Application of Unconventional Techniques for Evaluation and Monitoring of Physico-Chemical Properties of Water Streams

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Abstract - **Industrial processes requires the evaluation and the monitoring of some water properties like Electrical Conductivity (EC) and Langelier Saturation Index LSI), which are of interest for the estimation of water quality. Analytical representation of water properties can be useful in Process Modelling and Simulation of industrial systems. The use of common simulation softwares allows to test industrial processes under conditions that cannot be easily or safely tested; nevertheless estimation of Electrical conductivity and Langelier Saturation Index is usually not possible or requires particular software licenses. Application of unconventional techniques can fix the problem. In particular for Aspen Plus commercial software the availability of customizable calculator blocks allows the user to represent specific features. The paper presents the unconventional method developed to calculate EC and LSI values in Aspen Plus modelling FORTRAN based calculator blocks starting from literature information.**

Keywords - Electrical conductivity; Langelier Saturation Index; Process Modelling; Process Simulation; Water Properties Evaluation and Monitoring

I. INTRODUCTION AND LITERATURE REVIEW

Water use is fundamental for many industrial processes; only suitable water quality allows the correct process operations and prevents undesiderable phenomena like corrosion and fouling [1] [2]. Water properties evaluation and monitoring are fundamental in online measurement in order to avoid excessive water exploitation and frequent plant maintenance. Nowadays exploration of industrial system behaviour under conditions that cannot be easily or safely tested requires ad hoc developed studies, also supported by Process Modelling and Simulation (PM&S) [3]. PM&S allow in-depth investigation of future scenarios in order to evaluate the feasibility of process integration solutions [4] and multi-objective optimization to improve economic and environmental process sustainability according to [5] and [6]. Examples of PM&S applications can be the choice of the best industrial practice for a correct byproduct and waste generation and management [7] or the minimization of freshwater consumption and wastewater generation [8] [9]. For this reason, in addition to online measurements, analytical representation of water properties obtains leading importance. In a simplified approach basic models can provide results which are accurate enough for practical applications: for example simple Excel-based model can evaluate pH and electrical conductivity computing the ionic representation of a water streams [10]. In other cases only more complex models allow focused studies and simulation sotwares are useful to this aim. However, many common simulation softwares (e.g. Aspen Plus [11]) do not estimate important physico-chemical properties such as electrical conductivity or Langelier Saturation Index (or require dedicated licences). The present paper describes the development of FORTRAN based calculator blocks to evaluate above mentioned properties in Aspen Plus simulation software. After an introduction about electrical conductivity and Langelier saturation index and a basic description of Aspen Calculator blocks, Section III gives information about the assumptions and the methods exploited in the modelling of EC and LSI calculators. Section IV concerns models validation and examples of applications. The last section is about the important and potential contribution that the developed models can have in process simulation.

II. DESCRIPTION OF KEY CONCEPTS

Electrical conductivity and Langelier Saturation Index are common parameters used to monitor water quality in many industrial fields. Due to their qualitative nature, they don't give quantitative values but are very useful to monitor water contamination and suitability during the whole industrial process.

A. Electrical Conductivity

Electrical conductivity (EC) of water solution is a measure of its capacity to conduct electric current and it increases with temperature as the mobility of the ions increases [12]. Usually conductivity is linked with total dissolved solids (TDS) but it is important to consider that it does not take into account non conductive contaminants and therefore it is only a measure of electrolytes TDS.

Due to the EC dependence on concentration and nature of the ions, this variable is usually exploited in many industrial field (e.g. water treatment, desalination, leak detection, clean in place, interface detection) as a qualitative but not specific indicator of conductive ions content in a solution (Tab.I).

B. Langelier Saturation Index

The Langelier Saturation Index (LSI) is based on carbonate equilibrium in water and it is defined as [14]:

$$LSI = pH - pH_S \quad (1)$$

where pH is the actual pH of a water stream and pHs [15] the saturation pH of calcium carbonate based on the equilibrium expressions of calcium carbonate solubility and bicarbonate dissociation; the effects of temperature and ionic strength are considered.

