of them give them all. One particular case is our main reference, Zaytsev [1], which is a relatively complete collection of several data and their sensitivities, sometime obtained experimentally, sometimes calculated with dynamic molecular numerical simulations. But this reference, which is a general aqueous dissolved salts data handbook, is hardly accessible, expensive and contains a lot of unnecessary data for uninteresting salts for the alkaline water electrolysis process modelling. And also, Zaytsev [1] does not give all the needed data and fitting correlations for example for electrical conductivity σ ($S\ m^{-1}$) and mass transfer diffusion D ($m^2\ s^{-1}$) coefficients. Table 1 presents all the references that have been used to write the present article. Most of the reference data come from Zaytsev handbook [1]. For the electrical conductivity σ ($S\ m^{-1}$) and the density of KOH, the Gilliam work [3] has been used. See's work [2] also gives data and correlation but only for electrical conductivity σ ($S\ m^{-1}$). The electrical conductivity is widely available among the previous alkaline water electrolysis because this property is the main and only necessary one for simplest modelling (primary or secondary current density distribution for example). Klochko [4] has also given few points for the KOH and NaOH electrical conductivity σ ($S\ m^{-1}$) and viscosity μ ($kg\ m^{-1}\ s^{-1}$). Guo's work [5] provides correlations and experimental data for KOH density ρ ($kg\ m^{-3}$) and viscosity μ ($kg\ m^{-1}\ s^{-1}$). Laliberté's article [6] and Roux [7] furnish a method to determine from experimental data the KOH and NaOH density ρ ($kg\ m^{-3}$), viscosity μ ($kg\ m^{-1}\ s^{-1}$) and specific heat Cp ($J\ kg^{-1}\ K^{-1}$) correlations laws. Wang's [8] and Riedel's [9] work has been used to get correlation for KOH and NaOH thermal conductivity λ($W\ m^{-1}\ K^{-1}$). For the NaOH density ρ ($kg\ m^{-3}$), there are three mains article : Akerlof [10] and Olsson) [11] and Churikov[12]. Olsson also supplies correlations and data for NaOH viscosity μ ($kg\ m^{-1}\ s^{-1}$). The more difficult to find has been the diffusion coefficient D ($m^2\ s^{-1}$) and its temperature T and electrolyte mas fraction Y (-) for OH^- hydroxide anions at anode which is an essential data for ternary modelling of current density distribution. This is due of the small number of published works under this ternary modelling assumption. It was especially harder to find for NaOH instead of KOH. We have tried to give with the few data found a sensitivity correlation

Table 1-Summary of the references depending on their works

	Electrical Cond.								Density								Viscosity								Specific Heat								Thermal Cond.								Diffusivity Coef.							
	KOH				NaOH				KOH				NaOH				KOH				NaOH				KOH				NaOH				KOH				NaOH				KOH				NaOH			
	T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk		T		Yk	
	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C	D	C
Zaytsev [1]	X		X		X		X		X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X		X		X		X	
See [2]	X	X	X	X																																												
Gilliam [3]	X	X	X	X					X	X	X	X																																				
Klochko [4]	X		X		X		X										X		X		X		X																									
Guo [5]									X	X	X	X					X	X	X	X																												
Laliberté [6]										X		X		X		X		X		X		X		X		X		X				X																
Roux [7]																									X		X	X	X		X	X																
Wang [8]																																		X		X		X		X								
Riedel [9]																																		X	X	X		X	X	X								
Akerlof [10]													X	X	X	X																																
Olsson [11]													X	X	X	X					X	X	X	X																								
Churikov [12]									X	X	X	X	X	X	X	X																																
Le Bideau	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X

Nomenclature

Roman symbol

a	Heat diffusivity ($m^2\ s^{-1}$)
c	Species molar concentration (mol m^{-3})
Cp	Specific heat (J $kg^{-1}\ K^{-1}$)
D	Species diffusion coefficient ($m^2\ s^{-1}$)
g	acceleration constant
h	Heat convection coefficient (W $m^{-2}\ K^{-1}$)
I	Intensity (A)
j	Current density (A m^{-2})
k	Mass transfer coefficient (m s^{-1})
M	Molar mass (kg mol^{-1})
n	species quantity (mol)
J	Mass flux density (mol $m^{-2}\ s^{-1}$)
P	Power (W)
R	Electrical resistance (Ω)
r_m	atomic radius (Å)
S	Surface area (m^2)
T	temperature (°C)
t	time (s)
V	velocity (m s^{-1})
U	Electrical imposed potential (V)
U	Vector velocity
X	species molar fraction (-)
Y	species mass fraction (-)
x, y, z	Spatial coordinates (m)

Greek symbol

α	Wang's coefficient (-)
λ	heat conductivity (W $m^{-1}\ K^{-1}$)
ρ	density (kg m^{-3})
μ	Dynamic viscosity (kg $m^{-1}\ s^{-1}$)
η	Over potential (V)
σ	Electrical Conductivity (S m^{-1})
ν	Kinematic viscosity ($m^2\ s^{-1}$)
ϕ	electrical potential (V)

Subscripts

a	anode
act	activation
av	average
c	cathode
conc	concentration
i	species
l	limit
r	reactions

Constants

R	Ideal gas constant = 8,314 J $mol^{-1}\ K^{-1}$
F	Faraday's constant = 96485 C mol^{-1}

2 Electrolysis working point and Thermodynamics

To choose the right nominal point, thermochemistry theory is first involved. To be simple, the wished hydrogen production of the alkaline electrolyzer N_{H2} (mol m^{-2} s^{-1}) and its consumed electrical power P (W) depends of the cell imposed potential U (V), the average current density j_{av} (A m^{-2}) and the total surface electrolyzer area S (m^2):

1/ the more the average current density j_{av} (A m^{-2}), the more the hydrogen production. This fact can be checked by the faraday law equation (1).:

$$N_{H2} = j_{av} \, n_e^{-1} \, F^{-1} \quad (1)$$

2/ the less the cell imposed potential U (V), the cheaper the hydrogen production. This fact can be formalized with the potential equation:

$$U_{cell} = \underbrace{\underbrace{\underbrace{E_{rev} + \eta_{ohm}}_{1^{st}} + \Sigma\eta_{act}(j)}_{2^{nd}} + \Sigma\eta_{conc}(j)}_{3^{rd}} \quad (2)$$

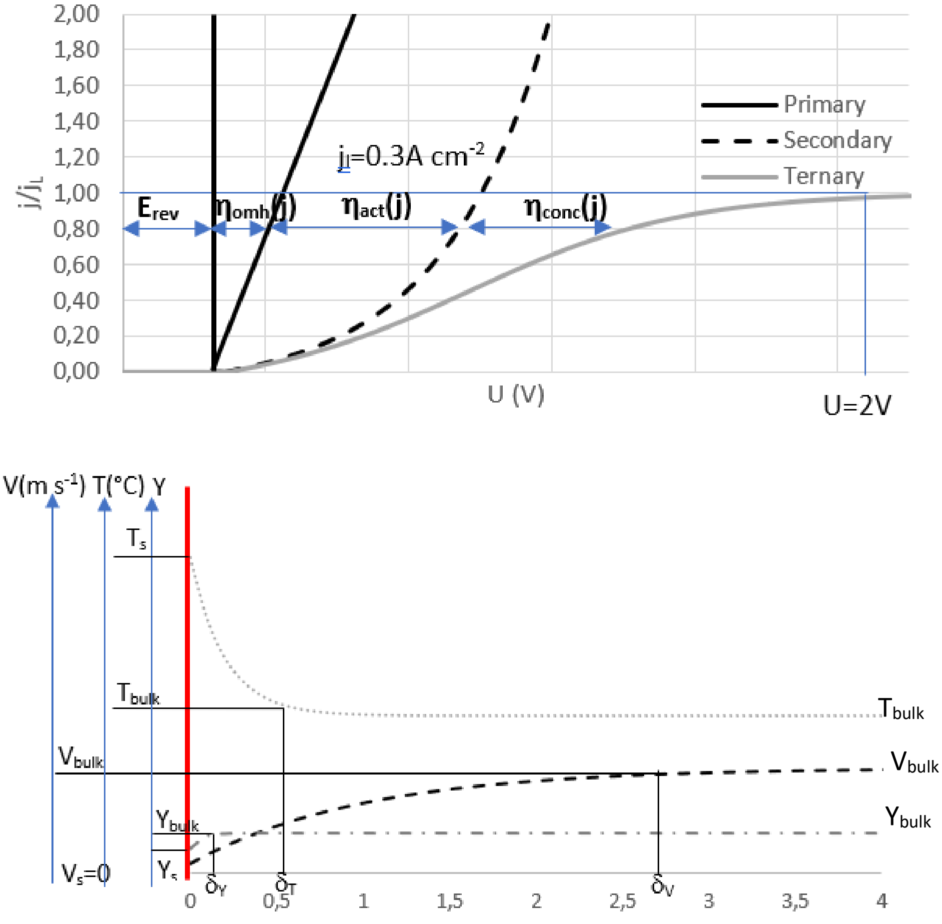

Figure 1-Top: Simulated normalized intensity evolution depending on the tension of the cell according to the three types of model. Bottom : Evolution of the local temperature, mass fraction and velocity near an electrode

With N_{H2} the molar flux in mol m^{-2} s^{-1}, n_e (mol) the electrons number that is exchange during the electrolysis process and F the Faraday constant.

