Performance Evaluation of GAF-CNN for Ion Concentration Prediction from Cyclic Voltammetry Data

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1. Introduction

Molten Salt Reactors (MSRs) employ molten salts as both fuel and coolant, enabling high-temperature, low-pressure operation with high thermal efficiency and strong passive safety [1,2]. However, chloride- and fluoride-based melts can corrode structural materials at elevated temperatures, making real-time diagnosis of dissolved metal-ion behavior essential. Cyclic voltammetry (CV), which records redox processes as current–potential waveforms, is a promising technique for this purpose.

In practice, reproducibility is hindered by peak overlap in multicomponent environments, non-ideal background currents, variations in electrode surface condition, and waveform drift with temperature and scan-rate. Traditional peak deconvolution and empirical models are vulnerable to such variability, and operator-dependent preprocessing introduces interpretive bias. While deep-learning–based representation learning offers an alternative for handling complex waveforms, rigorous validation remains limited as to whether image-encoded CV preserves information sufficient for both qualitative and quantitative analysis [3,4].

This study proposes a pipeline that converts CV time series into Gramian Angular Field (GAF) images and uses a pretrained EfficientNet-B0–based multi-task model to perform ion classification and concentration prediction simultaneously. Under leakage-controlled evaluation, the approach shows significant performance, substantiating the feasibility of automated CV analysis and quantitative prediction for MSR environments.

2. Methods

Fig. 1 outlines the overall workflow. A single CV cycle is normalized and converted into a three-channel GAF image, from which global-correlation features are extracted using a pretrained EfficientNet-B0. The learned features are concatenated with meta-variables (temperature, scan rate) and fed to task heads that perform ion-species classification and concentration regression jointly. Finally, performance evaluation was conducted.

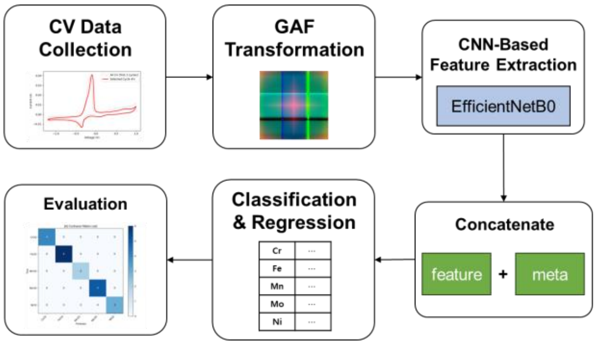

Fig. 1. Overview of the proposed pipeline

2.1 Data Description and Acquisition

All chemicals for this study were prepared and handled within a high-purity argon atmosphere glove box to minimize oxygen and moisture contamination. A eutectic mixture of NaCl and $MgCl_2$ was utilized as the solvent for the molten salt, to which five metal chlorides ($CrCl_2$, $FeCl_2$, $MnCl_2$, $MoCl_5$, $NiCl_2$) were added to form the molten salt samples.

Electrochemical measurements were performed under static conditions using a custom-designed quartz tube cell. Tungsten wires were used as working electrodes, quasi-reference electrodes, and counter electrodes. To ensure repeatable and reliable data collection, the electrodes were subjected to thorough polishing, ultrasonic cleaning, and drying processes to maintain clean surface cleanliness. The entire cell assembly was performed under an inert atmosphere to prevent contamination. CV measurements were performed under an inert atmosphere. CV data were obtained for each metal chloride species at three different temperature conditions: 723 K, 773 K, and 823 K, multiple scan rates of 0.8 mV/s at 0.05 mV/s, and three different concentration levels: 0.1 wt%, 0.3 wt%, and 0.5 wt%. All experiments followed a consistent laboratory environment and measurement protocol to ensure data reliability.

Each raw CV file was segmented into cycles of 2,000 points. When available, the representative cycle was chosen between the second and the third cycles, selecting the more reliable one; when only 1–2 cycles were present, the last available cycle was used. Files without any complete cycle were excluded, and the selected cycle was locally smoothed only for isolated extremes to preserve overall peak shapes.

To enhance the robustness of the model, a fivefold data augmentation was applied to each sample. This augmentation involved current jitter, which added Gaussian noise (standard deviation $\sigma = 0.02 \times$ current range), and current scaling, which applied a uniform factor within the range of [0.95, 1.05]. An example of the data augmentation is shown in Fig. 2.

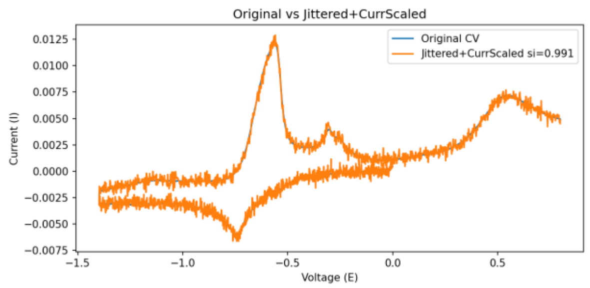

Fig. 2. Examples of data augmentation

2.2 GAF Image Representation

Time-series signals were embedded as images using the Gramian Angular Field (GAF). Given a univariate series X_t, we rescaled to [−1,1]:

$$(1)\quad x_t = \frac{2(X_t - \min X)}{\max X - \min X} - 1$$

$$(2)\quad \phi_t = \arccos\,(x_t)$$

then mapped in polar coordinates with angle $\phi_t = \arccos(\tilde{x}_t)$ and radius $r_t = t/N$. Because $\cos(\phi)$ is monotonic over $[0, \pi]$, the mapping is bijective, and the radius preserves absolute temporal order. The Gramian Angular Summation and Difference Fields are computed as:

$$(3)\quad GASF_{ij} = \cos(\phi_i + \phi_j)$$

$$(4)\quad GADF_{ij} = \sin(\phi_i - \phi_j)$$

We formed a 3-channel image by assigning R = GASF(E), G = GADF(I), and B = GASF(I), followed by linear scaling to 0–255 and resizing to 224×224×3 (RGB). Fig. 3 shows an example of a representative image for each ion.

