A Machine Learning Approach for Predicting Hydrogen Embrittlement in Storage and Transportation Steels

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Abstract: Sectors related to energy constitute the predominant source of CO2 emissions due to a global dependence on fossil fuels. As a substitute to fossil fuels, hydrogen emerges as a cleaner energy source, offering easy transportability and diverse storage options (e.g., compression, metal hydrides). However, hydrogen embrittlement (HE) in steel becomes a significant concern, which can lead to the premature formation of cracks, and may result in a more severe structural failure. Ensuring effective and safe hydrogen storage and transportation technologies is essential for hydrogen distribution. Simultaneously, Machine Learning (ML) is a potent tool for predicting and identifying HE in diverse steel grades due to its ability to analyze complex and large datasets effectively. This study suggests utilizing ML classification models to analyze HE factors, including material composition, environmental conditions, and testing methods. The methodology involves database creation, labeling process, and preprocessing techniques for improving the performance of models. The AdaBoost classifier demonstrates high accuracy in classifying steel examples as susceptible or not to HE, reaching an accuracy of 93%, supported by preprocessing approaches. Metrics such as precision, recall, and confusion matrix are also analyzed in the study. This research contributes to the early detection of susceptibility in different steel grades, promoting the use of hydrogen as a cleaner energy.

Keywords: Hydrogen Embrittlement, Machine Learning, Storage, Steels.

1. INTRODUCTION

Hydrogen has emerged as a promising solution to the dual challenge of climate change and carbon emissions reduction, offering a clean and renewable energy source with diverse applications across industries, as shown by Zou et al. (2022). It presents easy transportability, including long-distance transportation, through pipelines or electrical transmission lines. Additionally, hydrogen is associated with a diverse range of storage options (e.g., compression, metal hydrides), which is observed in Sharma et al. (2021). In this sense, Ratnakar et al. (2021) highlighted the importance of the development of effective and safe technologies for storage and transportation of hydrogen.

The hydrogen supply chain, consisting of tank trucks, pipelines, and storage facilities, becomes essential for widespread adoption, according to Almansoori and Shah (2006). However, hydrogen embrittlement (HE) in steel is a concern, which can lead to the premature formation of cracks, especially during pipeline transportation, as noted in the study of Ilyushechkin et al. (2023). Laureys et al. (2022) demonstrated that HE occurs when hydrogen atoms diffuse into the lattice of materials, leading to a degradation of mechanical properties, increased susceptibility to cracking, and potential structural failure. Hydrogen-assisted fatigue crack growth is significant even at low hydrogen partial pressures, caused by fluctuations in gas pressure and applied loads. In this sense, Maior et al. (2022) emphasizes that the focus on risk analysis and reliability (e.g., the maintenance of equipment) is essential to ensure operational efficiency and prevent unexpected failures.

In recent years, advancements in materials science and Machine Learning (ML) have provided innovative solutions for addressing the challenges associated with HE, as evidenced by Nachtane et al. (2023). ML algorithms can recognize patterns, correlations, and hidden relationships within the data that might be challenging for the human mind to discern, as seen in the research of Fragassa et al. (2019). By understanding the mechanisms of hydrogen interaction with steels including material properties, environmental conditions, and performance characteristics, Barrera et al. (2018) analyzed that it is possible the development of predictive models capable of assessing the susceptibility of steels to embrittlement. This strategy can facilitate the identification of more efficient steels for the transportation and storage of hydrogen.

This study discusses the construction of a tool utilizing different ML classification models, such as extra trees, k-nearest neighbors, adaptative boosting, and categorical boosting, to analyze factors contributing to HE, providing precise predictions and enabling early detection of susceptibility in different steel grades. In addition, it discusses the contribution to the use of cleaner energy and promotion the transition to more sustainable practices, particularly in the context of hydrogen transportation and storage. The study is organized as follows: Section 2 examines recent scientific literature to provide a comprehensive review. Section 3 outlines the methodology utilized, encompassing database creation, establishment of target values, preprocessing techniques, and predictive models. Lastly, section 4 involves the results obtained in the study, while section 5 offers concluding observations.