With E_{rev} (V) the reversible potential, η_{act} and η_{conc} respectively the activation and concentration overvoltage in Volt. 1^{st} stands for primary charge modeling, $2^{nd}$for secondary charge modeling and 3^{rd} the ternary modeling.

So, for massive and cheap hydrogen production, the intensity must be the largest as possible, and the applied cell potential must be the lowest as possible. But the consumed potential increases with the average cell applied current density j_{av}. The cell imposed potential is consumed for 3 different fees family (equation 2):

1/ the reversible tension and the ohmic drop defines the primary consumption;

2/ the addition of the activation overpotential at anode and cathode defines the secondary consumption;

3/ the addition of the concentration and bubbles effects over potential defines the ternary consumption.

This last consumption is particularly important for large average current density electrolyzor and really important due to the limiting hydroxide anions flux (N_{OH} in mol m^{-2} s^{-1}) and Oxygen Evolution Reaction (OER) at anode and Hydrogen Evolution reaction (HER) at cathode. Bubbles births, growings and departures lead to a lower effective electro active surface due to screening effect quantified with θ (-) and also change effective two-phase thermophysical properties according with the gas bubbles void fraction ε (-).

Each overvoltages needs optimization

Historically the reversible potential is optimized, then the Ohmic drop and after the activation over voltages… the concentration overoltages are not yet, for many electrochemical systems, optimized because it needs fluid mechanics and flow optimization. Which is our final goal.

The present part will focus here on the reversible tension because this one depends of the thermodynamic. Oliver [13] in his work proved that the reversible tension decreases with increasing temperature. The constraint is the boiling point, H_2O bubble are less conductive than liquid electrolyte and the appearance of more bubbles trigger off a new overpotential. The goal is to stay under this value of boiling point. He also proved that the reversible tension slightly increases with an increasing pressure. However, the increase is little and the higher the pressure is, the smaller the bubbles are and allow a better storage of the produced hydrogen. The pressure sensivity for the parameters:

- The electrical conductivity of electrolytes increases with the pressure according to Hamann [14] and Gancy [15]. Hamann says that the conductivity of KOH increases of 29% between 1bar and 75000 bar. Gancy proved that the electrical conductivity of aqueous KCl increase about

15% between 1 bar and 2000 bar at a temperature of 5°C and less than 1% at a temperature of 85°C. In addition, knowing that the maximum operating pressure for an electrolyzer is 200 bar and the temperature is about 80°C, we can assume that the electrical conductivity is not depending on pressure.

- The water is often considered as incompressible. In fact, its density increases of 2% in average between 1bar and 200 bar for all the temperature, in addition Fine [16] proved that the compressibility factor is 10^{-6} bar^{-1} which is 10^6 less than the compressibility of air.
- The effect of pressure on the viscosity is not negligible between 1 and 200 bar the viscosity of the water is multiplied by 100000 according to Le Neindre [17]. According to Schmelzer [18], the viscosity of water follows an inverse parabolic curve below 33°C.
- We have seen that the water can be considered as incompressible so the specific heat can be taken as constant depending on the pressure.
- For the thermal conductivity of the aqueous electrolyte, Le Neindre's [19] work says that the thermal conductivity of water increases only of 8% (in average) for all the temperature between 110MPa and 250MPa and 3% from 0.1MPa to 110MPa.

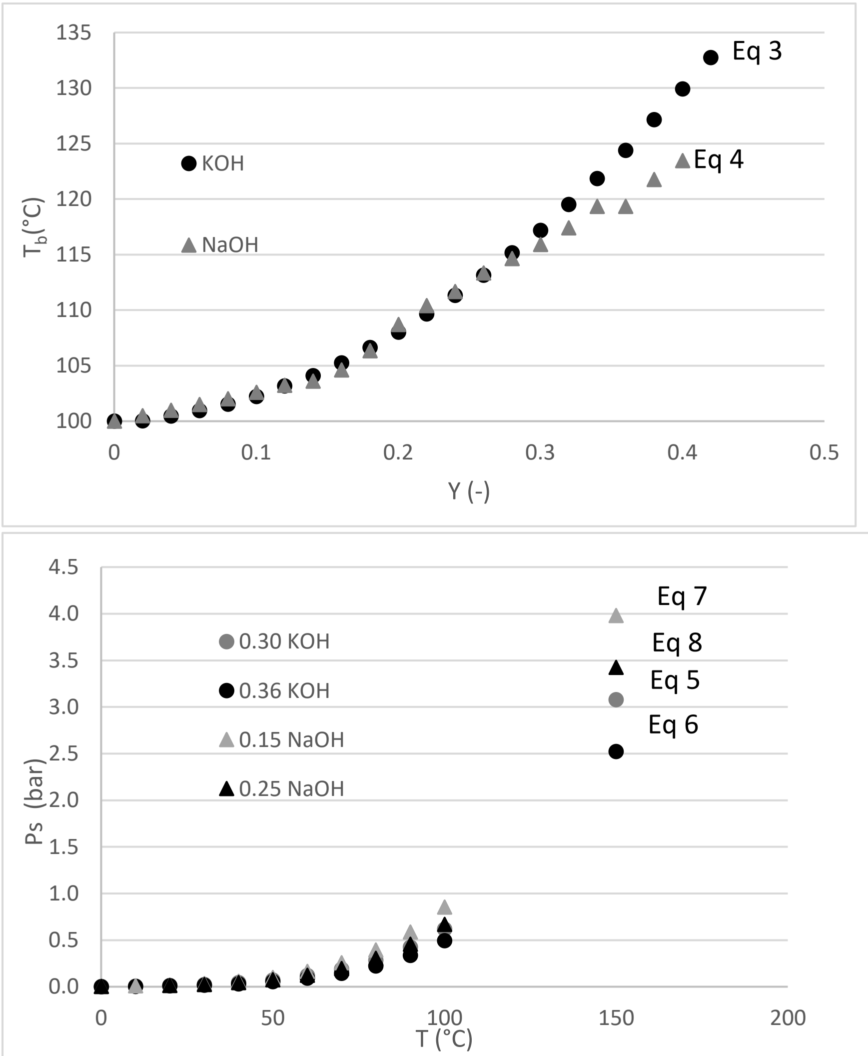

Figure 2-Up:Boiling point of the two electrolytes depending on their local mass fraction, black dot are for KOH boiling point and grey for NaOH boiling point. Bottomt: the saturated vapor pressure of the two electrolytes depending on the local temperature and with a sensitivity of 6% for the local mass fraction. Black is for KOH and grey for NaOH.

Boiling point

For KOH: $T_b(Y)= -5.933\ 10\ Y^3 + 1.756\ 10^2\ Y^2 + 5.533\ Y+9.995\ 10^1$ (3)

For NaOH : $T_b(Y)= -3.92\ 10^2\ Y^3 + 3.214\ 10^2\ Y^2 - 9.395\ 10\ Y + 1.005\ 10^2$ (4)

Saturated pressure

KOH Y=0.3 $Ps(T)= 1.763\ 10^{-6}\ T^3 – 1.633\ 10^{-4}\ T^2 + 5.460\ 10^{-3}\ T – 2.124\ 10^{-2}$ (5)

KOH Y=0.36 $Ps(T)= 1.479\ 10^{-6}\ 10^{-3}\ T^3 – 1.400\ 10^{-4}\ T^2 + 4.635\ 10^{-3}\ T – 1.862\ 10^{-2}$ (6)

NaOH Y=0.15 $Ps(T)= 2.143\ 10^{-6}\ T^3 – 1.843\ 10^{-4} T^2 + 6.103\ 10^{-3}\ T- 2.162\ 10^{-2}$ (7)

NaOH Y=0.25 $Ps(T)= 2.003\ 10^{-6}\ T^3 – 1.880\ 10^{-4}\ T^2 + 6.062\ 10^{-3}\ T- 1.822\ 10^{-2}$ (8)

3 Comparison tool

In this study, several models has been analyzed and compared to experimental data. To help the reader, this part will explain how the comparison has been performed. To compare the correlations with the data, the following equation has been used:

$$\Delta A=(A_{Zaytsev}-A_{correlation})/A_{Zaytsev} \quad (9)$$

$A_{Zaytsev}$ an experimental value of a parameter in SI units. $A_{correlation}$ a value obtained from a correlation. A can be replaced by ρ, λ, σ, Cp etc. Then the following equations have been used to compare correlation and model:

$$\Delta A_{av}=\Sigma\Delta A/N \quad (10)$$

With N the number of ΔA evaluated.

This one give the average errors percentage over a temperature and mass fraction range.

$$\Delta A_{max}=max(\Delta A) \quad (11)$$

This one the maximum errors percentage over a temperature and mass fraction range.

