SODIUM CONTENT IN ALUMINUM AND CURRENT EFFICIENCY – CORRELATION THROUGH MULTIVARIATE ANALYSIS

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Abstract

Current efficiency is an important indicator used in the aluminum reduction technology. Values for this indicator are usually determined among potlines and they are not representative of the fluctuations that may occur in a single electrolysis cell. To measure or calculate an accurate value on a monthly basis would be a very interesting tool for process technicians and engineers to help regulate and analyse the performance of the pot. The potential use of the sodium content of aluminum as an indicator of current efficiency is investigated. Many authors discussed its role and indicated a possible correlation with the current efficiency. Aluminerie Alouette Inc. performed some univariate statistical analysis to confirm this correlation on a potline scale. Furthermore, multivariate analysis is performed to strengthen the correlation according to other indicators. Results from these analyses and the possible implementation as an indicator is discussed in this paper.

Introduction

Aluminum is produced during electrolysis of alumina dissolved in cryolite-based melts. This process requires the use of high currents to maximize production. The performances are often compared by following the energy efficiency (kWh/kg Al) of the selected cell, potline or smelter. However, the energy efficiency is dependent on the current efficiency directly related to Faraday's law. The current efficiency is defined as the ratio between the metal produced and the theoretical production as shown in eq.1 for a 365 kA electrolysis cell. The current efficiency (CE%) for different smelters generally fluctuates between 85% and 96% [1].

$$CE\% = \left(\frac{\left(\frac{kg \ Al \ produced}{kA \cdot hr \ of \ current \ passed}\right)}{\left(\frac{2939.345 \ kg}{8760 \ kA \cdot h}\right)}\right) \cdot 100 \tag{1}$$

This indicator is commonly used in aluminum smelters. However, it can be very difficult to correctly identify the mass of aluminum produced on a daily basis for a particular cell. Fredrickson [2, 3] evaluated the statistical error from the CE% calculations based on the various smelter's technology and with consideration to the analytical measurement used for the calculation. The statistical error has been calculated for many cases of calculation of the CE%. The most interesting results for a single cell were calculated using: a) the aluminum production corresponding to 100 tapping cycles, b) a tracer metal dilution calculated on 48 hours, c) a tracer metal dilution on 48 hours with samplings every 2 hours, d) the CO/CO₂ ratio from exhaust gases

analysis. The estimated errors¹ were a) CE% ± 0.66 , b) CE% ± 3.79 , c) CE% ± 1.97 and d) CE% ± 4.91 . An important error is always present when calculating the CE% for a single cell. The most precise calculations require 100 tap cycles (125 days). This is considerably long.

In 1995-1996, Tabereaux [4, 5] postulated that the sodium concentration in the metal pad $(Na_{(Ab)})$ is directly related to the movement at the bath-metal interface. Polyakov et al.[6] indicated that the behavior at this interface is strongly related to the CE%. Therefore, Tabereaux showed that there is a strong relation between the sodium content of the aluminum and CE%. This relation has also been observed by other authors [7-9]. Othman and Ali [10] published a paper with conflicting results. However, the cells considered in their experiments had different excess %AlF₃. Consequently the relationship between sodium and CE% could not be clearly defined.

Some authors [4, 5, 7-9, 11-13] investigated on variation of the sodium content in the aluminum based from a theoretical aspect. During electrolysis, the transport of electrical charges in the electrolyte is mainly accomplished by the movement of the Na⁺ ions migrating towards the bath-metal interface. At the aluminum cathode, Na⁺ ions react with the fluoride complexes anions resulting from the cathodic reaction (eq.2), which increase the NaF ratio at the bath-metal interface.

$$AlF_4^- + 3 \cdot e^- \to Al + 4 \cdot F^- \tag{2}$$



Distance from Al cathode

Fig. 1: Sketch of the different concentrations next to the bathmetal interface. [12]

The increase of the cryolitic ratio, CR (fig. 1) causes a shift in the equilibrium at the bath-metal interface resulting in a higher content of sodium in the aluminum according to eq. 3. [5]

¹ These results are for a cell technology similar to the one analyzed further in this paper.

$$3 Na_{(Al)} + AlF_4^- \leftrightarrow 3Na^+ + 4F^- + Al \tag{3}$$

Alternatively, Solheim [8] studied a suggestion made by Sterten et al.[14] that the change in NaF/AlF₃ ratio increases the liquidus temperature, causing a solid cryolite precipitation at the bath-metal interface. Solheim examined this possibility using a 1D finite element method for the four main components of the bulk, hereby NaF, AlF₃, CaF₂ and Al₂O₃ and found that it is possible for the conditions to create such a precipitation. Kent [15] observed that a ring of solid material was formed on the measurement rod at the bath-metal interface height when measuring highly stable pots; this being the only published piece of evidence that this phenomena may occur in industrial cells.