2. LITERATURE REVIEW

Malitckii et al. (2020) combine hydrogen thermal desorption spectroscopy (TDS) and ML to quantitatively evaluate the susceptibility of steels and alloys to HE. They created a regression artificial neural network (ANN) to predict hydrogen-induced degradation of mechanical properties, using data augmentation to improve model generalization. Linear regression (LR) modeled the relationship between yield stress (YS), ultimate tensile strength (UTS), and hydrogen sensitivity parameter (HSP), achieving mean absolute errors (MAE) of 3.9% for YS and 5.5% for UTS. The ANN predicted HSP with an MAE of 1.4%. Similarly, Ahmed et al. (2024) used seven ML techniques, including random forest (RF) and categorical boosting (CatBoost), to predict HE based on the reduction of area in tensile tests of various low carbon and low alloy steels under pressurized hydrogen gas. The CatBoost model performed best, with low MAE and high coefficients of determination (R^2 of 77.62% for training and 72.50% for testing), identifying hydrogen gas pressure and UTS as key factors.

Kim et al. (2022) introduced an ML methodology to predict the Hydrogen Environment Embrittlement (HEE) index of austenitic steels by analyzing the relative reduction of area. They used Pearson's correlation coefficient and Maximum Information Coefficient to evaluate correlations between input features and the HEE index. They tested four ML models: RF, LR, Bayesian ridge, and support vector machine, finding that the RF model achieved the highest accuracy ($R^2 > 0.7$), significantly outperforming LR and Bayesian ridge models. Subedi et al. (2023b) also considered an index (i.e., Embrittlement Index) to a classification approach and to assess the safety of European natural gas pipelines for hydrogen transport using RF, AdaBoost, and ANN algorithms. Their study predicted material susceptibility to HE under different working conditions, aiming to prevent integrity loss and leaks. The RF algorithm correctly classified 84% of the evaluation database, while AdaBoost slightly outperformed it with 84.5% accuracy, and ANN achieved 77.5%. They ranked pipeline materials by their HE susceptibility, aiding hydrogen compatibility evaluations and strategies for integrity loss prevention.

3. METHODOLOGY

The methodology of the present study consists of database and target values creation, data preprocessing, modeling, and classification analysis. The methodology flowchart can be visualized in Figure 1.

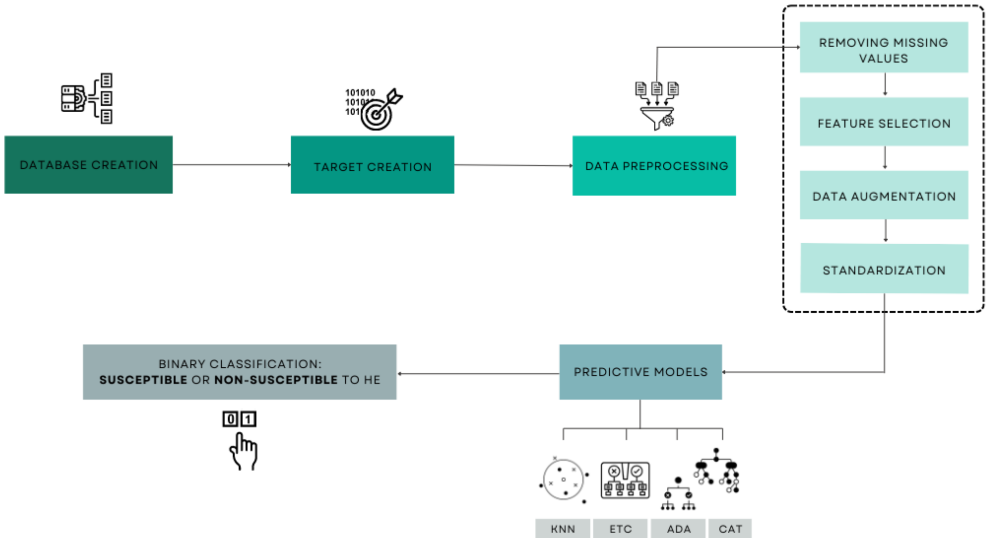

Figure 1. Methodology flowchart.