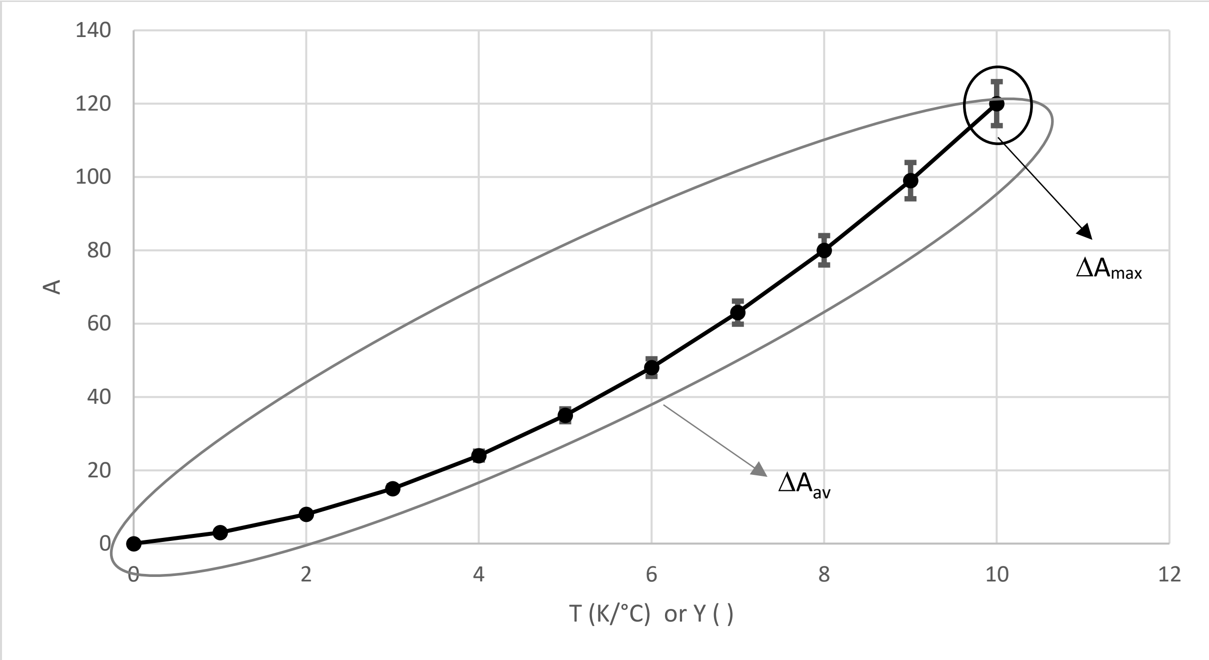

Figure 3-Comparison tools

4 Charge transfer: electrical conductivity

The simplest model to simulate the water electrolysis is a model that just calculate the potential gradient over the cell. It is a one-dimensional model that solve the electric potential equation.

$$\nabla(\sigma\Delta\varphi)+S=0 \tag{12}$$

Only one parameter is needed in this case: the electrical conductivity. This model gives a first approximation of the couple U, j but does not consider the motion of the electrolyte, neither the concentration of OH^-. The corresponding electrolysis model the Ohmic model.

$$U_{cell} = E_{rev} + \eta_{ohm} + \Sigma\eta_{act}(j) \tag{13}$$

In this part, the molarity will be used to calculate the electrical conductivity. Here is the equation used to calculate this molarity.

$$C = Y_{KOH}\, \rho_{KOH}\, M_{KOH}^{-1} \tag{14}$$

Y the mass fraction of KOH in the electrolyte, ρ_{KOH} the density of the electrolyte in kg m^{-3}, M_{KOH} the molar mass g mol^{-1}. C is in mol L^{-1} or M.

4.1 KOH

For the KOH electrical conductivity, the data was taken from Zaytsev [1] and the correlations from See [2] and Gilliam [3]. In Zaytsev [1], the electrical conductivity is available between 0 and 70°C and 0-0.48 of KOH mass. It can be noticed that Klochko supplies few data points for electrical conductivity[4]. All the authors agree on the evolution of the conductivity with temperature and KOH mass fraction. Indeed, the electrical conductivity increases linearly with the temperature and reaches a maximum before decreases for the KOH mass percentage. This maximum is reached between 0.28 (T<10°C) and

0.32 (60°C<T<70°C) in KOH mass. In Allebrod, the authors say that for a temperature superior to 100°C the maximum is superior to 0.375 KOH.

4.1.1 Gilliam equation

Gilliam [3] used a set of experimental data obtained from different other scientist (including See [2]) and from its own experiments. Then he developed this empirical model using a non-linear regression. The model is valid for temperature between T=[0-100°C] and Y=[0.01-0.48].

$$\sigma = -K_1 C - K_2 C^2 + K_3 C T + K_4 C T + K_5 C^3 - K_6 C T^2 \quad (15)$$

With Y the KOH mass fraction, T the temperature in K, σ the electrical conductivity of the electrolyte in S m^{-1}.

4.1.2 See equation

The See's model was developed from See's experimental data [2]: 0.15-0.45, -15-100°C. As Gilliam[3], he uses a non-linear regression to get this model.

$$\sigma = K_1 Y_{KOH} + K_2 T + K_3 T^2 + K_4 T Y_{KOH} + K_5 T^2 Y_{KOH}^{K6} + K_7 T Y_{KOH}^{-1} + K_8 Y_{KOH} T^{-1} \quad (16)$$

Y_{KOH} the KOH mass fraction, T the temperature in K.

4.1.3 Le Bideau equation

Gilliam[3] and See's equation are good model but can be hard to compute or too long. A simple model that can predict the electrical conductivity has been developed using a minimization method.

$$\sigma = K_1 + K_2 T + K_3 Y_{KOH}^2 + K_4 Y_{KOH}^3 + K_5 T Y_{KOH} \quad (17)$$

This model is accurate with an averaged error of 3.34% between T=[40-70°C] and Y_{KOH}=[0.16-0.32]

4.1.4 Comparison with Zaytsev

For See[3], the comparison with See's correlation gives an average difference of 5,36% and a maximum of 11,58% on the range of T=[40-70°C] and Y=[0.02-0.40]. The maximum difference is 11,58%. On this range, there are only two points above 10%, they are both at 40°C with a mass fraction of 0.22 and 0.24. For the Gilliam's correlation[3], an average difference of 4,12% is obtained with of maximum of 10,58%. The two points above 10% are the same as See. Le Bideau's model are simpler to use than the other but it can be used only in the range Y=[0.16-0.32] and T=[40-70°C]. In this range, the average difference is 3.34% with a maximum reached at T=40°C and Y_{KOH}=0.16 of 18.83%.

Table 2-Correlation constants for equation (9) (10) [3], the equations are evaluated between 40-70°C 0.16-0.32 for KOH.

	K_1	K_2	K_3	K_4	K_5	K_6	K_7	K_8	$\Delta\sigma$	$\Delta\sigma_{max}$	Validity range
See [2]	$2.80\ 10^{3}$	$-9.241\ 10^{-1}$	$-1.497\ 10^{-2}$	-9.052	$2.591\ 10^{-2}$	$1.765\ 10^{-1}$	$6.966\ 10^{-2}$	$-2.898\ 10^{1}$	4.63%	11.58%	-15-100°C 0.15-0.45
Gilliam [3]	2.041	$2.800\ 10^{-3}$	$5.332\ 10^{-3}$	$2.072\ 10^{2}$	$1.043\ 10^{-3}$	$3.000\ 10^{-6}$			5.88%	10.71%	0-100°C 10^{-3}-0.45
LeBideau KOH	$3.899\ 10^{1}$	$1.914\ 10^{-1}$	$9.993\ 10^{-3}$	$2.208\ 10^{-1}$	3.564				3.34%	18.83%	40-70°C 0.16-0.32
LeBideau NaOH	$2.658\ 10^{-2}$	$8.671\ 10^{-1}$	$-2.808\ 10^{3}$	$7.112\ 10^{2}$	$8.761\ 10^{1}$				3.71%	17.7%	0-50°C 0-0.25

4.2 NaOH

Zaytsev[1] gives experimental data but unfortunetly no correlations and even after a long research zero correlations has been found to described the evolution of NaOH with temperature and mass fraction. However, a correlation has been designed thanks to the least square method. Zaytsev [1]gives data from 0 to 50°C and Y=[0-0.25]. According to Zaytsev[1], the evolution of the electrical conductivity of the aqueous NaOH is the same as the aqueous KOH but the maximum is reached between 0.16 and 0.20. The designed model has been developed using data between 35-50°C and Y_{NaOH}=[0.08-0.3]. The equation of the model is:

$$\sigma = K_1 + K_2\,T + K_3\,Y_{NaOH}^3 + K_4\,Y_{NaOH}^2 + K_5\,Y_{NaOH} \qquad (18)$$

Over the all domain Y_{NaOH}=[0-0.25] and T=[0-50°C], the average difference is around 3.71% with a maximum of 17.7% at 50°C and 8%.