Independently of the mechanism dictating its migration, the sodium content is highly dependent on the bath-metal interface stability. Therefore, it is postulated in this paper that the variables having an impact on the current efficiency will affect the sodium accordingly. With consideration of the theoretical explanation for this phenomenon, we suppose that current efficiency and sodium content of the aluminum should be strongly related to the cell's bath-metal interface stability.

The purpose of the present study is to examine which measured cell variables permit to establish potential links between the Na-content of the metal and the current efficiency of a cell. Univariate and multivariate analyses were performed on the measured cell variables at Aluminerie Alouette Inc. (AAI). Table I lists some of the variables included in this study that were previously discussed in the literature.

Table I: Published parameters having a correlation² with sodium and/or current efficiency.*

Sodium in the Al	Current efficiency
Temperature [7, 16]	Temperature [17-19]
Current density [7, 11, 12]	Current density [18]
Bath ratio (% AlF ₃) [5, 7, 16]	<u>Bath ratio (% AlF₃) [5, 18, 19]</u>
Pot's noise [5, 7]	<u>Pot's noise</u> [17, 18]
<u>% Alumina</u> [7, 9]	<u>% Alumina [18, 19]</u>
Lithium content** [9, 11]	Anodic incidents [17, 20]
	Anode-cathode distance[18]
	Metal height [18, 21]
	Power modulation [18]
	Impurities (P,Fe,Si,)[22, 23]

*Underlined parameters are common to both groups.

** LiF is not used at AAI and will not be considered in this paper.

In this paper, we attempt to correlates the parameters from Table I with data collected from the potrooms. Discussions are focused on results agreeing or in conflict with the publications as well as other parameters briefly discussed in the literature.

Smelter - Monthly Correlations

Data Collection and Analysis Procedure

In order to accomplish the proposed analysis without errors inherent to the data collection, it was necessary to collect the

monthly data for the overall smelter performances. Moreover, in order to minimize the errors from the correlations, the collection of data points must cover an extensive time period. A period of 60 consecutive months was selected, from 2007 to 2011.

To minimize the error related to the calculations of the current efficiency as observed previously [2], data according to the metal produced were measured directly at the cast house. It includes the solid metal collected at the crucible treatment center and the aluminum siphoned when cryolite is tapped. The current efficiency calculation is based on the total number of pots in operation, the total mass of metal cast and the average intensity of the current for the corresponding months.

Other parameters were collected with the control system either continuously (avg. value per day) or with the weekly routine of data collection (2-6 values per weeks). From these, an average value or the total sum was calculated for the smelter based on every value collected from the cells in operation for every corresponding month.

The correlation analysis was performed using STATISTICA [24]. A correlation matrix was computed using the collected data from the plant. The analysis takes into account every parameters considered and assumes a linear correlation between them. Results regarding the strength of the correlations between the different parameters are illustrated as a matrix to ease the comparison. For this analysis, correlations in regard to current efficiency and sodium content will be discussed.

Results and Discussion

Results from the analysis are presented in Table II. The parameters in the Table were selected either from Table I or added because of their high correlation with CE% and/or the sodium dissolved in Al. Furthermore, each correlation will be discussed one at a time with comparison to the published results. Before entering into an exhausting analysis, it was necessary to determine if a correlation between the sodium content and the current efficiency was present. Results from Fig.2 compare the correlation observed at AAI with the one observed by Tabereaux [5] in 1996.



Fig.2: Relationship between the sodium content of the metal and the current efficiency as observed at AAI and by Tabereaux in industrial cells.

 $^{^2}$ The term correlation used through the text refers to the strength of the relation between two variables. Values for these relations are expressed as correlation coefficient. These values correspond to the square root of the R² generally used in linear correlations tools.

