

Article

Corrosion Effect in Carbon Steel: Process Modeling Using Fuzzy Logic Tools

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Abstract: Acid mine drainage (AMD), resulting from mining activities, poses a significant environmental concern. It adversely affects metallic materials, particularly carbon steel composites used in mining machinery and structures. Highly acidic and oxidizing compounds like sulfuric acid and ferric ions cause corrosion, iron oxide formation, and hydrogen gas release, which degrade carbon steel. AMD also alters the solvent's properties, dissolving heavy metals and contaminants, and intensifying the environmental impact of mining. A 30-week experiment immersed metal plates in AMD to study its effects. Weekly observations of the plates and solvent were made. The plate measurements and physicochemical data were analyzed using graphical–statistical analysis and fuzzy logic techniques to assess the data quality and identify errors. The results reveal consistent findings with prior studies, such as material degradation with weight loss and alterations in acid drainage media, including increased pH and total dissolved solids (TDS). These changes in the solvent characteristics stem from the dissolution of metal ions from corroded surfaces, reacting with the acid solution. Overall, this study discusses the effects of AMD (acid mine drainage) on metallic materials and emphasizes the significance of monitoring and reducing the environmental consequences of mining activities.

Keywords: corrosion; exposure time; carbon steel; acid mine drainage



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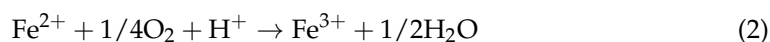
1. Introduction

The extraction of iron sulfides is a process that is common in all continents. It can be found throughout Europe, such as in the Iberian Pyrite Belt. However, it also exists in other places like South America, for example, in Peru (Antamina) and Chile (La Escondida mine), in Asia (Grasberg mine), in the USA (Morenci), in Canada (Abitibi area), and in Africa (Kansanshi).

Acid mine drainage (AMD) is formed in metal sulfides and coal mining operations when sulfides come into contact with oxygen and water, resulting in the release of hydrogen ions and a subsequent decrease in the pH of waters. Consequently, the resulting leachate exhibits extraordinary acidity, an elevated concentration of great metals, metalloids, rare earth elements (REEs), and sulfate concentrations in solution [1,2]. This process leads to the degradation of water environments, rendering them unsuitable for purposes other than mining. Also, as previously mentioned, the water becomes too acidic with high metal and sulfate [3–6] concentrations, occasionally reaching a negative pH [7]. AMD also represents the most severe and long-term environmental problem that is associated with metallic and non-metallic sulfide mining, for example, the case of arsenic, as identified by the authors

of [8]. So far, no uniform solution for AMD-affected systems has been found, so mining facilities and receiving rivers remain severely affected.

Younger et al. (2002) described the primary sulfide oxidation reactions [9]. The oxidation of sulfides under the action of water, oxygen, and microorganisms [10–15] are detailed in Equations (1) and (2) as follows [16]:



Apart from the myriad of environmental problems caused by AMD, water quality stands out as the most prominent exponent because it undergoes physicochemical changes, leading to extremely low pH values. However, additional concerns in terms of safety and economy persist. Machines, gear, structures, and equipment are exposed to these highly harsh ambiances and conditions, posing challenges to their durability, user safety, and repair costs.

In the scientific literature, many studies about the effects of saline water on metal alloys exist [17–19]. Although, the effect of AMD on machine components in a mining environment is an emerging area of work. This study examines the corrosion effects of AMD in steel, which is a recent topic because just few works on this topic exist, and in addition, it analyses the data using fuzzy logic, which is also new in this topic.