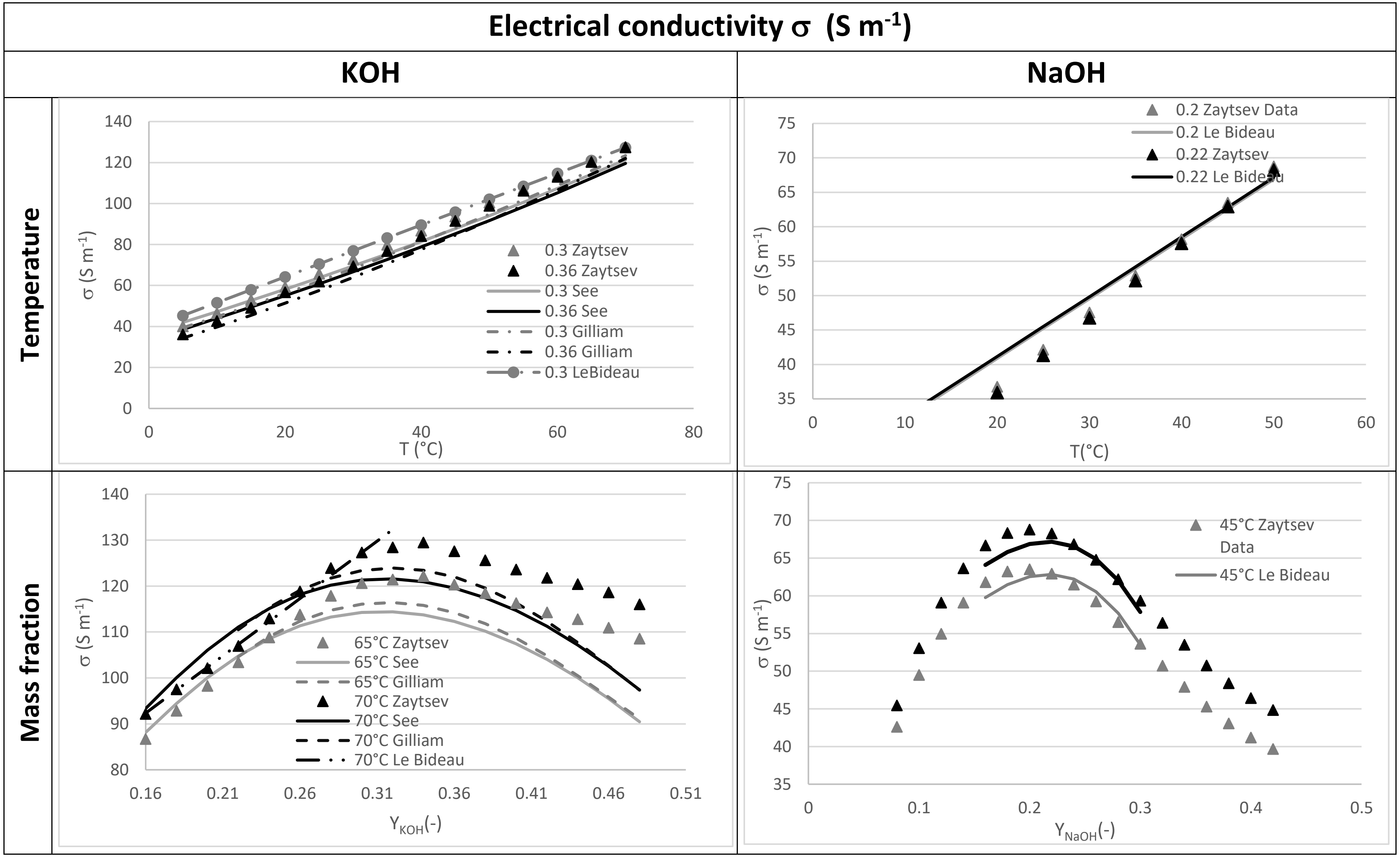

Figure 4- Electrical conductivity of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle rrepresents the experimental point from Zaytsev[1]. For KOH (left) the dotted line is the correlation from Gilliam [3] and the solid line is the correlation from See [2]. For NaOH (right) the dotted line represents the correlation from [12]. For temperature sensitivity (top), black is for Yk=36%(KOH°, 22%(NaOH) and grey for 30% (KOH) 20% (NaOH). For concentration sensitivity, grey is for 75°C and black 85°C

5 Momentum transfer

The calculation of the momentum conversation equation becomes necessary in the case of the ternary charge distribution. Indeed, the species distribution is roughly dependent of the electrolyte motion. This calculation gives the pressure and velocity gradient and it is composed of two equations: the continuity and Navier-Stokes equations. Two parameters are needed here: the density and viscosity.

$$\rho\, d\mathbf{U}/dt + \nabla\cdot(\rho\mathbf{U}\,\mathbf{U}) = -\nabla p + \nabla\cdot\tau + \rho\mathbf{g} \quad (19)$$

5.1 Density

5.1.1 Zaytsev Model

Zaytev[1] used the pycnometric method to determine the density of the two aqueous electrolyte up to 90°C . Then two correlations have been used to extrapolate the results up to 200°C.

For both KOH and NaOH Zaytsev[1] uses the same model.

$$\rho = (K_1 + T\, K_2 + K_3\, T^2)\, 10\text{^}((K_4 + K_5\, T)\, Y_i)) \quad (20)$$

5.1.2 KOH

For the KOH, data between T=[0-200°C] and Y_{KOH}=[0-0.50] KOH are available in Zaytsev[1]. Those data will be compared to correlations from Zaytsev[1] (20) Gilliam [3](21). The sensitivity of the density to the temperature between 0 and 105°C is -0.5 kg m^{-3} K^{-1}. This value varies between -0.47 kg m^{-3} K^{-1} (at 2% KOH) and -0.68 kg m^{-3} K^{-1}. In comparison, the sensitivity with mass fraction is around 10 kg m^{-3} $\%_{KOH}$ and it is constant with temperature. To sum up, the density decreases slowly with temperature but increases rapidly with mass fraction. The two evolutions are almost linear. The two correlations give good results compared to the data (less than 1% of difference).

5.1.2.1 Gilliam model

Gilliam[3] used data set from Zaytsev [1] and other scientist to develop a model of density. In its paper, he choose to use the following form:

$$\rho = \rho_{water}\ \exp(K_4\, Y_{KOH}) \quad (21)$$

With K the value of the water density for one temperature. In order to facilitate the use, the factor K has been remplaced by a thermodependent quadratic polynomial(20).

$$\rho = (K_1\, T^2 + K_2\, T + K_3)\, \exp(K_4\, Y_{KOH}) \quad (22)$$

T the temperature in °C, Y the KOH mass fraction, ρ in kg m^{-3}.

5.1.3 NaOH

Zaytsev [1] gives data for density of NaOH form 0°C to 200°C and Y_{NaOH}=[0-0.40]. The sensitivity with temperature of the density of aqueous NaOH is a little bit smaller than the KOH's aqueous density (-0.78 kg m^{-3} K^{-1} but the progression goes from -0.6 to -1.1 kg m^{-3} K^{-1} from 0.02 to 0.30). The sensitivity with mass fraction is almost the same as KOH's sensivity with mass fraction (17.4 kg m^{-3} $\%_{NaOH}^{-1}$), almost linear for both temperature and mass fraction. The two selected model has been developed by Zaytsev (21) [1] and Churikov[12] (23).

5.1.3.1 Churikov model

First, Churikov[12] has performed pycnometry to measure the density of NaOH, then he used identification to determine the coefficient of the following model:

$$\rho = K_1 + K_2\,T + K_3\,T^2 + (K_4\,Y^2 + K_5\,Y) \tag{23}$$

T the temperature in °C, Y the KOH mass fraction, ρ in kg m^{-3}.

5.1.3.2 Comparison with Zaytsev

The correlation given by Zaytsev[1] is very accurate for highly concentrate NaOH (0,54% of difference T=[60-105°C] Y_{NaOH}=[2-30%] with a maximum of 0,66% for 100°C and Y_{NaOH}=0.3). Churikov's [12] correlation gives satisfactory results for low and high concentration of KOH (1% of difference in average T=[60-105°C] and Y_{NaOH}=[2-40%] with a maximum of 4%).

5.1.4 Multilinear interpolation

For both electrolytes model using the equation of experiment has been developed. Those models are easy to use and since the evolution with the temperature and mass fraction is quasi linear then the models are valid.

$$\rho(Y_i,T) = K_1 + K_2\,Y_i + K_3\,T + K_4\,T\,Y_i \tag{24}$$

T the temperature in °C, Y_i the KOH mass fraction

Table 3-Comparison between model of density. The comparison has been performed between 0.02-0.4 kg⁻¹ and 60-100°C for KOH and 0.02-0.22 and 60-100°C for NaOH.

	K_1	K_2	K_3	K_4	K_5	T	Y_i	$\Delta\rho_{av}$	$\Delta\rho_{max}$
Zaytsev KOH [1]	$1\ 10^3$	$6.20\ 10^{-3}$	$-3.55\ 10^{-3}$	$3.76\ 10^{-1}$	$5.94\ 10^{-4}$	0-200°C	0-0.5	<1%	
Gilliam KOH [3]	$-3.25\ 10^{-3}$	$1.11\ 10^{-1}$	$1.00171\ 10^3$	$8.6\ 10^{-1}$		0-200°C	0-0.5	<1%	
Le Bideau KOH	$1.02\ 10^3$	$1.06\ 10^3$	$-6.09\ 10^{-1}$	$-7.89\ 10^{-1}$		60-100°C	0.02-0.40	0.78%	1.33%
Zaytsev NaOH [1]	$1\ 10^3$	$6.20\ 10^{-3}$	$-3.55\ 10^{-3}$	$4.25\ 10^{-1}$	$-1.15\ 10^{-4}$	0-200°C	0-0.5	<1%	0.66%
Churikov NaOH [12]	$1\ 10^3$	$6.2\ 10^{-3}$	$-3.55\ 10^{-3}$	$-1\ 10^1$	$1.057\ 10^3$	0-50°C	0-0.5	1%	
Le Bideau NaOH	$1.02\ 10^3$	$1.15\ 10^3$	$-6\ 10^{-1}$	-1.25		60-100°C	0.02-0.22	0.25%	0.54%