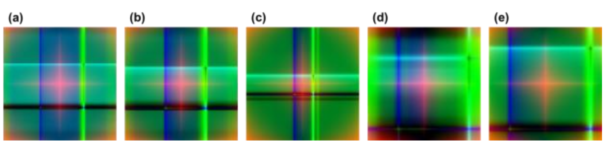

Fig. 3. Representative GAF Images of CV Signals for Various Metal Salts: (a) $CrCl_2$, (b) $FeCl_2$, (c) $MnCl_2$, (d) $MoCl_5$, and (e) $NiCl_2$

2.3 CNN Model Architecture, Training and Evaluation

This study used a single multi-task CNN. The input is a 224×224×3 GAF image, and ImageNet pre-learning EfficientNet-B0 was used as the feature extraction backbone and frozen during learning. After combining the temperature and scanning speed meta-variables with the 1,280th characteristics of the backbone, ion classification (softmax) and concentration regression (linear) were simultaneously calculated through the shallow complete connection layer. The data were divided into group-based 80:20 to block leakage of augmented samples derived from the same original, and the meta-variable and target concentration were standardized with learning set statistics, and the regression prediction was reverse-transformed and reported as wt%. Learning was conducted based on Adam, and early termination and learning rate reduction were applied to prevent overfitting. For the evaluation, the accuracy of classification and macro-F1 and RMSE·MAE·R^2 of regression were used, and the convergence characteristics were confirmed by confusion matrix and learning curve. The model and learning settings are summarized in detail in Table 1.

Table I: Problem Description

Item	Setting
Input	GAF image 224 × 224 × 3
Backbone	EfficientNetB0
Feature dimension	1,280
Meta-variables	Temperature, Scan rate
Prediction head	Dense 256–ReLU–Dropout 0.3 → Dense 128–ReLU–Dropout 0.2
Data split	Group-based 80:20; leakage prevented by grouping augmented samples with their originals
Standardization	Both meta-variables and targets standardized using training-set statistics; regression outputs inverse-transformed to report wt%
Optimizer	Adam (default settings)
Batch/Epochs	batch 32 / up to 80 epochs
Callbacks	EarlyStopping (restore best weights), ReduceLROnPlateau
Classification metrics	Accuracy, macro-F1 (confusion matrix shown)
Regression metrics	RMSE, MAE, R^2
Diagnostics	Training/validation loss & accuracy curves; parity (prediction–ground truth) plot (optional)

3. Results and Discussion

This section presents the performance evaluation of the developed pipeline, which incorporates GAF-based image representation, a frozen EfficientNet-B0 backbone, and a two-layer multi-task head. The pipeline was assessed on a group-based 80:20 data split (with a validation set of n = 20 samples), ensuring a balanced representation of species. To prevent any data leakage between partitions, all augmented samples derived from the same original source were confined to their respective sets. All reported metrics in this section refer exclusively to the validation set.

3.1 Species Classification

The proposed pipeline demonstrated exceptional performance in species classification, achieving a perfect accuracy of 1.00 and a macro-F1 score of 1.00. The corresponding confusion matrix, presented in Fig. 4, exhibited a completely diagonal structure, clearly confirming the flawless identification of all five chloride species without any misclassification errors. This result highlights the strong capability of the methodology for qualitative analysis of molten-salt species under the tested conditions.

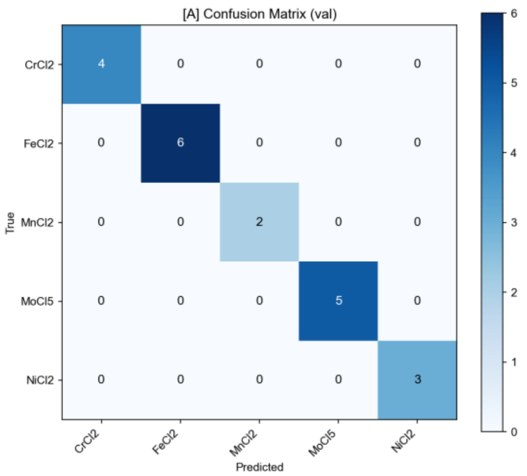

Fig. 4. Confusion Matrix on the Validation Set

3.2 Concentration Regression

For the quantitative prediction of concentration (wt%), the pipeline yielded a RMSE of 0.1212 wt%, a MAE of 0.0847 wt%, and an R^2 value of 0.567. The positive R^2 value signifies that the model provides a meaningful degree of predictability, indicating a substantial improvement over a simple mean predictor. While not representing a perfect fit, this R^2 demonstrates the pipeline's capability to capture a considerable portion of the variance in target concentrations, marking a significant step towards automated quantitative analysis for this specific dataset. Further efforts to enhance this predictive power will be a focus of future work.

4. Conclusion

The developed GAF + EfficientNet-B0 pipeline demonstrated excellent species classification and meaningful concentration regression performance on the evaluated dataset. While the R^2 value indicates moderate predictive capability, it highlights room for improvement in quantitative accuracy. A notable limitation is the relatively small size of the validation set, which may affect the generalizability of the results. Future work will focus on extensive validation using larger and more diverse datasets, including external test sets and cross-laboratory comparisons. Additionally, stress testing across an expanded range of temperatures and scan rates will be conducted to better assess robustness. These steps are essential to establish the pipeline's reliability and applicability in real-world molten-salt electrochemical analysis.

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