3.1. Database creation

Mechanical tests conducted on materials immersed in hydrogen environments were utilized for our research. For analysis, we considered deformation and fracture results from tensile tests using slow strain rate tensile tests (SSRT) with smooth specimens. The information from these tests was extracted from the "Technical Reference for Hydrogen Compatibility of Materials" by San Marchi and Somerday (2012), and the "Influence of Gaseous Hydrogen on Metals," the final report by Walter and Chandler (1973). Both reports are freely available and can be used to assess HE in materials.

From the reports, we extracted a dataset containing 137 data samples. This dataset includes 26 distinct features, which encompass various aspects such as chemical material composition (i.e., percentage of specific elements), environmental conditions (e.g., the pression and temperature of environment), mechanical properties (i.e., characteristics of the steel in response to a force or stress), and label classifying in susceptible or not to HE. Table 1 illustrates the format of the final database.

Table 1. Database overview

	ID	Gr.70; Gr. B; 1080; X42 ...	Environment		
Chemical components	Fe	66.54; 52.780; 99.240 ...		H2 pressure (MPa)	6.9; 69.00; 138; 172; 34.5 ...
	B	0; 0.002; 0.006; 0.005 ...			
	Ti	0; 1.930; 2.090; 2.100 ...			
	V	0.35; 0.350; 0.420; 0 ...		Temperature of thermal precharging (K)	295.15; 470; 620; 473 ...
	Nb	0.83; 0.190; 0; 0.230 ...			
	Sn	0.005; 0; 0.084 ...			
	Al	0.005; 0.005; 0.012 ...		Temperature of environment (K)	295.15; 375; 200 ...
	N	0.3; 0.170; 0.70; 0.270 ...			
	Cu	0.04; 0; 0.420; 0.300 ...	Mechanical properties	Strain rate (s^{-1})	0.0001; 0.00033; 0.00054 ...
	Ni	0.084; 0.300; 0; 0.040 ...			
	Co	4.450; 0 ...		Sy (MPa)	375; 364; 462; 414; 366 ...
	Cr	0.93; 1.670; 1.460; 0.540 ...			
	Mo	0.2; 0.420; 0.430; 1.010 ...		Su (MPa)	535; 566; 559; 814 ...
	C	0.21; 0.400; 0.160; 0.130 ...			
	Mn	1.04; 0.830; 0.320; 0.300 ...		RA (%)	69; 72; 58; 16; 14 ...
	P	0.009; 0.0016; 0.005 ...			
	S	0.020; 0.014; 0.019 …	Label		0; 1
	Si	0.21; 0.310; 0.020; 0.630 ...			

During the data curation process, certain missing values were addressed by applying standard assumptions to avoid data loss for temperature and strain rate. For instance, a room temperature of 295.15K (equivalent to 22ºC) was assumed, which is widely used for normal conditions, as seen in Maeda et al. (2012). Additionally, a strain rate of $0.0001 s^{-1}$ was utilized, as it serves as a nominal rate commonly employed in both experimental setups and modelling. This finding was shown by Jin et al. (2021).

3.2. Creation of target values

A method for quantifying HE in materials is the use of an embrittlement index (EI). This index measures the degree of material susceptibility to hydrogen exposure, as seen in Moro et al. (2010). The EI is expressed as a percentage ranging from 0 to 100. At 0%, hydrogen has no impact on the toughness of the material, and at 100%, HE completely reduces the steel toughness to zero, as mentioned by Álvarez et al. (2021).