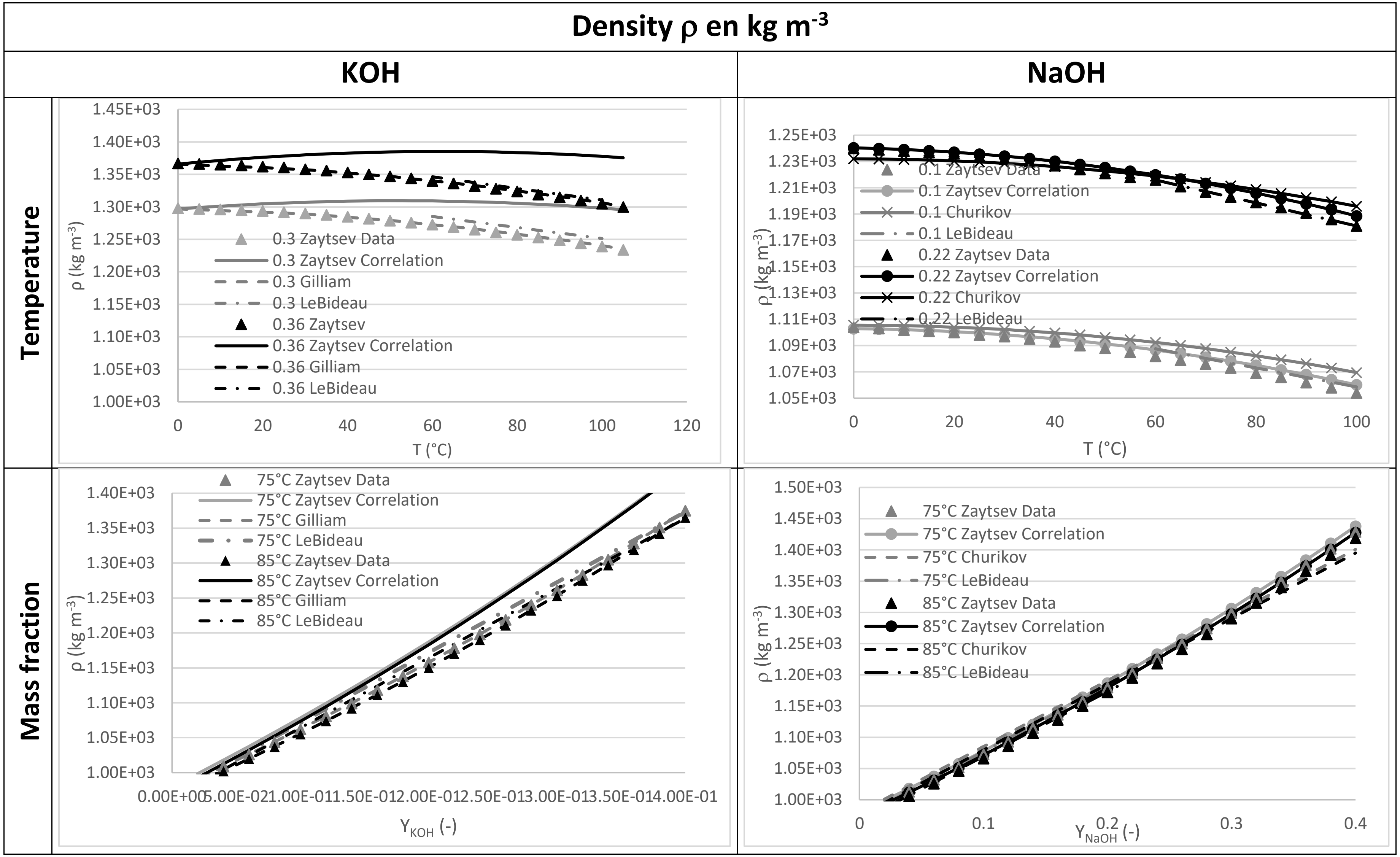

Figure 5-Density of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle and the solid line represents the experimental point and their correlation of Zaytsev [1]. For KOH (left) the dotted line is the correlation from Gilliam [3]. For NaOH (right) the dotted line represents the correlation from Churikov [12]. For temperature sensitivity (top), black is for Yk=36%(KOH°, 22%(NaOH) and grey for 30% (KOH) 20% (NaOH). For concentration sensitivity, grey is for 75°C and black 85°C.

5.2 Viscosity

5.2.1 Zaytsev model

For the viscosity, for both electrolytes, Zaytsev [1] collected data from other Russian/Sovietic scientists. From those data, he determined two correlations (one for the isotherms and one for the isomasses) then he extrapolated the data from 90°C to 200°C.

$$\mu=(K_1\ (K_2 + T) - K_3 + 10\hat{}((K_4 + K_5\ T)\ Y)))\times 10^3 \quad (25)$$

T the temperature in °C, Y_i the mass fraction, µ in mPa s.

5.2.2 KOH

5.2.2.1 Guo model

The following model was firstly designed for a ternary electrolyte system K_2CrO_4-KOH-H_2O. The empirical model (24) was determined using capillary viscometer data. The measurements go from 15 to 60°C.

$$\mu= \exp(\ K_1 + K_2\ T + K_3\ T^2 + K_4\ C) \quad (26)$$

T the temperature in °C, M_{KOH} the molar mass of KOH in g mol^{-1}; Y_{KOH} the mass fraction, the density in kg m^{-3}, µ in mPa s.

5.2.2.2 Analysis and comparison with Zaytsev

The available data in Zaytsev's books [1] are 0-200°C and Y_{KOH}=[0-0.50]. The viscosity decreases exponentially with the temperature whereas it increases exponentially with the mass fraction. To model this evolution, two models has been chose: Zaytsev's model [1] (13) and Guo's model [5] (24). The Zaytsev's model [1] described the evolutions in mass fraction and temperature with an average difference of 2,9% a maximum of difference of 18% is reached at 60°C and Y_{KOH}=0.40 of mass. Guo's model [5] has an average difference with Zaytsev [1] of 5% for temperature between 20-60°C and mass fraction between Y_{KOH}=[0-0.40] KOH but for temperature superior to 60°C a divergence is observed (10% of difference for all the mass fraction). Due to high divergence with linearity of viscosity evolution and the availability of model to describe this evolution, no additional model has been developed. The best model is Zaytsev.

5.2.3 NaOH

5.2.3.1 Olson model

Olsson [11] collected data from other studies and used them to make a correlation.

$$\mu = (10\hat{}(\log_{10}((\ 5.98\ 10^{-1}\ (4.32\ 10^1 + T) - 1.54\ )) + (\ 3.39 - 1.12\ 10^{-2}\ \ T)\ Y))\ 10^3 \quad (27)$$

$$\ln(\mu_{NaOH}/\mu_{H2O}) = d_1 + d_2\ T^{1/2} + d_3\ T\ (20)$$

$$d_1 = k_1\ YNaOH +\ldots + k_4\ Y_{NaOH}^4$$

$$d_2 = l_1\ Y_{NaOH} +\ldots+ l_5\ Y_{NaOH}^5$$

$$d_3 = m_1 Y_{NaOH} + \ldots + m_5 YNaOH5$$

$$\mu_{H2O} = \exp(n_0 + n_1 T + n_2 T^{1.5} + n_3 T^{2,5} + n_4 T^3)$$

Table 4-Parameter for Olson's correlation [11]

k_1	-6.14	l_1	2.32	m_1	$-1.152\ 10^{-1}$	n_0	$5.87\ 10^{-1}$
k_2	$1.25\ 10^2$	l_2	$-2.3\ 10^1$	m_2	1.05	n_1	$-3.98\ 10^{-1}$
k_3	$-2.47\ 10^2$	l_3	$4.93\ 10^1$	m_3	-2.37	n_2	$2.47\ 10^{-3}$
k_4	$1.47\ 10^2$	l_4	$-3.697\ 10^1$	m_4	2.10	n_3	$-4.94\ 10^{-6}$
		l_5	6.58	m_5	$-5.25\ 10^{-1}$	n_4	$1.49\ 10^{-7}$

This correlation can be used only in the following interval.

Table 5-Domain of validity of the Olsson's correlation [11]

Y_{NaOH}	T (°C)
Between 0.02-0.4	20-30°C
Between 0.02-0.45	30-50°C
Between 0.02-0.55	50-70°C
Between 0.02-0.70	70-150°C

5.2.3.2 Comparison with Zaytsev

For NaOH, data in Zaytsev [1] are available between 0 and 200°C and 0-0.50 of mass fraction. The two models are Zaytsev's (25) and Olson's model (27). The Zaytsev's model [1] has an average difference of 5% whereas Olson's one is 6% over the range T=[60-105°C] and Y_{NaOH}=[2-0.40]. Also, for Olson's model the difference reaches 15% for 100°C and 18% and this model has the particularity to have an interval of validity depending the mass fraction. This domain of validity is presented in the table5.