Therefore, LSI represents the degree of saturation of calcium carbonate and its magnitude and sign are qualitative indicators of whether water will be corrosive or calcium carbonate will precipitate (Tab. II). If water pH is equal to pHs and LSI = 0, water is balanced. If LSI is positive, water is defined nonaggressive, it is oversaturated of *CaCO3* and scale-forming; LSI value in the range 0-0.4 shows a water tendency to inhibit corrosion with the formation of a protective film that insulates pipes and other plant components from direct contact with water. Finally, if LSI is negative, water is corrosive (or aggressive) because it is unsaturated of *CaCO3*, it will dissolve carbonates and prevents the formation of the protective carbonate coatings. In spite of its qualitative nature, LSI is a good medium in assessing water treatment requirements in industrial processes.

III. ASPEN PLUS CALCULATOR BLOCKS

Calculator blocks in Aspen Plus allow the user to carry out flowsheet calculations using equations printed in ad hoc made Excel spreadsheet or FORTRAN statements. The calculator results must be viewed by directly examining the values of the variables modified by block as in [16]. The use of Aspen calculator block is possible following a fixed sequence: 1) model variables to sample (import variables) or manipulate (export variables) must be identified;

TABLE I.
TYPICAL WATER ELECTRICAL CONDUCTIVITY [13]

Water Solution	Electrical Conductivity
High quality deionized water	about 5.5 μS/m
Typical drinking water	about 5 to 50 mS/m
Sea water	about 5 S/m

TABLE II.
WATER CHARACTERISTICS [18]

Water Characteristics	LSI
Highly aggressive (corrosive)	< −2.0 to −0.5
Slightly aggressive	< −0.5 to 0.0
Balanced	0
Nonaggressive (Protective film formation)	0.0 to 0.4
Nonaggressive (Scale-Forming)	> 0.4

2) FORTRAN statements (or Excel spreadsheet) must be compiled;

3) the sequence in which the blocks are executed during flowsheet calculations must be specified. Aspen Plus Help describes import and export variables as follows [17]:

- import variables establish information flow from the block or stream containing a sampled (read-only) variable to the calculator block;
- export variables establish information flow from the calculator block to Aspen Plus. Using the FORTRAN statements user has to pay particular attention in the variables definition and related unit of measurements: possible change in units must be handled by the user specifing directly in Fortran statements conversion factors because of Fortran limits in units managing.

IV. MODELS FOR EC AND LSI CALCULATION

Models have been developed using literature information about EC relationship with ion concentration and LSI relationship with alkalinity, calcium concentration, total dissolved solids and water temperature. After the characterization of simulated water stream, import and export variables have been specified to allow the use of FORTRAN compiler. Modelling informations are now presented for each model developed: a calculator for electrical conductivity and two different calculators for Langelier saturation index (Fig. 1).

A. Modelling of Electrical Conductivity Calculator

Electric current is carried differently by cations and anions. Therefore in the modelling of EC calculator the conductivity factors for major ions have been considered (Tab. III).

The following approximate relationship is used in the FORTRAN compiler to obtain electrical conductivity in μS/cm [19]:

$$EC = \sum XCi \cdot fi \quad (2)$$

where Ci is the concentration of ionic specie i in solution expressed in mg/L and fi is the conductivity factor for ionic specie i.

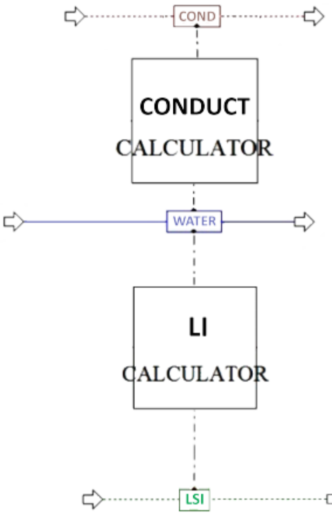

Figure 1. Electrical conductivity and Langelier Saturation Index Calculators in Aspen Plus.