In this study, the EI of each steel was determined by calculating the tensile properties loss according to the Equation (1):

$$EI = \frac{\delta_0 - \delta_H}{\delta_0} \times 100\% \tag{1}$$

Where, δ_H and δ_0 represent the tensile properties of the specimens with and without hydrogen pre-charging, such as elongation (El) and reduction of area (RA), based on the research of Zhao et al. (2021). Our study considered RA for the calculation of EI.

The EI value was utilized for the labelling process, which proceeded according to condition:

$$EI \geq 50 \begin{cases} 1 \\ 0, otherwise \end{cases}$$

The labels indicate two different classifications. Class 1 is defined by an EI higher than 50%, indicating materials not recommended for hydrogen applications under the specified testing conditions. Alternatively, class 0 represents materials with EI lower than 50%, which indicates low susceptibility to HE but does not guarantee the suitability of a material for hydrogen transport or storage under the specified operating conditions, and further analysis is required, as stated by Subedi et al. (2023a). Quantitatively, 101 data samples of this dataset received label 0 (73.72%), and 36 received label 1 (26.28%).

3.3. Data preprocessing

3.3.1. Removing missing values

In the initial stage of the preprocessing, we addressed missing data by eliminating instances with incomplete information which were not filled in by standard values during data curation. Handling missing values ensures the integrity and reliability of a dataset, establishing a robust base for subsequent analyses. This finding was presented by Needham et al. (2009).

Immediately after the process of removing missing data, the dataset was partitioned into an 80:20 ratio, with 80% allocated for training and 20% for testing.

3.3.2. Feature selection

Feature selection is an important step in ML, aiming to identify the most relevant features to enhance model performance, as described by Maior et al. (2023). In our study, we employed gradient boosting (GB) for this purpose. GB utilizes a metric called feature importance to assess the significance of each attribute in decision-making during tree construction. According to Upadhyay et al. (2021), Feature importance scores are computed by comparing and ranking all features in the dataset, with the importance of a feature determined by the number of splits associated with it, weighted by the observations from each split. Purity metrics like the Gini Index are commonly used to select split points, and the feature importance of each tree is averaged across all trees in the model. Figure 2 shows all the features ranked by its importance value.

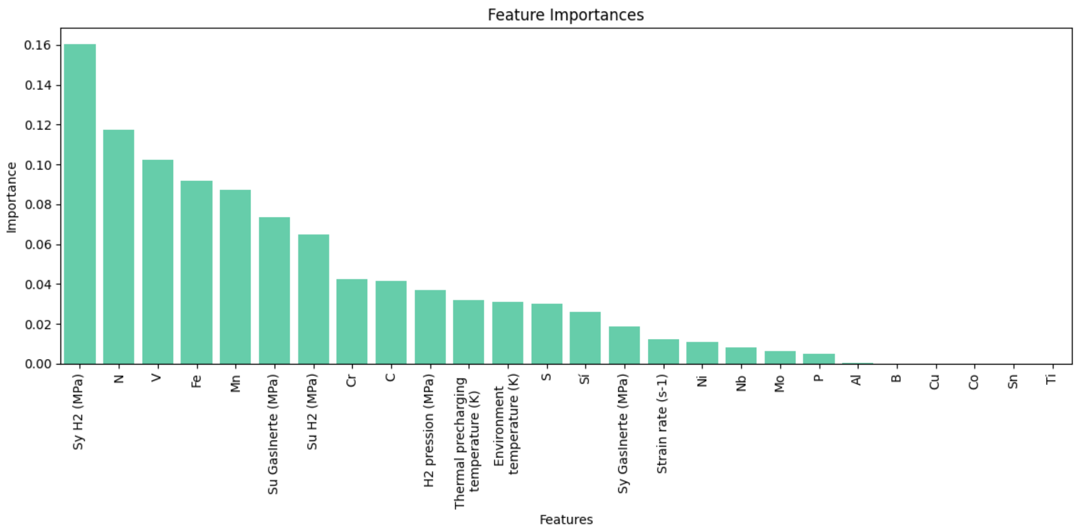

Figure 2. Ranking of features by importance.