Mining equipment and machinery with a high metal content, whether internally (pumps, drills, screws, electrical engines, internal support structures, tracks, wagons) or externally (trucks, ore and wet classifiers, floated cells, etc.), are susceptible to high levels of wear caused by corrosion, with associated economic and safety costs [20–24]. Metallic materials deteriorate through a corrosion process of chemical and electrochemical reactions that can be accelerated depending on the environment to which they are exposed [25,26], which is evident when they are in a particularly aggressive environment such as AMD [27,28]. This process results in a loss of material and strength in metallic materials, reducing their performance and raising issues of security, stability, and economics, and underscoring the relevance of the analysis process. In mining, as in other industries, corrosion is a constraining obstacle due to the increase in mechanization, so the problem of corrosion is becoming worse. Therefore, it is crucial to understand the factors contributing to and causing corrosion, including elements that are part of AMD; these factors include low pH, microorganisms, dissolved metals and minerals, temperature, and dissolved oxygen. The presence of certain bacteria, such as *Acidithiobacillus ferrooxidans*, contribute to the increase in the acidity of mine water, with an acid formation rate that is greater than in the absence of bacteria, thus increasing the corrosion rate, particularly when the bacteria contact the surface of the metal.

Material selection is based on a range of chemical, physical, mechanical, and technical properties that meet the expected adequate value, safety, duration, and cost. For example, a material failure in a structure or machine can cause the rest of the system to go wrong or even break down, which leads not only to quality defects, but also to safety issues. In this case, the used material failure due to corrosion becomes an important factor to be considered.

An essential prerequisite for proposing measures to increase the durability of these materials is the successful analysis of the processes, reactions, and consequences that occur when alloys come into contact with these waters. This knowledge lays the foundation for opening dedicated production lines.

To complete this paper, an extensive bibliographic review was conducted, revealing that this area of the scientific literature is scarce.

The primary objective of this study is to analyze the corrosion progression of carbon steel plates exposed to the action of AMD in the laboratory. This work aims to establish a causal relationship between the changes observed over 30 consecutive weeks and the physicochemical reaction components that require resolution.

2. Materials and Methods

This section comprehensively describes the methods used to develop the experiments and describes the principles and tools used to analyze the data obtained by immersing the plates in solvents.

2.1. Development of the Experiment

Thirty “ $0.05 \times 0.06 \times 0.006$ m” carbon steel metal plates (% C < 0.25) were used for this experiment, the chemical compositions of which are given in % (material properties extracted from certification tests) as follows: C = 0.20; manganese = 0.49; Si = 0.21; S = 0.017; p = 0.011; copper = 0.22; chromium = 0.08; nickel = 0.11; C_{eq} = 0.32. We immersed the plates in the reagent solution, employing separate 0.8 L plastic containers. Plates were placed on a rounded plastic ring to guarantee the maximum amount of contact with the reagent solution.

The water used was sampled from a creek affected by AMD and taken to the laboratory in polypropylene containers and was protected from direct light in polypropylene bottles that were previously cleaned using diluted nitric acid. Then, we put the water in an open container and kept it in constant motion using a whisk. Submerged panels were taken every week and weighed, metered, and analyzed for solvent water. The obtained data were checked via Statgraphics Centurion, a powerful tool for exploratory data analysis, and then using fuzzy logic. The reaction solution was discharged from a stream affected by AMD located in the Iberian Pyrite Belt. The river receives wastewater polluted by the Tharsis Mine [29], and it is the iconic waterway of one of the world’s largest sulfide-forming provinces, with a mining history of more than 5000 years, and has not undergone any preventative or corrective measures [30]. The initial physicochemical properties of the reaction solution were pH 2.9, redox potential (Eh) 220 mV, total dissolved solids (TDS) 2.41 mg/L, and electrical conductivity (EC) 4.9 mS/cm. We left a 50 L stock of the same water in an open recipient and stirred it to replenish the reagent solution as it evaporated. Water was moved to the laboratory in airtight containers, protected from direct light in polypropylene bottles that were cleaned earlier using diluted nitric acid. In the laboratory, it was stored in an open container with a stirrer that kept it in constant motion.

On 30 January 2021, 30 plates were submerged in the reagent solution. The plates remained in contact with the solution for different durations, with the first plate being immersed for one week, and the last plate being immersed for a period of 30 weeks until 9 June 2021. Throughout the experiment, the plates were kept still.