Concentrations of ionic species are import variables for the model; due to some limits of Aspen Plus, each ionic concentration in the calculator block can be imported only as mass fraction (Ci,w [mg/mg]) The use of the previous EC correlation is possible if the concentration (Ci[mg/L]) is expressed as mass/volume concentration. Therefore, the desidered expressions for concentrations have been evaluated as follows:

$$C_i[mg/L] = C_{i,w} \cdot \rho \cdot 10^3 \frac{mg/kg}{dm^3/m^3} \tag{3}$$

with ρ expressed in *kg/m³*

The results of the previous formula are considered "auxiliary" exported variables. The conductivity factors have been inserted as constants in FORTRAN compiler. The only real export variable is the calculated value of electrical conductivity of water streams. Table IV lists all the import and export variables used in the model.

TABLE III. CONDUCTIVITY FACTORS FOR COMMON IONS IN WATER [19]

Ion species	Conductivity factor
Ca^{2+}	2.6
Mg^{2+}	3.82
K^+	1.84
Na^+	2.13
HCO_3^-	0.715
Cl^-	2.14
SO_4^{2-}	1.54
NO_3^-	1.15

TABLE IV. EC MODEL VARIABLES.

ID	Unit	Variable	Type
CA	mg/mg	Ca^{2+} Ion mass fraction	Import
MG	mg/mg	Mg^{2+} Ion mass fraction	Import
K	mg/mg	K^+ Ion mass fraction	Import
NA	mg/mg	Na^+ Ion mass fraction	Import
HCO	mg/mg	HCO_3^- Ion mass fraction	Import
CL	mg/mg	Cl^- Ion mass fraction	Import
SO	mg/mg	SO_4^{2-} Ion mass fraction	Import
NO	mg/mg	NO_3^- Ion mass fraction	Import
DENS	kg/m^3	Solution density	Import
CAC	mg/L	Ca^{2+} Concentration	Auxiliary
MGC	mg/L	Mg^{2+} Concentration	Auxiliary
KC	mg/L	K^+ Concentration	Auxiliary
NAC	mg/L	Na^+ Concentration	Auxiliary
HCOC	mg/L	HCO_3^- Concentration	Auxiliary
CLC	mg/L	Cl^- Concentration	Auxiliary
SOC	mg/L	SO_4^{2-} Concentration	Auxiliary
NOC	mg/L	NO_3^- Concentration	Auxiliary
COND	$\mu S/cm$	Electrical conductivity	Export

B. Modelling of Langelier Saturation Index Calculator

Langelier Saturation Index Calculator has been developed with two different approaches. In both cases the calculation of LSI from pHs follows eq. (1) considering alkalinity, total dissolved solids, pH, temperature and calcium hardness. The first method (LSI-1 calculator) is based on a simpler relationship to calculate pHs than the one used in LSI-2 calculator, where all the aspect that influences pHs estimation are considered in detail.

Each model uses temperature, pH and ions concentration [mg/mg] as import variables from water stream. Table V lists all common import variables. Total dissolved solids are evaluated as the sum of all cations and anions referring to any minerals, salts, metals dissolved in water [20] and it is compiled in FORTRAN as follows:

```
TDS = (NA+CL+CLO+NO3+F+SO4+ZN+CA+CO3+MG+CAOH+
      HCO++HSO4+ZNOH4+ZNOH3+FE+AL+K)*((10**3)*DENS)
```

with 10**3 the conversion factor

1000·(*mg/kg)/(dm3/m3*).

Alkalinity is calculated as the sum of bicarbonates and carbonates equivalents and it estimates the trend of water to neutralize acids as explained in [21] and it is inserted in mg/L in the model as follows:

```
ALC = (HCO3*((10**3)*DENS)/61+CO3*
*((10**3)*DENS)/30)*50
```

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with 10**3 the conversion factor 1000·(mg/kg)/(dm3/m3):

61 the HCO_3^- equivalent weight,

30 the CO_3^{2-} equivalent weight and
50 the CaCO3 equivalent weight.