We then established a threshold of 0.05 for feature importance values and selected the top 7 features from the dataset based on this criterion. Notably, this threshold was chosen empirically, without a technical procedure, serving as a practical guideline for selecting the features that most contribute to the model's predictive performance.

3.3.3. Data augmentation

Considering the imbalance in the database, where the majority class is significantly overrepresented compared to the minority class, we implemented a random oversampling data augmentation to address this issue. This technique involves randomly selecting instances from the minority class and replicating them to increase their representation in the dataset. This process continues until the number of instances in the minority class matches that of the majority class, as seen in Amin et al. (2016). The objective is to provide the model with more instances of the minority class, optimizing the performance of the models by ensuring that it has sufficient data to capture patterns and make predictions across all classes accurately, as analysed in the study of Nemade et al. (2023). It is also important to mention that data augmentation is performed after the separation of the dataset into training and test and is only applied to the training data, based on the approach of Maior et al. (2021).

3.3.4. Standardization

In the standardization step, we applied StandardScaler to the data. This technique works by calculating the mean and standard deviation of each feature in the dataset. Then, it subtracts the mean from each feature and divides by the standard deviation, according to Gelman (2008). This transformation results in a distribution with a mean of 0 and a standard deviation of 1 for each feature, effectively centering the data around 0 and scaling it to unit variance. This process improves the performance of ML models, especially those sensitive to feature scales. This outcome was presented by Sales da Cunha et al. (2023).

3.4. Modeling

3.4.1. Optimization of hyperparameters

The optimization of hyperparameters was a part of the ML model's modeling process. We employed GridSearchCV to determine the most optimal hyperparameters for our classification models. This algorithm systematically explores a predefined grid of hyperparameter values, evaluating the performance of the model for each combination using cross-validation, as stated by Shams et al. (2023). Thus, it performs an exhaustive search through the hyperparameter space, facilitating the identification of the hyperparameter values that achieve best performance metrics, enhancing the robustness of the models. These findings are observed in the study of Belete and Huchaiah (2022).

3.4.1. Predictive models

Supervised classifiers models were utilized to predict the target variable based on the features in our dataset. These models have the ability of learn from labeled data, where each data point is associated with a known class or category, as shown in Maior et al. (2020). According to Lo Vercio et al. (2020), the classifiers aim to identify complex patterns within the data to make predictions about the class labels of unseen instances. They are trained on a labeled dataset, which is then used to identify or make decisions on new unseen data. The models utilized are defined bellow:

K-Nearest Neighbors (KNN): KNN is a classifier that works by storing all available cases and classifying new cases based on a similarity measure (e.g., distance functions), as described by Maillo et al. (2015). This model has the concept that classes are determined by a majority vote of its neighbours. The choice of k, the number of neighbors, is a critical hyperparameter that influences the performance of the model. This observation is presented in the research of Won Yoon and Friel (2015).

Adaptive Boosting (AdaBoost): AdaBoost is an ensemble learning method that combines multiple weak learners (typically decision trees) to create a strong classifier, as seen in Subasi et al. (2018). According to Cao et al. (2013), this algorithm trains a sequence of weak learners, each focusing on the instances that were misclassified by the previous learners. The final prediction is made by combining the predictions of all weak learners, weighted by their performance,

Categorical Boosting (CatBoost): CatBoost is a powerful ML algorithm designed for dealing with categorical variables without the need for extensive preprocessing, such as one-hot encoding, as noted by Zhang and Jánošík (2024). Based on Dong et al. (2021), this classifier utilizes ordered boosting, which optimizes the sequence of trees built during training, resulting in faster convergence and improved performance.