Results from Tabereaux strongly correlate with a logarithmic curve. From the data collected at AAI, the correlation coefficient was similar when using a linear or logarithmic regression. The logarithmic curve shown on Figure 2 has been chosen for an easier comparison with the results of Tabereaux. Moreover, a previous study at Aluminerie Alouette [25] showed an asymptotic behavior close to 30 ppm of sodium, reinforcing the logarithmic pattern. The correlation of this relationship is however only half as strong as the one observed by Tabereaux. Coursol et al.[26] described Aluminerie Alouette Inc. as a smelter working at a low anode-cathode distance (ACD) close to the critical ACD where CE% would drop rapidly. This critical ACD is not yet reached, but the actual ACD does slightly lower the CE%. Therefore, the range of values for the current efficiency is narrower than the one observed by Tabereaux, so small deviations will have a greater impact on it. On the other hand, the premise from the introduction could explain the difference observed. In fact, if the sodium content of the metal and CE% are related not directly but indirectly through other parameters, the impact of each factor can be of different proportions. It is unlikely that the cells used in this correlation have similar operating conditions as the ones used in the study of Tabereaux [5].

Table II: Goodness of the correlation for 16 variables with regard to current efficiency and sodium content in molten aluminum.

	Variables	Correlation coefficient* with	
	, and out	CE%	Na _(Al)
ves	Excess AlF ₃ in bath	0.12	-0.26
	Daily doses of AlF ₃	-0.24	-0.46
	% CaF ₂ in bath	-0.27	-0.52
liti	Bath depth	0.34	0.46
٧dc	Added liquid bath	0.33	0.73
4	\checkmark Daily doses of Al ₂ O ₃	-0.18	-0.40
	Number of point feeder's action	0.14	0.66
gy	Current intensity	-0.59	-0.71
	Cell's resistivity	0.61	0.68
er	Cell's voltage	0.57	0.59
Ш	Bath temperature	-0.04	0.32
	Silicon content in molten Al	-0.43	-0.66
thers	Noise	-0.26	-0.20
	Total number of anodic incidents	-0.42	-0.25
	Iron content in molten Al	-0.43	-0.66
0	Total number of anode effect	-0.08	0.25

*Values in bold are within a 95% confidence margin according to the number of points considered [27]. Values are listed between -1 and 1. Extremums correspond to a perfect correlation and 0 is equivalent to no correlation.

Additives Dissolved in Molten Bath

An increase in the fluoride content showed no interesting results from a statistical point of view even though theory indicated that a relation existed with the CE% and the $Na_{(AI)}$. The divergent results are explained by the daily variations of the cells. Only 20% of the pots are analyzed daily for the bath ratio. This low number can hardly correlate with 100% of the CE% value as it does not consider the daily variation of the remaining pots. Results according to the total number of daily doses are recorded daily and a weak correlation can be observed. A negative correlation with CE% and sodium dissolved in aluminum is

related to the reactions of the fluoride. When more AlF_3 is dosed to the cell, there is an increase in the superheat of the cell causing the sidewall to melt. This melting causes the metal pad to expand more in the cell to fill the melted ledge volume. As a consequence, it lowers the metal height, hence causing less metal to be tapped and affecting the calculation for current efficiency.

In opposite to the theoretical work of Thonstad et al.[7], a decreasing correlation has been observed between the CaF_2 and sodium content in molten aluminum. By examining carefully the correlation matrix, the relationship between CaF_2 and the bath height is also weakening. Therefore, it is highly plausible that the correlation between calcium fluoride and sodium is indirect and both parameters are related to the bath level in the cell.

The bath height correlation has an importance on the sodium content because it is an indicator of the total mass of cryolite in the cell. Supposing the ledge thickness is constant, a higher bath value would indicate more cryolite present in the cell. It is supposed that by increasing the total mass of bath, the magneto hydrodynamic movements of the metal are diminished due to an increase in pressure resulting from the extra bath mass to support. Results from Kurenkov et al.[21] indicated that the depth ratio (metal/bath) had an importance on the stability. It was indicated that higher bath could lead to higher stability of the metal pad. This change will increase the CE% and facilitates the sodium transfer towards the aluminum. When liquid bath (200 to 500 kg) is added to the electrolysis cell, a similar correlation can be observed with CE%. Moreover, a strong correlation with the sodium content of the aluminum is observed. It may be explained by the mixing that occurs between aluminum and molten bath when cryolite is poured in the cell. Before a new equilibrium is reached in the pot, a small fraction of the cryolite may react according to eq.4 while it is still dissolved in the aluminum metal pad. If only 1 kg of bath reacts with the aluminum according to this reaction. An increase of nearly 30 ppm could be observed in the sodium content of the aluminum. Experimental studies are necessary to confirm if the reaction occurs in such conditions.