Table 6-Comparison of viscosivity model. Comparison has been performed between 40-100°C 0-0.4

	K_1	K_2	K_3	K_4	K_5	T	Y_i	$\Delta\mu_{av}$	$\Delta\mu_{max}$
Zaytsev KOH [1]	$5.98\ 10^{-1}$	4.33 10	1.54	1.12	$2.03\ 10^{-3}$	0-200°C	0-0.50	2.9%	18%
Guo KOH [5]	$4.3\ 10^{-1}$	$-2.51\ 10^{-2}$	10^{-4}	$1.3\ 10^{-1}$		20-60°C	0.02-0.4	5%	10%
Zaytsev NaOH[1]	$5.98\ 10^{-1}$	4.33 10	1.54	3.39	$-1.12\ 10^{-2}$	0-200°C	0-0.50	5%	

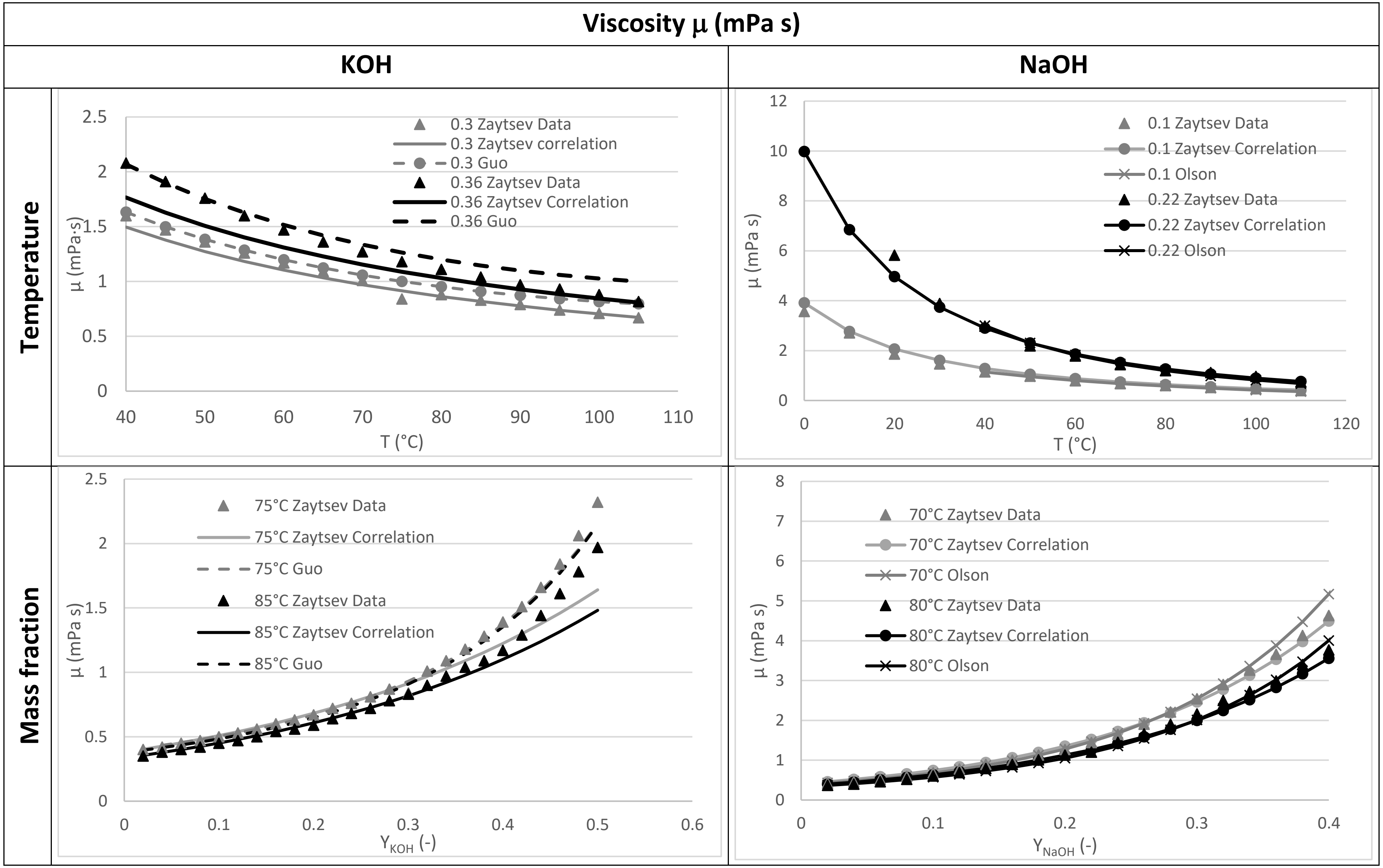

Figure 6- Viscosity of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle and the solid line represents the experimental point and their correlation of Zaytsev [1]. For KOH (left) the dotted line is the correlation from Guo [5]. For NaOH (right) the dotted line represents the correlation from Olsson [11]. For temperature sensitivity (top), black is for Yk=36%(KOH°, 22%(NaOH) and grey for 30% (KOH) 20% (NaOH). For concentration sensitivity, grey is for 75°C(KOH) 70°C(NaOH) and black 85°C(KOH) 80°C(NaOH).

6 Heat transfer

The calculation of the heat equation can bring more precision on the calculation of the pressure and velocity gradient and can influence also the species transport. Indeed, a temperature gradient trigger off natural convection and most of the parameters have a temperature dependency. To calculate the temperature gradient three parameters are needed: the density, the specific heat, the thermal conductivity. The two first are necessary only in time dependent studies because they characterize the inertia of the system. The last one is needed for both stationary and time dependent study.

$$\rho\ Cp\ dT/dt + \rho\ Cp\ \nabla \cdot UT = \Delta(\lambda\ T) + P \quad (28)$$

6.1 Specific heat

6.1.1 Zaytsev model

The experimental method used to get data by Zaytsev [1] is :

- For KOH, he used an isothermal glass calorimeter then he used the same technic of extrapolation as for the other parameters
- For NaOH, he collected data from other sovietic scientists and exprotaled them.

The same model is used by Zaytsev [1] to describe the evolution of specific heat for NaOH and KOH.

$$Cp = K_1 + K_2 \ln(T/100) + (K_3 + K_4 Y + 8 T) Y_i \quad (29)$$

With T the temperature in °C and Y the mass fraction.

6.1.2 Method of Lalibertée

Laliberté [6] use the following equation to model the specific heat of the KOH and NaOH. In his article, Laliberté[6] explain how to use his method. First, experimental must be collected then initial coefficient must be chose. The squared difference between the experimental and model data is made and this difference gives a criterion to minimize using a solver.

$$Cp = Y_i (K_1 \exp(\alpha) + K_5 Y_i^{K6}) + (1-Y_i) Cp_{water} \quad (30)$$

$$\alpha = K_2 T + K_3 \exp(0.01 T) + K_4 Y_i \quad (31)$$

6.1.3 Multilinear interpolation

$$Cp(Y_i,T) = K_1 + K_2 Y + K_3 T + K_4 T Y_i \quad (32)$$

With T the temperature in °C and Y the mass fraction.

Due to the quasi linearity of the specific heat evolution with temperature and mass fraction a Multilinear interpolation has been developed. The table gives the good parameters to use and the domain of validity.

6.1.4 KOH

The study of the temperature sensitivity shows that the evolution with temperature of the specific heat is hyperbolic because the derivative of the specific heat depending on the temperature increases slighty before decreases (the increase and the decrease is in average around 3 kJ kg^{-1} K^{-2}). This evolution is negligible compared to the evolution depending on the mass fraction, which linearly decreasing (around -33 kJ kg^{-1} $K^{-1}\%_{KOH}^{-1}$). To model has been selected: Zaytsev's model(29) [1] and Laliberté's(30) (31) model[6]. Zaytsev's model (29) [1] has an accuracy of 2% in average between Y_{KOH}=[0-0.40] KOH and 0-100°C with a maximum to 8%. Laliberté's model [6] does not follow the temperature sensitivity but this problem can be due to a solver problem. In contrast, the evolution depending on the mass fraction is respected.

6.1.5 NaOH

In his book Zaytsev [1] gives data from 0 to 200°C and Y_{NaOH}=[0-0.42]. The correlation given by Zaytsev [1] has an average difference of 2% with a maximum at 4.32% at 90°C and 20% NaOH. The method of Laliberté[6] was used to get another model, the same problem as for KOH can be observed. This model is valid between 60 and 100°C. The average difference is around 3% with a maximum of 8% for 100°C and Y_{NaOH}=0.20.