Extra Trees Classifier (ETC): According to Ampomah et al. (2020), ETC is an ensemble technique that builds multiple decision trees and combines their predictions through voting or averaging. Unlike traditional decision trees, ETC introduces additional randomness by selecting random thresholds for each feature at each split, reducing overfitting and improving the generalization of the model, as shown in the study of Joshi et al. (2021).

4. RESULTS

Our investigation into classification ML models achieved different accuracies through the implementation of diverse preprocessing techniques, such as feature selection (FS), data augmentation (DA), and hyperparameter tuning. Additionally, we considered balanced accuracy when evaluating our findings. This calculation is expressed in Equation (2) for binary classification, according to Chicco et al. (2021).

$$Balanced\ accuracy = \frac{1}{2} \times \left(\frac{TP}{TP + FN} + \frac{TN}{TN + FP}\right) \tag{2}$$

From our analysis, all models have relatively high accuracy, indicating a correct classification ability of most examples. AdaBoost was the model with best accuracy of the study with 93%, followed by ETC and KNN classifiers. All results obtained can be found in the appendix section of this study. Table 2 presents results from models with high accuracy performance when utilizing preprocessing techniques.

Table 2. Balanced accuracy results.

	Preprocessing		
	tuning	tuning + DA	tuning + DA + FS
ETC	0.75	0.75	0.90
KNN	0.60	0.85	0.90
ADA	0.60	0.83	0.93

In Figure 3, we can observe the significant impact of hyperparameter tuning and the FS technique on models trained with DA, particularly the ETC and KNN, which led to considerable increases in accuracies. However, the CAT model did not perform well with the FS and tuning technique, presenting a slight fall in its accuracy.

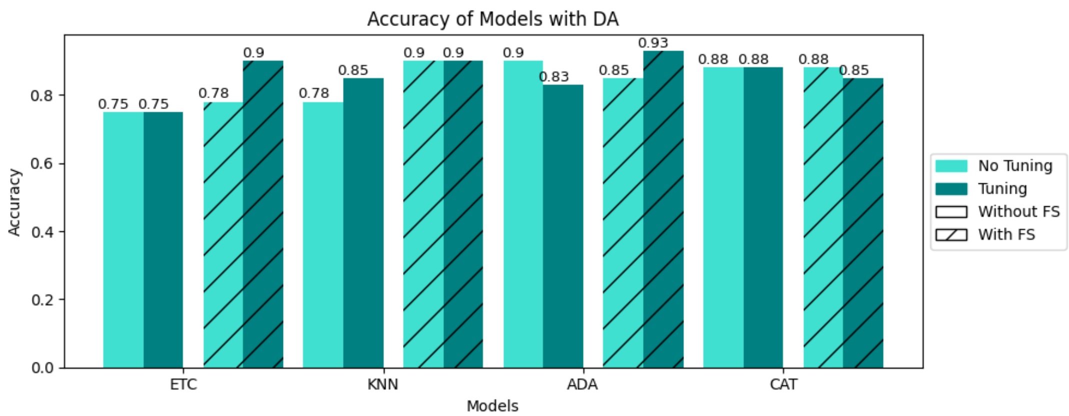

Figure 3. Accuracy of models utilizing data augmentation.

Examining the precision, recall, and F1-score metrics from the models with high accuracies helps to better understand their performances as classifiers. Considering the utilization of tuning, DA, and FS approaches, the

ETC exhibits the best precision (0.96) and F1-score (0.95), indicating great ability to avoid false positives and maintain a balance between precision and recall. ADA also shows a respectable precision of 0.93 and a recall of 0.88, resulting in an F1-score of 0.89, proving to be a strong competitor. However, the KNN shows relatively weaker performance, especially in terms of recall (0.84) and F1-score (0.85), despite a reasonable precision of 0.91 as shown in Table 3.

Table 3. Precision, recall, and f1-score from best models.