Every withdrawn metal plate was washed and dried before being measured (weight, width, height, and thickness). Various physicochemical parameters of the reagent solution were measured, including pH, EC, TDS, Eh, and temperature (T). A HORIBA LAQUA PC-110-K multi-parameter meter was employed to measure and record these parameters.

2.2. Statistical Treatment

The results obtained from the measurements of the studied test tubes and the physicochemical data of the resulting reagent solution were compiled into matrices for further graphical–statistical treatment. The Statgraphics Centurion XVI version 16.2.04 software from Statgraphics Technologies, Inc. (The Plains, VA, USA, EE. UU.) was utilized for this analysis. It is a technique that is widely used in environments affected by mining activity, as reported in [31–34], and offers various features for exploratory data analysis, statistical summary, analysis of variance, statistical control, multivariate analysis, time series, etc., and allows for the different studied variables to be classified into categories or proximity ratios [35].

Descriptive statistics were applied to process and analyze the data for their subsequent treatment using fuzzy logic, ensuring their quality and identifying any possible errors in the methodology [36].

2.3. Data Mining and Fuzzy Logic

In the data analysis using fuzzy logic, the fuzzy stage gives the input data a degree of membership within the different possible expressions; for this, it searches for the correspondence between the variables' states, and the defined membership functions. Therefore, for a certain value of a variable, its degree of membership will be greater in one expression than in the others. Once the variables' states are expressed in linguistic form, logical relationships can be established between them, typically through rules such as if...then (IF—THEN). To do this, a series of relationships are defined that interpret the common sense and allow for the generation of a desired action (in a linguistic state). Subsequently, this must be translated into a number (defuzzification), so that the digital–analog converter converts it to a signal.

Fuzzy logic [37] allows for more complex and precise relationships to be established between variables, since it is not limited to a binary response (yes or no, 0 or 1, white or black). Fuzzy logic admits infinite intermediate values such as an infinite range of grays between white and black, which makes it possible to observe phenomena or interrelationships that are not detectable using classical statistics [38]. It is also more intuitive and easier to use.

Fuzzy logic can consider a continuous range of values for each variable and can establish dependency relationships within a discourse universe, which means that the relationships between variables can be analyzed based on the range of values allowed for each of them. In short, fuzzy logic offers a great potential for data analysis and allows for more precise and complex relationships to be established between variables.

The usefulness of this data analysis technique has already been corroborated in the field of acid mine water [8], as well as in various fields of engineering [39–42]. The usefulness and feasibility of this technique for data analysis lies in the complex analysis that is performed, as it can relate quantitative and qualitative variables in different units and perform an analysis very similar to that which a human being would perform after a long period of study. It is therefore possible to establish clear relationships that are also represented in graphical models. In this way, with a simple glance, and thanks to the graphic representation, it is possible to distinguish the possible existence of a relationship between an antecedent and an established consequent; this consequent is the main study variable that is conditioned by the different variables (antecedent).

MATLAB R2022b software [43] was used for the analysis of the data detailed earlier in this research. This mathematical software has sufficient capacity for the analysis of a large mass of data, as well as the possibility of implementing a fuzzy logic model that interprets the variables and establishes the cause–effect relationships between antecedents and consequents. In addition, the use of this software allows for very intuitive graphs to be obtained that relate the various variables to each other, as can be seen in Figure 1.