Table 7-Comparison of different specific heat correlation. The comparison has been performed between 60-100°C for NaOH and KOH and 0-0.4 for KOH and 0-0.2 for NaOH

	K_1	K_2	K_3	K_4	K_5	K_6	T	Y_i	ΔCp_{av}	ΔCp_{max}
Zaytsev KOH [1]	$4.236\ 10^3$	1.075	$-4.831\ 10^3$	8			0-200°C	0-0.4	2%	8%
Zaytsev NaOH [1]	$4.236\ 10^3$	1.075	$1.576\ 10^3$	$1.59\ 10^1$			0-200°C	0-0.4	2%	4.32%
Laliberté KOH [6]	1	$1.160\ 10^{-5}$	$4.037\ 10^{-1}$	$1.500\ 10^{-3}$	$7.048\ 10^3$	2.99	60-100°C	0.02-0.4	2%	8%
Laliberté NaOH [6]	$2.426\ 10^1$	0	1.68	0	$1.141\ 10^2$	1.77	60-100°C	0.02-0.2	3%	8%
Le Bideau KOH	$4.101\ 10^3$	$-3.526\ 10^3$	$9.644\ 10^{-1}$	1.776			60-100°C	0.02-0.4	1.79	4.02%
Le Bideau NaOH	$3.879\ 10^3$	$-2.068\ 10^2$	$6.63\ 10^{-1}$	$-2.36\ 10^{-1}$			60-100°C	0.02-0.2	1.09%	1.95%

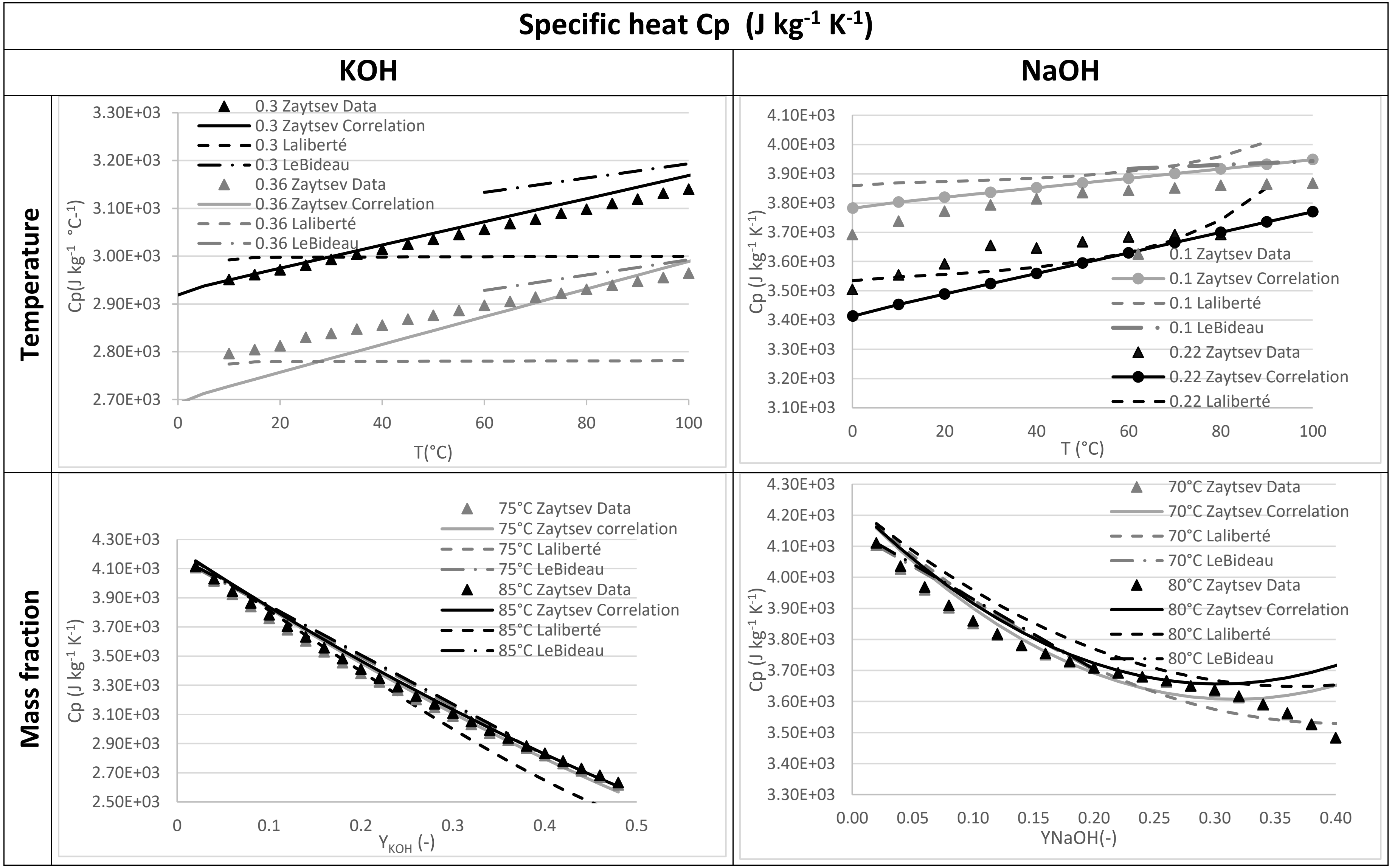

Figure 7- Specific heat of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle and the solid line represents the experimental point and their correlation of Zaytsev [1]. For KOH (left) and NaOH the dotted line is the correlation determined using method from Laliberté [6]. For temperature sensitivity (top), black is for Yk=36%(KOH°, 22%(NaOH) and grey for 30% (KOH) 20% (NaOH). For concentration sensitivity, grey is for 75°C(KOH) 70°C(NaOH) and black 85°C(KOH) 80°C(NaOH).

6.2 Thermal conductivity

6.2.1 Zaytsev model

Zaytsev [1] has selected data from other scientists and then used the same method as before to extrapolate the results.

$$\lambda = (K_1 + K_2 T - K_3 T^2) (1-Y_i K_4) \quad (33)$$

6.2.2 Wang model

Wang[8] assumed that the thermal conductivity can be modeled by taking into account the water thermal conducitivity, the interaction between the solvent ($\Delta\lambda^s$) and the ion species, the interaction between two ion species($\Delta\lambda^{s\text{-}s}$).

$$\lambda_{elec} = \lambda_{water}(T) + \Delta\lambda^s + \Delta\lambda^{s\text{-}s} \quad (34)$$

For our binary system (NaOH, KOH-H_2O), the previous model becomes:

$$\lambda_{elec} = \lambda_{water}(T) + X_i (\alpha_{1i} + \alpha_{2i} \exp(-AT)) + X_k (\alpha_{1k} + \alpha_{2k} \exp(-AT))) (\beta_1 \exp(\beta_2 T)) X_k X_i) \quad (35)$$

For KOH i=K^+ k=OH^-, for NaOH i=Na^+, k=OH^- A=-0.023

6.2.3 KOH

For the KOH, data are available between T=[0-155°C] and Y_{KOH}=[0-0.40] of KOH mass fraction. The thermal conductivity increases with the temperature but decreases with the mass fraction. The sensitivity depending on the mass fraction and temperature is the same order of magnitude. The two chose models are Zaytsev's [1](33) and Wang's (35) model[8]. The Zaytsev's model [1] between T=[20-115°C] and Y_{KOH}=[0-0.40] deviate from its values about 1.5% with a maximum of 4.4% for 115°C and 0.2 in mass. In his publication Wang[8] does not give the values of the coefficient for his model for KOH but they have been identified. The resulted model is valid for 60°C and 100°C and Y_{KOH}=[0.02-0.40]. It deviates of 0,6% with a maximum of 3%.

6.2.4 NaOH

The Data available in Zaytsev [1] are for temperatue between 0-155°C and Y_{NaOH}=[0-0.35]. The correlation given by Zaytsev (28) is accurate for T=[20-115°C] and Y_{NaOH}=[0.05-0.35] with an average difference of 4.92% and a maximum 12.04% for 20°C and 0.35. The original correlation given by Wang [8] (32) has an average difference of 10% with the Zaytsev's data and reaches a maximum of 35% for T=40°C and Y_{NaOH}=0.35. However, after using a minimization method, another interaction parameter has been found. By replacing the original parameters by the new one, the average difference falls to 3% and the maximum with a maximum of 6% for T=40°C and Y_{NaOH} =0.35.

Table 8-Parameters thermal conductivity model

	α_{1i}/K_1	α_{2i}/K_2	α_{1k}/K_3	α_{2k}/K_4	β_{wang}		$\beta_{LeBideau}$		$\Delta\lambda_{av}$	$\Delta\lambda_{max}$
					1	2	1	2		
Zaytsev KOH[1]	5.545 10^{-1}	2.460 10^{-3}	1.184 10^{-5}	1.280 10^{-1}					1.5%	3%
Wang KOH[8]	-3.8249 10^{-1}	4.49 10^{-2}	4.923 10^{-1}	-1.8 10^{-2}	--	--	-2.5	--	0.6%	4.5%
Zaytsev NaOH[1]	5.545 10^{-1}	2.460 10^{-3}	1.184 10^{-5}	1.260 10^{-1}					4.92%	12.04%
Wang NaOH[8]	0	0	4.923 10^{-1}	-1.8 0^{-2}	-4.95	-2.5409 10^{-4}	-1.95	-2.5409 10^{-4}	$10\%_{wang}/3\%_{LeBideau}$	$35\%_{wang}/6\%_{LeBideau}$