	ETC	KNN	ADA
Precision	0.96	0.91	0.93
Recall	0.96	0.84	0.88
F1-Score	0.95	0.85	0.89

When analyzing the confusion matrices, we can observe that both KNN and ADA exhibit similar rates of true positives and false positives, suggesting that they both tend to incorrectly classify some negative examples as positive. On the other hand, ETC demonstrates a perfect rate of true positives but incurs a rate of false negatives, indicating that it lacks some positive examples. This may suggest that the variations introduced by data augmentation technique may not be fully captured by the model, leading to a loss of some positive examples during the classification process. The confusion matrices of these models are presented in Figure 4.

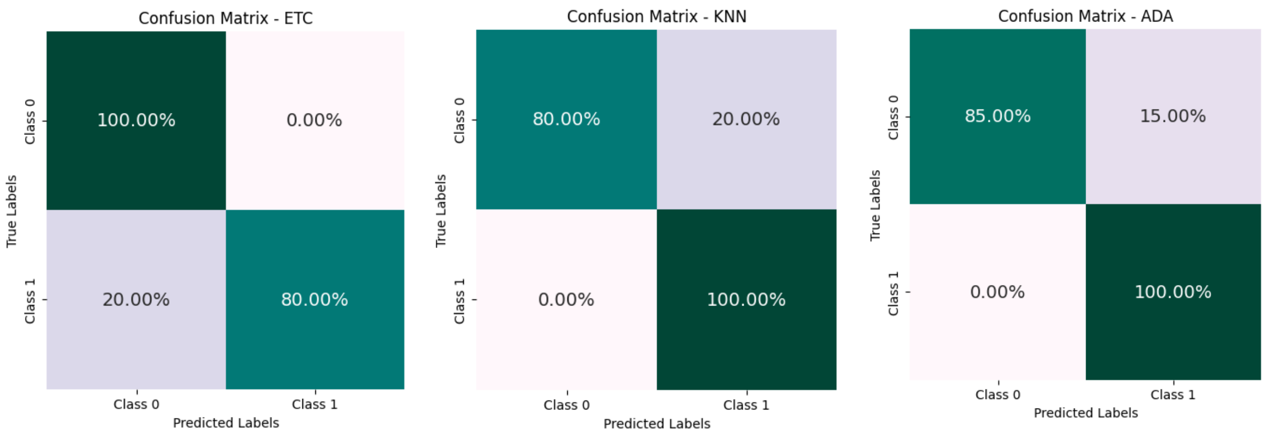

Figure 5. Confusion matrices from best models.

5. CONCLUSION

This study applied Machine Learning (ML) classification models to analyze factors contributing to Hydrogen Embrittlement (HE) susceptibility, considering material chemical composition, environmental conditions, and testing methods. Among the models tested, AdaBoost achieved the highest accuracy at 93%, followed by ETC and KNN classifiers. However, some models did not show significant improvements with hyperparameter tuning, indicating the need for a deeper exploration of parameter combinations and tuning techniques (grid search). Although ETC demonstrated high recall and f1-score values, the confusion matrix suggests that these metrics might be influenced by data augmentation issues inherent to the model. For future analysis, we recommend using an alternative data augmentation technique with ETC to enhance the model's performance. Another challenge faced was data acquisition. Investing in expanding the database is important for this work to significantly contribute to the development of a robust tool for classifying different steel types intended for hydrogen transport and storage. The advancement of this methodology can be essential to support the expansion of the hydrogen supply chain, promoting the wider adoption of hydrogen as a clean energy source.

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Appendix

Table 4. All tests result.

		without tuning		with tuning	
		without DA	with DA	without DA	with DA
ETC	without FS	0.75	0.75	0.75	0.75
	with FS	0.75	0.78	0.75	0.90
KNN	without FS	0.68	0.78	0.60	0.85
	with FS	0.68	0.90	0.68	0.90
ADA	without FS	0.90	0.90	0.60	0.83
	with FS	0.85	0.85	0.63	**0.93**
CAT	without FS	0.80	0.88	0.78	0.88
	with FS	0.68	0.88	0.60	0.85

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