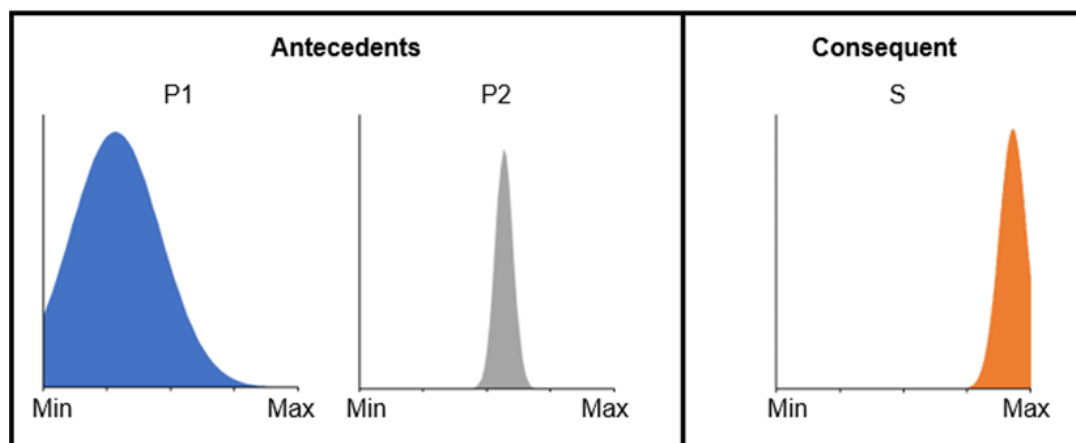


Figure 1. Fuzzy rules graphical representation.

Figure 1 shows a simple example that allows us to understand the relationships between variables that are generated using fuzzy logic data analysis. On the one hand, the figure shows two variables (P1 and P2) called antecedents and, in turn, a main variable called the consequent (S1). These variables also have certain values determined, in this case, between the maximum and the minimum, which is what is called the universe of discourse. In short, the universe of discourse comprises those values that can be obtained via the different variables between a maximum and a minimum. Therefore, in view of Figure 1, it can be seen how variable P1 must have low to intermediate–high values for variable S (Consequent) to obtain intermediate–high to high values. In turn, variable P2 must have values within the universe of discourse from intermediate to intermediate–high in order for the consequent, S, to obtain intermediate–high to high values.

Consequently, it can be seen how the example in Figure 1 shows the immense potential of fuzzy logic, since it does not detail a single value of the variables P1 and P2 to obtain high values of the consequent, S, but rather, details a range of values for each variable, which is something much more realistic than what is proposed by classical statistics.

Moreover, the use of Gaussian curves for the presentation of the different graphs associated with the various variables also provides very important additional information. On the one hand, it indicates which is the most usual mean value, i.e., the one corresponding to the crown of the Gaussian curve. On the other hand, it shows the dispersion of the results, i.e., the possibility of choosing different values for a variable within the universe of discourse. This fact is easily seen in Figure 1, by observing the difference between variable P1 and P2. Variable P1 has a much wider Gaussian curve that occupies a larger number of values within the universe of discourse, whereas P2 is a narrow curve that occupies a narrow interval within the universe of discourse. Consequently, it can be stated that there are more values within the universe of discourse of variable P1 that can lead to intermediate–high or high values of the consequent S than those of variable P2. Therefore, variable P2 is a variable that is highly dependent on the consequent S, since it must obtain very localized values within its universe of discourse in order to obtain intermediate–high or high values of the consequent.

In this research, after the analysis of the different data using fuzzy logic techniques, several graphs were obtained in which the different variables were represented as antecedents and the main consequent variables. The consequent variables selected for the study were weight loss and exposure time. The plates' weight loss and exposure time to the acidic medium are clear indicators of how the metallic elements are affected. Therefore, the interpretation of the results and the subsequent analysis was possible with a simple overview of the graphs and a detailed physicochemical interpretation of the results.

3. Results and Discussion

An introduction to the data and results of the experiment are shown in Table 1, through a statistical summary of the studied variables.

Table 1. Statistical summary.

	EC (mS/cm)	Eh (mV)	Exposure Time (days)	pH	Surface (cm ²)	Temperature (Celsius)	Total Dissolved Solids (mg/L)	Volume (cm ³)	Weight Loss (g)
Count	30	30	30	30	30	30	30	30	30
Mean	18.7	258	112	2.63	71.1	18.4	7564	16.3	14.79
Coefficient of variation (%)	63.1	21.2	56.8	15.6	2.62	22.7	56.9	7.30	73.7
Minimum	5.40	190	9	2.05	66.9	10.5	2600	14.1	1.98
Maximum	45.646	392	219	3.60	74.0	24.5	17100	17.9	37.0
Range	40.2	202	210	1.55	7.06	14.0	290	3.75	35.0

Figures 2 and 3 show the fuzzy rules, using the exposure time as a consequent, and using the other variables as antecedents.