TABLE V. LSI MODELS COMMON IMPORT VARIABLES

ID	Unit	Variable
T	K	Solution temperature
PHO		Solution pH
DENS	kg/m^3	Solution density
NA	mg/mg	Na^+ Ion mass fraction
CL	mg/mg	Cl^- Ion mass fraction
CLO	mg/mg	ClO^- Ion mass fraction
NO3	mg/mg	NO_3^- Ion mass fraction
F	mg/mg	F^- Ion mass fraction
SO4	mg/mg	SO_4^{2-} Ion mass fraction
ZN	mg/mg	Zn^{2+} Ion mass fraction
CA	mg/mg	Ca^{2+} Ion mass fraction
CO3	mg/mg	CO_3^{2-} Ion mass fraction
MG	mg/mg	Mg^{2+} Ion mass fraction
CAOH	mg/mg	$CaOH^+$ Ion mass fraction
HCO3	mg/mg	HCO_3^- Ion mass fraction
HSO4	mg/mg	HSO_4^- Ion mass fraction
ZNOH4	mg/mg	$ZN(OH)_4^{2-}$ Ion mass fraction
ZNOH3	mg/mg	$ZN(OH)_3^-$ Ion mass fraction
FE	mg/mg	Fe^{3+} Ion mass fraction
CACO3	mg/mg	$CaCO_3$ mass fraction
CAF2	mg/mg	CaF_2 mass fraction
CACL2	mg/mg	$CaCl_2$ mass fraction
CANO32	mg/mg	$Ca(NO_3)_2$ mass fraction
CASO4	mg/mg	$CaSO_4$ mass fraction
CAOH2	mg/mg	$CaOH_2$ mass fraction

Calcium hardness is determined by the concentration of calcium ion in water normally related to the dissociation of calcium sulphates, nitrates and chlorides.

Total dissolved solids, alkalinity and calcium hardness are considered in both model as "auxiliary" export variable (Tab. VI). 1) LSI-1 Calculator: the method employed to develop LSI-1 calculator evaluates pHs as (4) [22].

$$pH_s = (9.3 + A + B) - (C + D) \quad (4)$$

where A is the TDS (in *mg/L*) index, B is the absolute temperature T correction, C is linked to calcium hardness (*mg/L* Ca^{2+} as *CaCO3*) and D is the alkalinity (Alk as mg/L in CaCO3) index.

TABLE VI COMMON "AUXILIARY" EXPORT VARIABLE.

ID	Unit	Variable
TDS	mg/L	Total Dissolved Solids
ALC	mg/L in $CaCO_3$	Alkalinity
CAM (LSI-1)	mg/L in $CaCO_3$	Calcium Hardness
CAM (LSI-2)	mol/L in $CaCO_3$	Calcium Hardness

The above listed quantities are evaluated as follows:

$$A = (\log(TDS) - 1)/10 \quad (5)$$

$$B = -13.12 \cdot \log(T) + 34.55 \quad (6)$$

$$C = \log(CalciumHardness) - 0.4 \quad (7)$$

$$D = \log(Alk) \quad (8)$$

In LSI-1 Calculator block A, B, C, D and pH_S are considered "auxiliary" export variables and the only real export one is the Langelier Saturation index (Tab. VII).

2) *LSI-2 Calculator*: LSI-2 calculator exploits the following more complex equation to obtain pH_S according to [23]:

$$pH_s = pK_2' + pCa - pK_s' - \log(2 \cdot (Alk)) - \log(\gamma_m) \quad (9)$$

where *pCa* value is the negative common logarithm of the concentration of Ca^{2+} in *mol/L*.