$$Al_{(l)} + Na_3AlF_{6(s)} = 2AlF_{3(s)} + 3Na_{(Al)}$$
(4)

Thonstad et al.[7] indicated that the alumina concentration had no important relation with the sodium and Haupin [9] quantified it to be very small, but negative. Our results indicate that more doses of alumina negatively correlate with the sodium level in aluminum. Finally, when the point feeders are activated, they push the alumina more rapidly into the cell than a normal dosage by gravity. When dosed too fast, part of the alumina can accumulate at the bottom of the metal pad as sludge. The Na₂O content of this sludge can react with the aluminum to form alumina and dissolved sodium in the metal pad according to eq. 5. This could explain the strong correlation observed between the number of point feeder's action and the sodium content of the metal.

$$2Al_{(l)} + 3Na_2O_{(s)} = Al_2O_{3(s)} + 6Na_{(Al)}$$
(5)

Energy Transfer in the Electrolysis Cell

Increasing the line current intensity has a direct impact on the cathode current density. Theory[12] indicates that a positive correlation should be observed by increasing the current density. However, a strong and negative correlation has been observed from the smelter's results. As mentioned previously, the ACD is very small at AAI. Therefore, a small increase in intensity

obtained by diminishing the ACD will have a more important impact on the bath-metal interface stability than it would have at a higher ACD. This difference will cause the negative correlation to overcome the positive increase we could observe in both cases.

The cell voltage and cell resistance are highly linked to each other in an electrolysis cell because the intensity is maintained on a target. A strong positive correlation can be observed for the current efficiency and the sodium content of the aluminum. If these parameters are at higher values, it is plausible that the ACD is larger, causing a reverse effect that one observes when increasing current intensity. On the other hand, with this increase, the chances for eq. 6 to happen, as described by Welch and Tabereaux[28], are higher and would favor the sodium dissolution.

$$Na^+ + e^- = Na_{sat}(Al) \tag{6}$$

In opposite to results from Tarcy and Sorensen [17], the bath temperature had no impact on the current efficiency. It is supposed that temperature variations between the cells caused the average temperature to be non-representative. A weak correlation with the sodium content can be observed but the statistical analysis is not sufficient to explain this correlation.

A negative correlation has been observed for the silicon content in aluminum. The CE% correlation is easily explained as silicon is directly linked to the cell power. The cell power can easily be unbalanced when a lot of reoxydation occurs. This phenomenon generates heat in the cell and increases the superheat causing the sidewalls to melt. Hence, silicon is a consequence of a low CE% and not a cause. It is supposed that the strong correlation between silicon and sodium is indirectly related to the high instability at the bath-metal interface that would cause low current efficiency in the first place.

Other Indicators

The pot noise is an indicator of the variations in the cell resistance. An increase in the noise is generally due to strong fluid movement in the cells [29] or caused by an incorrect anode. Both of these phenomena should lower the CE% according to theory. However, our results did show a negative correlation but it is not as strong as one would expect. Dissolved sodium is also related to noise with a similar coefficient as the CE%. The increase in movements at the bath-metal interface and the impacts observed correlate with the hypothesis of this paper less than expected. To add precision, it would be necessary to identify the origin of the instabilities (metal pad or anode incidents) and analyze the results in two groups separately.

The total number of anodic incidents shows a stronger influence than noise on the current efficiency. Data collected at the Voerde smelter [20] indicate that spikes can contribute to lower CE% for as much as 1.6%. The relationship according to anodic incidents was present but unclear with the present analysis. Data from the analysis also included air burned anodes. These cases do not lower the current efficiency, therefore weakening the correlation. The correlation with the sodium is weak and negative. It is highly plausible that spikes absorb sodium from the aluminum when the carbon is dipped into the metal pad. Chemical analysis[30] from anode incidents taken at AAI showed an important increase in the sodium content of the anode. This high sodium concentration is specific to the first centimeters of carbon. This region has the highest probabilities to be in direct contact with the aluminum.