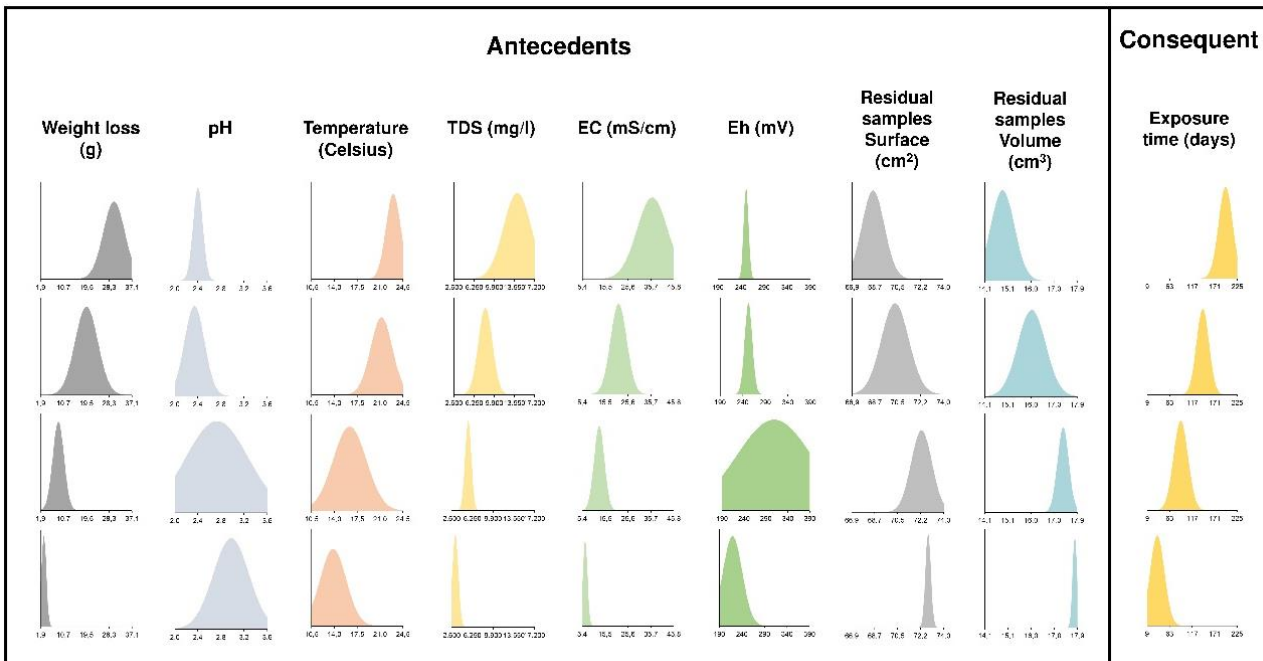


Figure 2. Fuzzy rule using exposure time as a consequent and the rest of the variables as antecedents.

