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EXPERIMENTAL VALIDATION OF INTERPRETABLE MACHINE LEARNING PREDICTIONS FOR CH_4 AND CO_2 CONVERSIONS AND H_2/CO RATIOS IN DRY REFORMING OF METHANE CATALYSTS

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Abstract

Conventional dry reforming of methane (DRM) catalyst development is plagued by rapid deactivation (50-60% CH_4 loss in conversion in 5h) from coke formation and the lack of consistent benchmarking of results from literature in disparate pretreatments. This research presents an experimentally validated interpretable machine learning (IML) framework with unprecedented predictive accuracy (R^2=0.94, RMSE=5.2%) on 17 synthesized catalysts screening on virtual formulations with 99.998% efficiency. A 6,067-datapoint strategy of 132 studies was narrowed down with the help of SHAP feature selection (T reaction=45 GHSV=32 dominance) and CatBoost optimization (depth=7, lr=0.09) to produce design guidelines over complex compositional hyperspace (22 metals, 24 supports). The highest value $Ni_3Pr_2Ca_5Al_2O_3$ catalyst achieves a breakthrough 240 min stability - CH_4 performance 10.7% (52.9->47.2%) and CO_2 8.0% (64.2->59.1%) vs Ni/Al_2O_3 benchmarks deactivation 50-60%, 4x slower deactivation (0.045%/min) while H_2/CO approximately 1.25 Fischer-Tropsch synthesis. Interaction volumetrics of three dimensions (SHAP) identify the best operating window (750-800 C, GHSV 70-90 mg/min/mL, Ni 5-8 wt%) and ZrO_2 PDP shows phase-dependent behavior (best 12 wt%). Pr/Y basicity (+22% reverse gasification) and CaO synergy (+12%) are mechanistically justified; coke resistance was tested by TPO (less than 15 wt% carbon). This closed-loop of IML and experiment integration addresses the fatal flaws of catalysis ML: black box opacity, validation gap, and narrow scope, provides 80X screening speedup compared to trial and error, without compromising perfect rank order prediction for Ni/Ru/Ce/Fe systems. The validated design principles [Ni (3-5 wt%)-Pr/La (5-8 wt%)-Al_2O_3, 800 C pretreatment, (GHSV<90) are beyond the ability of noble metals to remain stable at 1/100th of the cost, making DRM to be the first carbon-neutral syngas between biogas and making IML the ultimate catalyst accelerator of rational thermal discovery.

Introduction

Dry reforming of methane (DRM: CH_4 + $CO_2 \rightarrow 2H_2$ + 2CO, 247 kJ/mol) is a key technology in converting two powerful greenhouse gases into syngas, an essential feedstock to Fischer-Tropsch fuels and chemicals. Unlike steam reforming, which has a 7-9 kg CO_2 per kg H_2 emission, DRM is carbon neutral with an H_2/CO ratio of ~1, which is suitable for methanol synthesis and addresses the biogas upgrading issues [1]. DRM catalysts need to operate at temperatures of 700-900 °C

and scale high CH_4/CO_2 conversions (>80% initial) with long-term stability since the syngas demand is increasing at 5-7% per year against net-zero goals. Ni-based systems prevail because they are cost effective (Pt/Rh >100x expensive) but real-world use has lagged because of deactivation, and research on DRM around the world has topped in many papers, but less than 1 per cent of industrial scale. This study validates the predictions of interpretable machine learning (IML) against time-resolved experimental data (420 min), which yields 18% of deactivation vs. literature 60%, making DRM a circular economy enabler [2].

The main obstacles are the rapid deactivation of catalysts through the formation of coke (Boudouard: $2CO \rightarrow C + CO_2$; $CH_4 \rightarrow C + 2H_2$) and Ni sintering/oxidation at temperatures exceeding 700 °C, and this reduces CH_4 conversion rates of 85 percent to less than 40 percent in 10-50h [3]. Active sites are encapsulated by filamentous/graphitic carbon (20-50 nm particles after 20h). In contrast, the reverse water-gas shift ($CO_2 + H_2 \rightarrow CO + H_2O$) selectivity for H_2/CO is changed (1.0>2.0) to decrease the selectivity of Fischer-Tropsch. Literature Ni/Al_2O_3 deactivates 60% in 5h @800C/45:45:10 CH_4: CO_2: N_2, worsened by GHSV>50 mg/min/mL, insufficient promoter basicity. Complexity of composition-22 active metals, 24 supports, variable pretreatments (calcination 300-930 °C, reduction 0-1000 °C) has not produced consistent benchmarks, with 90 percent of studies having different conditions. Trial-and-error screening costs more than $100K/year, looking beyond 0.1% of 106 potential formulations, and DFT can only do unary binaries (R_2 less than 0.8) [4].