Iron content in metal usually comes from anode stubs which are exposed to molten cryolite.[31, 32] No correlation was expected nor observed with the sodium. However, as described by Sterten and Al.[23] from laboratory experiments, the iron had a negative correlation with the CE% (0.23 ± 0.04 % per 100 ppm Fe_(bath)). Results from AAI in production cells show a decrease of 0.17% per 100 ppm Fe_(Al) with regard to the CE% It is explained by Sterten and al. that the different states of Fe ions produce a loss in the efficiency by changing constantly.

No particular correlation is applicable between the total number of anode effects and the current efficiency. An overview of the data indicates that a small number of cells have contributed to a large number of anode effects. These few cells caused an important increase in the number of anode effects, but the real impact on CE% is diluted in the total number of cells considered. A small positive correlation is observable between the anode effects and the sodium content. When an anode effect occurs, the cell voltage will considerably increase. It is plausible that the reaction from the eq.6 occurs during this erratically high voltage. However, anode effects generally last less than a minute and the impact of this reaction should be less than what is observed.

Cell to Cell Performances - Multivariate Analysis

Data Collection and Analysis Procedure

Most algorithms used in multivariate analysis require an important number of values to correctly represent the system. In order to achieve a high number of values for the analysis, every electrolysis cell was considered, from January 2007 to November 2011. The data considered for the analysis were divided into periods of four months. This period length was chosen because it almost corresponds to 100 tap cycles. For this time step, and according to the results from Fredrickson [3], the accuracy on the calculation of the current efficiency is known to be a little higher than ± 0.66 %, for the calculated value. The remaining variables included results from the average value of the measurements attributed to this time period.

Calculations were effectuated using the "STATISTICA" software. A total of 40 predictors and over 8000 values were considered. The data used were filtered using Henry's chart to eliminate extremely out-of-range values and validate the normal distribution of the values. In some cases, data were transformed using a logarithmic function to approach a normal distribution. The analysis was divided in three studies:

- 1. Defining the most reliable predictors for CE% (excluding sodium dissolved in aluminum).
- 2. Defining the most reliable predictors for the sodium dissolved in aluminum
- 3. Developing a model based on the predictors that correlates with the observed value of current efficiency (including sodium dissolved in aluminum)

The values were computed using a boosting trees algorithm. This algorithm generates an important number of simple and poorly accurate decision trees. Then it uses the results from one tree as input for another. Different weight is assigned to every tree during iterations to increase the accuracy of the model. Elith and al. [33] described the importance of setting the input parameters and their

impact on the resulting model. Results from analysis 1 and 2 obtained using this algorithm, will be discussed simultaneously. Study #3 will be discussed according to the efficiency of the model and the possible implementation as a tool for aluminum production.

Reliable Predictors for CE% and Na(Al)

From the computed results, it was possible to determine which parameters have the most of influence on the dependent variables considered in this article. The 10 most important predictors are listed in Table III in decreasing order of importance.

Table III: Most important predictors for current efficiency and sodium content of the aluminum resulting from multivariate analysis

Current efficiency	Sodium _(Al)	
Silicon in Al	Year of measurements	
Line current	Line current	
Total bath transfer	Alumina dosage speed	
Daily dose of AlF3	Silicon in Al	
Target for cell resistivity	Target for cell resistivity	
Metal Height	CaF_2 content in bath	
Phosphorus in Al	Metal height	
Cell's days in operation	Number of point feeder action	
Cell's resistivity	Iron in aluminum	
Vanadium in Al	Cell's voltage	

Four of the ten strongest indicators were found to have an important impact on both the CE% and the $Na_{(AI)}$; line current, silicon in molten aluminum, metal height and the target for cell resistivity. Two of these have been discussed in the previous section of this paper. According to a model from Biedler and Banta [34], an increase in the metal depth tends to lower the bath ratio. This is because the surface area adjacent to the metal pad region increases, while the amount of heat available for dissipation remains the same. Therefore, a significant amount of bath is consumed to thicken the ledge in the metal pad region. The change in cryolitic ratio can explain the change observed towards the CE% and the Na_(AI).