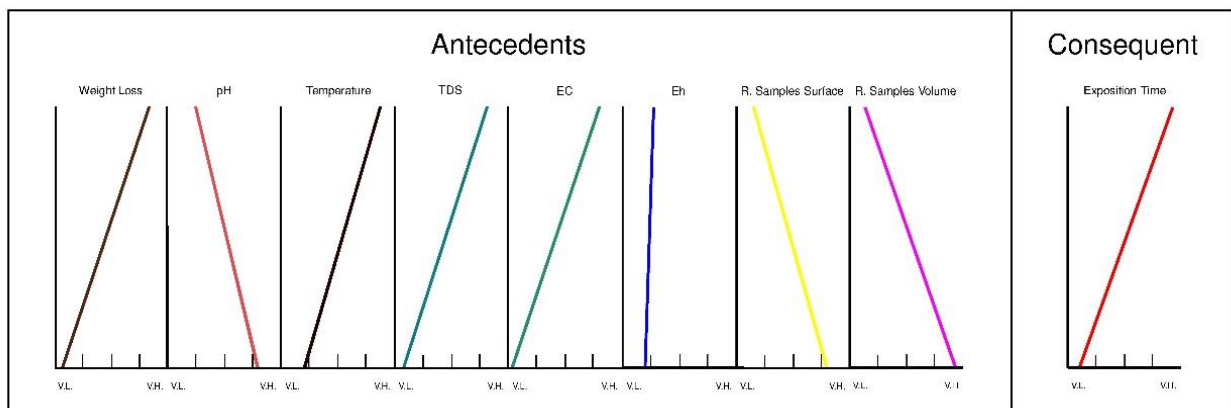


Figure 3. Simplification of the cause–effect relationships of antecedents and consequent represented in Figure 2, based on the trend line of the mean values for each variable.

On the one hand, we can observe that as the exposure time increases (Figure 2), there is an increase in the weight loss of the plates, reaching extreme low values when the exposure time varies between extremely low to low values, and taking values that range from high to extremely high when the exposure time takes the same range of values.

In contrast to what was previously described, it can be observed how both the residual surface area and the residual volume of the samples show a decrease in their values. Thus, the residual surface presents very specific high values when the exposure time takes values that range from extremely low to low, while the residual volume takes extremely high values that are also very defined, and for the high to extremely high values of the exposure time, both parameters take more scattered values from their discourse universe, ranging from low to medium.

Regarding the physicochemical characteristics of water, it can be seen that the pH presents an opposite behavior to the Eh, the TDS, conductivity, and temperature.

In this way, when the exposure time varies from extremely low to medium values, both the pH and the Eh show scattered values, being able to take any value in their discourse universe; however, as the exposure time increases and takes values that are medium to extremely high, the pH and Eh present more defined low values.

On the other hand, it can be observed that as the exposure time increases, the temperature, TDS, and conductivity also increase, presenting an identical behavior of the TDS and conductivity in all cases. Thus, for low to extremely low values of exposure time, the TDS and conductivity present very peculiar extremely low values, and present a broader discourse universe with values ranging from medium to extremely high when the exposure time takes values that range from high to extremely high. On the other hand, the temperature presents a greater amplitude of values from extremely low to medium when the exposure time is low–extremely low, while for high–extremely high values of exposure time, the temperature takes more specific high–extremely high values.

In turn, if we use weight loss as a consequent and the other variables as antecedents (Figures 4 and 5), it is possible to observe how the exposure time presents a similar behavior to this, while the residual surface and the residual volume of the samples exhibit a completely opposite behavior.