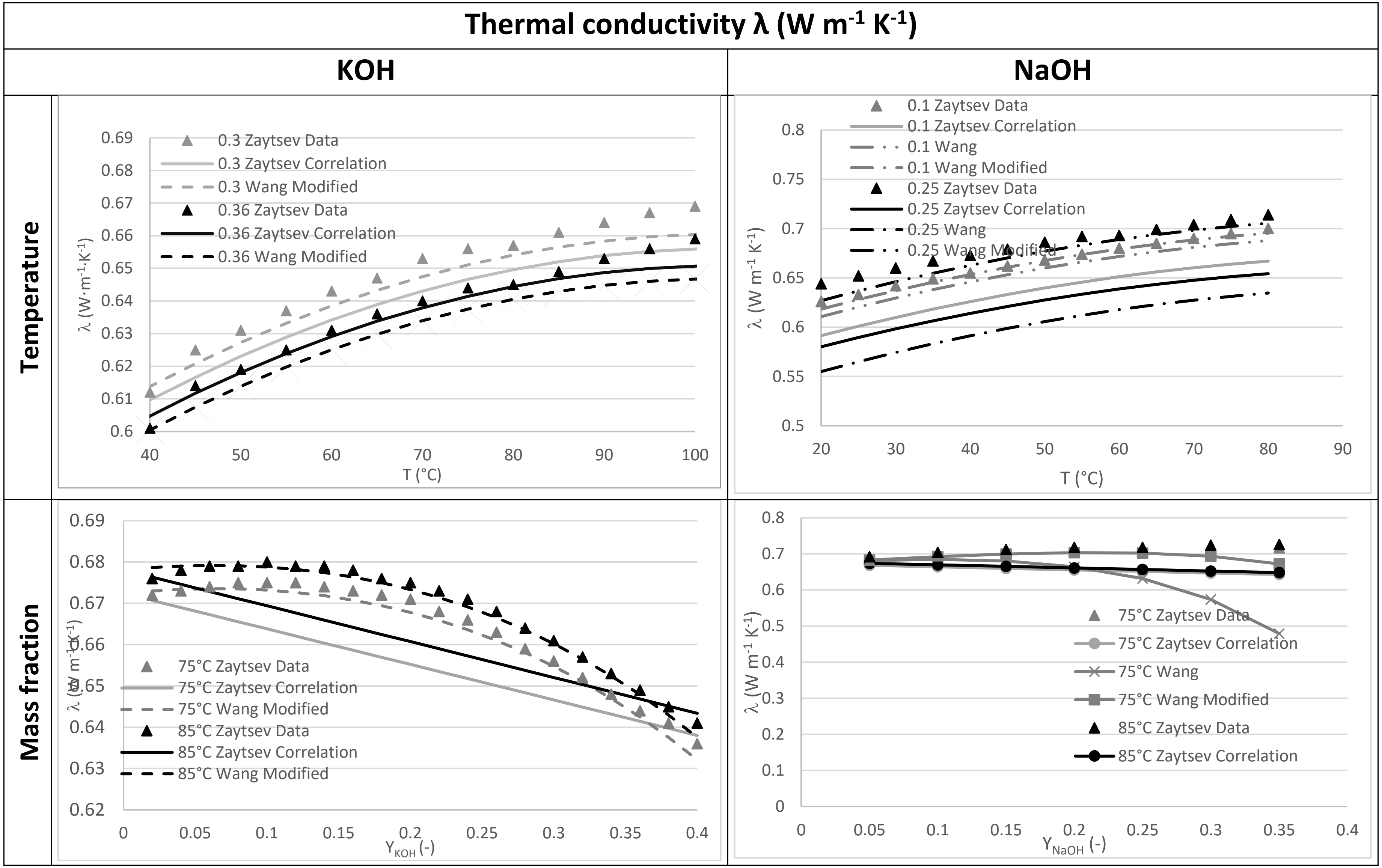

Figure 8 Thermal conductivity of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle and the solid line represents the experimental point and their correlation of Zaytsev [1], the dotted line is the correlation from [8] and the dotted line is a modified correlation using Wang [8]]. For temperature sensitivity (top), black is for Yk=36% and grey for 30%. For concentration sensivity (bottom), grey is for 75°C and black 85°C.

7 Mass transfer: mass transfer coefficient

The ternary charge distribution shows a limit in current density. This limit is governed by the mass transfer. Indeed the limit current density is depending on the coefficient of diffusion, the hydrodynamic/mass transfer and the bulk concentration of active species.

$$j_L = z\ F\ D\ C_{bulk}\ \delta^{-1} = z\ F\ k\ C_{bulk} \quad (36)$$

The coefficient D must be known. The data given by Zaytsev [1] are not numerous and are presented on the form of D_{KOH} this means that's the ions OH^- and K^+/Na^+ respect the electroneutrality which is true in the bulk but not true near the electrodes[E. L. Clusser] due to the presence of the layer of negative charge.

7.1 Multilinear interpolation

Zaytsev [1] does not give correlations modelling the diffusion coefficient and the data for NaOH were rare. However, a multilinear interpolation has been developed to model the evolution of this parameter for NaOH and KOH. Nevertheless, due to the lack of data for NaOH the model must be used with keeping in mind that it is a extrapolation of few data points.

$$D(Y_i,T) = K_1 + K_2\ Y + K_3\ T + K_4\ T\ Y \quad (37)$$

Table 9-Parameters for the Multilinear interpolation to calculate the mass transfer coefficient

	K_1	K_2	K_3	K_4	T	Y_i	ΔD_{av}	$\Delta DMax$
KOH	$-1.05\ 10^{-1}$	2.45	$9.20\ 10^{-2}$	$1.148\ 10^{-2}$	40-70°C	0.05-0.40	2.24%	5.78%
NaOH	1.05	-4.70	$3.32\ 10^{-2}$	$4.04\ 10^{-2}$	15-20°C	0.004-0.02		

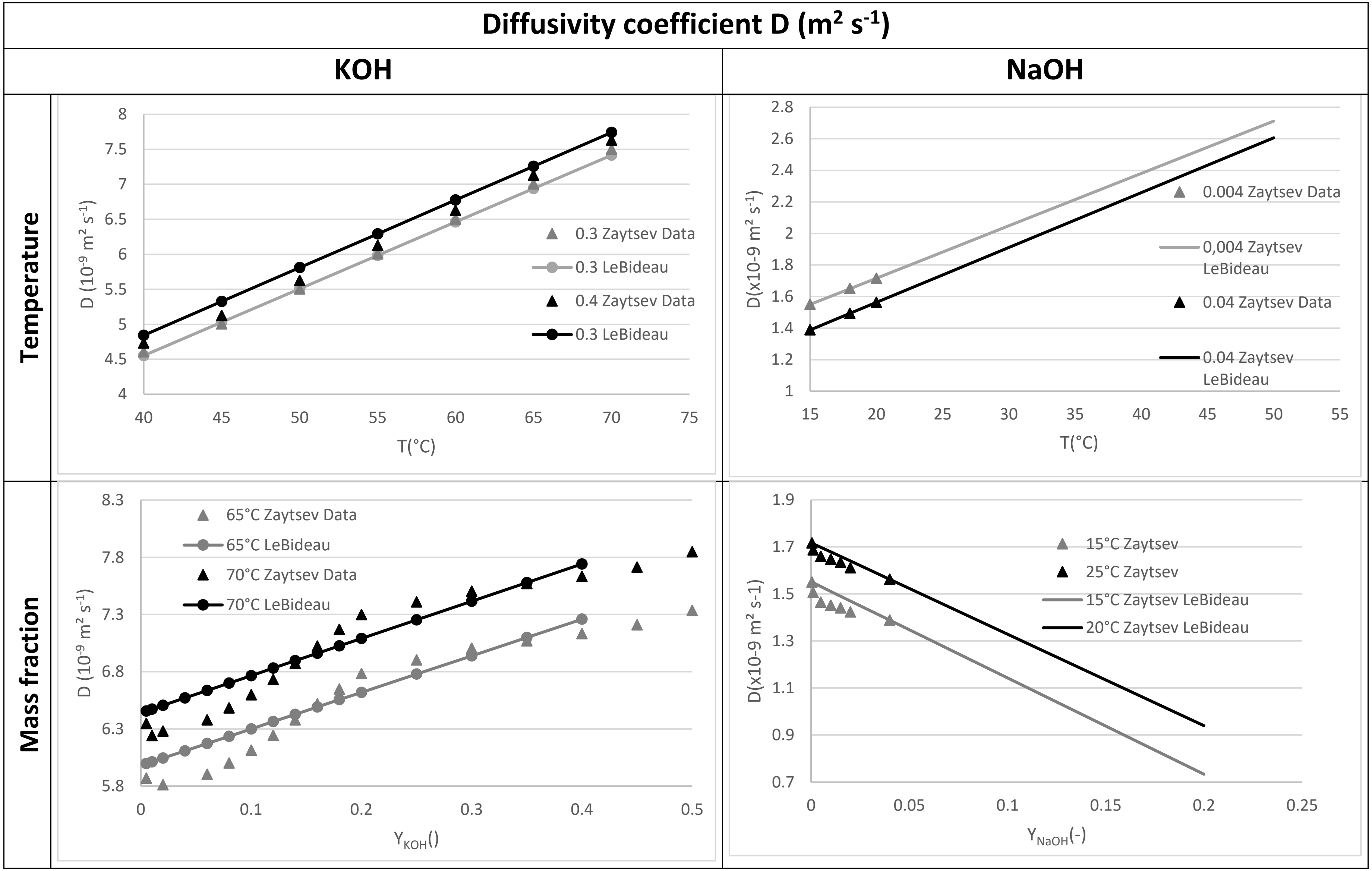

Figure 9 Diffusion coefficient of KOH and NaOH depending on temperature and mass fraction. For both KOH and NaOH, triangle are data from Zaytsev and solid line are LeBideau's model.

8 Conclusion

All the parameters for simulating with accuracy the alkaline water electrolysis using CFD. However, it must be reminded that this model is monophasic whereas real electrolysis is at least biphasic due to the presence of H_2 and O_2 bubbles and further work must be performed in order to review thermal, electro-kinetic parameter and transfer properties of electrode material. In addition, the values summarized in this article are accurate for monophasic flow only. The influence of gas-bubble will be investigated in another article.

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9 Figure