The target for cell resistivity was not considered in the first section because of the strong correlation it had with the resistivity itself. Out of the remaining indicators, many were already discussed in the first section. However, it is interesting to notice that in some case, (e.g. iron in aluminum, total bath transfer) the correlations were not observed in univariate analysis. This can be attributed to non-univariate relations that were not considered in the first part of this paper. On the other hand, some relations observed previously are confirmed by the multivariate analysis, i.e. CaF_2 relation with sodium, number of point feeder's action.

Two of the remaining indicators (P & V dissolved in Al) have been previously studied by Sterten et al. [23] and many others [35-37]. The observation from this paper correlates with the literature.

The year of the measurements had a major impact on the sodium concentration in aluminum. An important part of this correlation is attributed to the increase in the potline current with the years.. However, it is supposed that some parameters, that were not included in the analysis, were more important than expected. Therefore, if important variations of these hidden parameters occurred during the different years, it would be in agreement with the observed results. It could be the case of the sodium oxide concentration in primary alumina and could be the reason why the alumina dosage speed (time in overfeed vs underfeed) have this much of an importance on the $Na_{(AI)}$ predictions.

The developed models that are based on the previous indicators had a correlation coefficient of 0.53 and 0.9 respectively to the CE% and the $Na_{(Al)}$ between the predicted and observed values.

Developing a Model that Correlates the Calculated Current Efficiency of a Cell to its Design and Performance Parameters.

Using a boosting tree analysis, the most efficient predictors provided by study #1 were considered. Moreover, the sodium dissolved in molten aluminum was included in the model predictors to identify its impact on the calculated CE% value. In this case, sodium is the second strongest predictor, closely following the cell's intensity. The obtained correlation between the predicted and observed values is illustrated in Figure 3. The correlation coefficient for this model is the same as study #1 : 0.53. The results indicate that Na_(Al) and CE% are evolving similarly with a change of parameters. However, no increase of the correlation has been observed when including Na_(Al). This indicates that the sodium content variation is strongly related to the same parameters that were used in the study #1. These results are in agreement with the premise of this paper.

Predicted vs. Observed values for current efficiency using multivariate analysis



Fig. 3: Predicted vs. observed values of the current efficiency, obtained with a multivariate model. (Full line represents the ideal model; the area between dotted lines represents the error attributed to the calculation of CE%)

The model developed is a very good start and indicates that it is possible to use multivariate analysis to approximate the current efficiency of a particular cell. The present results need to be optimized for the model to be used efficiently. To be optimal, the model should be able to adequately represent a time step of one month operation at the most. Moreover, the correlation coefficient needs to be increased so that the deviation is lower than the potential difference between two cells.

The following adjustments of the model will be examined in a future analysis to possibly improve its efficiency:

- To increase the impact of the sodium content of the aluminum, indicators from the study #2 need to be considered in the analysis.
- The sodium level could also be classified according to the level (low, medium, high). The use of categorical values instead of continuous ones could create an important difference in the results.
- The daily fluctuations in the sodium could be a predictor much more accurate about the day that passed than the current level.
- By grouping the electrolysis cell in small groups with similar current efficiency, it would be possible to decrease the error in the calculation of the current efficiency.

Conclusion

This paper describes the relation between the current efficiency and several performance parameters of an electrolysis cell. The correlation between these indicators and the sodium dissolved in aluminum is also investigated trough the performance of pot lines for five years of operation.

Results from the literature were investigated and many indicators related with the current efficiency were in strong agreement with the published results. However, bath temperature and AlF_3 content did not correlate as expected. Most results according to sodium were not in close agreement with the literature, e.g. temperature and noise. Strong correlations were found with the dosage rate of alumina and with the number of point feeder's action. Both the sodium and the current efficiency were strongly correlated with the silicon level in metal, the current intensity and cell's resistivity. These last two indicates that the anode-cathode distance probably has a strong influence on the results.

Finally, it was possible to develop a model with multivariate analysis that can approximately calculate the current efficiency of an electrolysis cell based on the performance results of four operating months. The use of the sodium indicates that this predictor is strongly correlated with the other variables used in the model, illustrating the premise of this paper. This model is still in development and needs to be optimized to be efficiently used as a production tool for follow-up of the cells.

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