Alkalinity (Alk) is reported as *mol/L* of *CaCO3*.

pK'_2 and pK'_S are the negative common logarithm of the activity constants K'_2 and K'_S, which are expressed as:

$$pK_2' = -\log\left(\frac{10^{-(2902.39/T + 0.02379 \cdot T - 6.498)}}{\gamma_D}\right) \quad (10)$$

$$pK_s' = -\log\left(\frac{10^{-(0.01183 \cdot t + 8.03)}}{\gamma_D^2}\right) \quad (11)$$

respectively with temperature *T* in *K* and *t* in °C.

Values of γm and γD are respectively the activity coefficient of monovalent and divalent ions, determined using the following Davies relationship:

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$$I < 0.5M \Rightarrow \gamma_i = 10^{-(1.82\cdot10^6\cdot(78.3\cdot T)^{-3/2})\cdot Z^2\cdot(\frac{\sqrt{I}}{1+\sqrt{I}}-0.2\cdot I)} \quad (12)$$

TABLE VII. "AUXILIARY" AND EXPORT VARIABLES IN LSI-1 CALCULATOR

ID	Unit	Variable	Type
A	–	TDS Index	Auxiliary
B	–	T Correction	Auxiliary
C	–	Calcium Hardness Index	Auxiliary
D	–	Alkalinity Index	Auxiliary
PHS	–	$CaCO_3$ saturation pH	Auxiliary
LI	–	Langelier Saturation Index	Export

$$0.5M < I < 1M \Rightarrow \gamma_i = 10^{-(1.82\cdot10^6\cdot(78.3\cdot T)^{-3/2})\cdot Z^2\cdot(\frac{\sqrt{I}}{1+\sqrt{I}})} \quad (13)$$

where Z represents the oxidation number of the chemical species (Z = 1 for monovalent ions and Z = 2 for divalent ions) and I the Ionic Strength of the water in *mol/L* (as in (14) with TDS in *mg/L*).

$$I = 2.5 \cdot 10^{-5} \cdot (TDS) \quad (14)$$

In LSI-2 Calculator block all the previous variables are considered as "auxiliary" export variables and are obtained using other supporting variables. The only real export variable is the Langelier Saturation index. The following table (Tab. VIII) lists these variables.

Some examples of applications of the developed water properties calculators are provided in the next section.

TABLE VIII "AUXILIARY" AND EXPORT VARIABLES IN LSI-2 CALCULATOR

ID	Unit	Variable	Type
IS	mol/L	Ionic Strenght	Auxiliary
ALCM	mol/L	Alkalinity (as $CaCO_3$)	Auxiliary
LGM	–	Log of γ_m	Auxiliary
LGD	–	Log of γ_D	Auxiliary
PK2P	–	$-Log$ of K_2'	Auxiliary
PKSP	–	$-Log$ of K_s'	Auxiliary
PCA	–	$-Log$ of Calcium Hardness	Auxiliary
LALC	–	Log of 2·Alkalinity	Auxiliary
PHS	–	$CaCO_3$ saturation pH	Auxiliary
LI	–	Langelier Saturation Index	Export

V. RESULTS

Validation of the calculator models has been carried out simulating different water streams and evaluating electrical conductivity and Langelier saturation index. The analyzed streams are listed below:

- fresh water A used in a steelmaking plant (FWA);
- fresh water B used in a steelmaking plant (FWB);
- process water of a steelmaking plant (PW);
- blowdown stream of a steelmaking plant (BW);
- sea water (SW) [18]; • tap water (TW) [18].

Water streams modelling has been carried out using laboratory or literature data about stream composition [18]. Results of EC and LSI calculators have been compared with laboratory data, literature informations or open source web calculator (OSC) results [18].

Table IX shows the following simulation data: absolute temperature T (K), water pH, TDS (*mg/L*), alkalinity Alk (*mg/L* as *CaCO3*), calcium hardness (*mg/L* as *CaCO3*).

TABLE IX SIMULATION DATA

	FWA	FWB	PW	BW	SW	TW
T	300	300	300	300	293	293
pH	8.09	7.91	9.60	9.23	8.00	8.60
TDS	229	16.40	22.45	18.47	34483	273
Alk	76	80	5	3	260	46
Ca hard	251	431	313	431	998	122

EC (in *µS/cm*) data coming from laboratory tests (ECLab) and result values as output of developed calculator (ECCalc) are compared in table X and in figure 2.