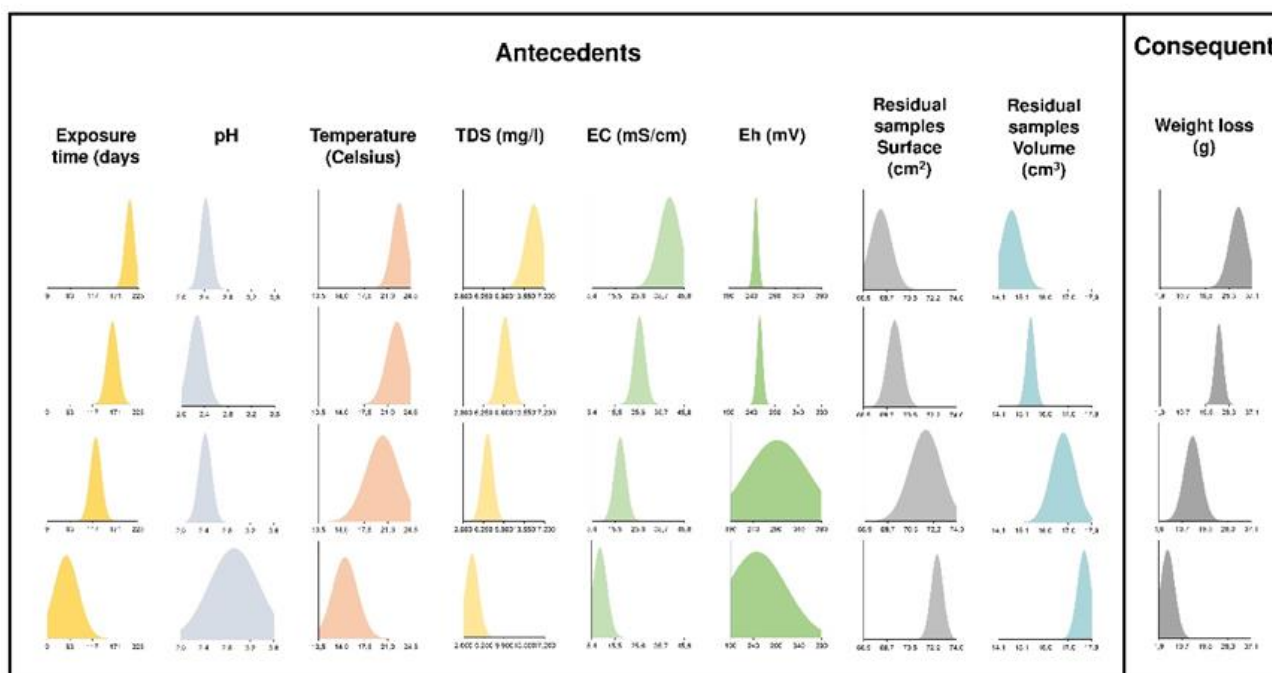


Figure 4. Fuzzy rule in which weight loss is used as a consequent and the rest of the variables are used as antecedents.

In this way, it is observed that as the weight loss increases from extremely low–low to high–extremely high values, the exposure time increases from extremely low–medium values to extremely high values, while the volume and residual surface decrease from high–extremely high values to extremely low–low values.

Taking into account the physicochemical parameters of the water, similar behaviors of the Eh, temperature, TDS, and conductivity are observed with respect to weight loss; however, the pH presents an opposite behavior to these last three.

Regarding the pH, it is also observed how it can take any value from its discourse universe for weight loss values that range from extremely low to low, showing more defined low–extremely low values as the weight loss increases from low to extremely high values.

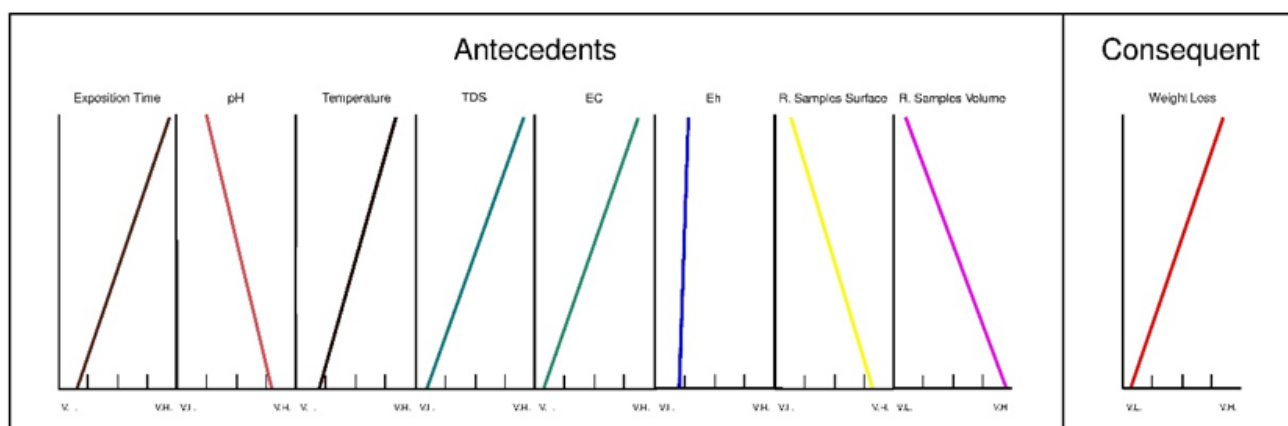


Figure 5. Simplification of antecedents and consequent cause–effect relationships represented in Figure 4, based on the trend line of the mean values.