The EC calculator and laboratory values are similar; in the FWA case the error is higher than 5.5% but EC absolute value is comparable with laboratory data, which is related to water content of salts. The model reliability is proved. On the other hand, table XI shows pH_S and *LSI* results and figure 3 compares them: pH_S -1 and LSI-1 and pH_S -2 and LSI-2 are developed calculators results and pH_S-OSC and LSI-OSC derive from an open source web calculator [18]. Due to the qualitative nature of LSI, which gives an idea of water quality, it is possible to notice that model results are comparable. In most cases, each water sample falls within the same range of LSI (water quality) using the three different considered calculators: model validation is possible. Gaps between absolute values can be justified by the lower accuracy of some used relationship or by lack of information about water modelled streams. In particular, as expected, method LSI-1 responds more rapidly due to reduced computational cost but with a higher error on the results.

TABLE X. ELECTRICAL CONDUCTIVITY RESULTS

	FWA	FWB	PW	BW	SW	TW
EC-Lab	330	3400	4500	4000	–	–
EC-Calc	387	3362	4736	4010	72992	307
Error [%]	17.27	1.12	5.24	0.25	–	–

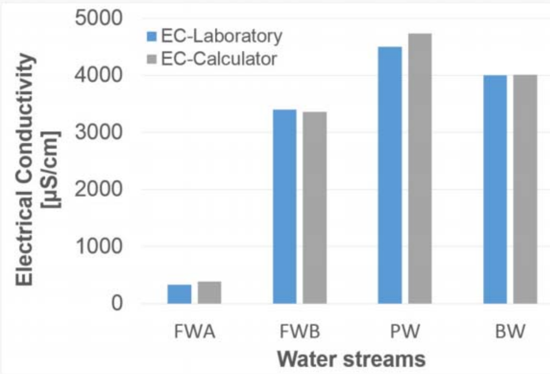

Figure 2. Electrical conductivity results comparison.

TABLE XI. pH_S AND LSI RESULTS

	FWA	FWB	PW	BW	SW	TW
pH_s-1	7.60	7.43	8.81	8.89	7.06	8.21
pH_s-2	7.56	7.53	8.94	9.00	7.89	8.17
pH_s-OSC [18]	7.60	7.60	9.20	9.10	8.00	7.90
LSI-1	0.49	0.48	0.79	0.34	0.98	0.41
LSI-2	0.53	0.38	0.66	0.23	0.15	0.68
LSI-OSC [18]	0.53	0.36	0.41	0.10	0.05	0.68

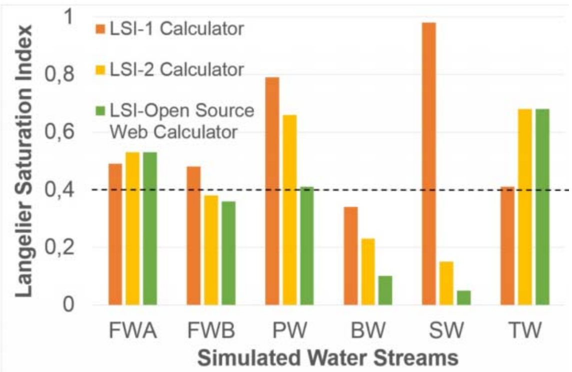

Figure 3. Langelier Saturation Index results comparison.

VI. CONCLUSION

In the current paper, the possibility of estimating and monitoring electrical conductivity and Langelier Saturation Index in Aspen Plus through FORTRAN based calculators has been presented. Two LSI calculators based on different relationship in terms of accuracy have been described. Both EC and LSI calculators have been validated. Their implementation allows to control water contamination and its features in a complex process simulation, giving important informations that can support plant revamping or maintenance choice. For this reason, the realized water properties calculator models turn out to be important instruments in many process simulation studies.

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