As occurred in the previous fuzzy rules, the TDS and conductivity present an identical behavior among themselves and are very similar to weight loss, while the temperature presents a broader discourse universe, increasing their values in all cases in the same way that the weight loss does.

On the other side, it is observed how the Eh presents a more linear behavior, being able to take any value of its discourse universe, ranging from extremely low to medium weight loss values, and presenting very defined low values when the weight loss takes medium to extremely high values.

The fact that weight loss and a decrease in the volume of the plates were observed as time went by is something to be expected due to the mechanical abrasion processes that eliminate the oxide and sulfate layers formed during the process to continue with the Fe oxidation of the steel. Corrosion in steel has different natures depending on the environment, material, and exposure. In this case, generalized corrosion develops over the entire surface, resulting in the progressive steel oxidation and, consequently, the section reduction. This type of corrosion, which is typical in metallic materials exposed to very aggressive environments, is identified with a continuous penetration of oxidation into the material and its subsequent weakening, causing the reduction of the section.

Regarding the pH variable, the great negative influence of exposure time was observed; since the pH variable is a logarithmic expression that indicates the acidity degree of the water, the lowest pH value indicates a higher acidity of the water. However, the Eh has a moderate and positive influence due to the solubility of the metal in water, which decreases as the solvent loses its oxidation capacity.

The similar behavior between the TDS and the conductivity corroborates the one observed by classical statistics [44] and may be due to an increase in the Fe^{2+} ion in the water as the oxidation process of the plates progresses. The Fe in the plates is like $\text{Fe}(0)$, and when it oxidizes in this acidic medium, it initially becomes Fe^{2+} , which is soluble in an acidic pH. Subsequently, by hydrolysis, the Fe^{2+} becomes Fe^{3+} , which precipitates as ferric oxide, ferric hydroxide or ferric hydroxysulfate that are less soluble in an acidic pH than the previous ion.

This process is an exothermic reaction that provokes, over time, the water heating in which the immersion plates are submerged at the expense of their metallic corrosion, which may explain the time–temperature correlation.

4. Conclusions

The main objective of this work was to study the effect of acidic waters in the corrosion processes of carbon steel through the use of fuzzy logic tools, which allowed us to corroborate the conclusions obtained through classical statistics, as already verified in other

studies [45]. A novelty of this study is that it is the first time that fuzzy logic has been used to study the corrosion processes of steel, which showed to be an effective technique.

Thus, it was possible to demonstrate how acidic water can affect the corrosion of this material, one of the most used materials in the mining field, that can be found both in mining machinery and in the structural elements of mines, and it was possible to define the cause–effect relationships between the physicochemical variations of the water and the condition suffered by the metal plates submerged in it.

In this way, it can be concluded that acidic water significantly affects carbon steel, producing a decrease in its volume and weight.

It was observed how this corrosion process of the steel plates caused changes in the physicochemistry of the water and led to a notable increase in the TDS and conductivity, as well as a decrease in the pH due to the oxidation–reduction and precipitation–dissolution processes that occurred in the system. The Eh presented similar values to the pH due to the high saturation degree of the water.

The temperature increase throughout the experiment was due to the fact that the corrosion process is an exothermic reaction.

On the other hand, it was expected that the volume of the plates would decrease since the oxidation process causes a material loss.

Logically, all of this is closely linked to the exposure time, since the longer the immersion time of the plates, the greater the weight loss and the consequences described above.

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