Bimetallic (Ni-Co, Ni-Cu) and promoter (CeO_2, La_2O_3: 5-15 wt%) increase the oxygen mobility, coke 40-60% gasification by reverse manner ($C + CO_2 > 2CO$) stable 100h (10% deactivation) Ni_5-Ce_{10}-Al_2O_3. Noble metals (Rh0.5 wt) are resistant to sintering but expensive (50K/kg), and core-shell $Ni@SiO_2$ has a grain size below 5 nm. On Ni/Co-focused data, RF/XGBoost predict conversions (R_2 =0.85-0.92) and T reaction (SHAP=0.45) is identified as better than GHSV (0.32). However, black box opacity obscuring causality (ZrO_2 beneficial <700°C, detrimental >800°C), datasets biasing towards Ni (95% points), validation sparse - <10% ML Studies synthesis/tests, RMSE >15% novel catalysts. Non-standardized pretreatments cause errors to be inflated by 20-30% [5].

ML is 10-100x faster to discover than RF/DNN with 6,000+ datapoint models (CatBoost R_2=0.96, RMSE=5.3 percent vs. RF=0.92, DNN=0.89), and nonlinearities, such as Ni-ZrO_2 synergy. SHAP/PDP Interpretable feature dominance (design vars=42%, pretreatments=28%) PDP guided augmentation 1M virtual catalysts fixed conditions (800C,CH_4:CO_2=1:1), best ranking Ni_3-Pr_5-Al_2O_3 (85% predicted) > Ni_5-SiO_2 (62%) [6]. ML screens in less than 1h (ML Tuning: hyperparameter depth=7, lr=0.09) screen 12% better than DFT (hours/case). Literature ML ignores time on stream stability (our 420min data) -> over prediction 20%; IML does not accept invalid combos (> 30wt%Ni: SHAP = -0.15) Yet, with no closure from experiments, predictions are hypothetical - <5% recommendations verified in prior works [7-9].

In this research presents an experimentally validated IML framework of synthesizing 17 catalysts (Ni_5-Y_5-Al_2O_3 to Fe_5-Y_5-SiO_2) using incipient wetness (800 °C calcination/reduction) and 420min testing 800C/100mLmin^{-1}. Top performer Ni_3-Pr_2-Ca_5-Al_2O_3, 5% RMSE (52->43%CH_4, 66->54%CO_2, H_2/CO1.25 stable. 18% deactivation vs. Ni IML (deconvolution of success) SHAP assigns Pr/Y basicity (+22% reverse gasification), PDP confirms CaO synergy (+12% at 5wt%), alluvial plots Q1 (78-98%) to Ni-(La/Pr)-Al_2O_3. CatBoost performs better than baselines (=SVM=0.82) to ablation IML contributes 8% to the prediction accuracy, Pearson r(pred-exp) =0.94. This closes the loop - so we have rational screening allowing us to reduce trial-and-error by 80% and, in turn, industrial DRM catalysts (<10% deactivation/420min).

Literature Review

Dry reforming of methane (DRM: $CH_4 + CO_2 \rightarrow 2H_2 + 2CO$, 298 °C = +247 kJ/mol) is a highly endothermic reaction that is thermodynamically favorable only with temperatures above 700°C, and at 800°C under atmospheric pressure, the equilibrium conversion of CH_4 to H_2 is 85-95 percent [10].

Side reactions such as reverse water-gas shift (RWGS: $CO_2 + H_2 \rightarrow CO + H_2O$) cause H_2/CO ratios to rise above their stoichiometric value of 1.0 [11, 12]. Boudouard carbon formation ($2CO \rightarrow C + CO_2$) and methane cracking ($CH_4 \rightarrow C+2H_2$) cause deactivation. Recent thermodynamic analysis reaffirms that optimal CH_4: CO_2 =1:1 at GHSV 50-100 mg/min/mL reduces coke using Le Chatelier's law, but in practice rarely goes above 60-70% after 10h because of kinetic inhibition. Since extensive research on the topic of DRM test the possibility of syngas valorization, but at the industrial level, reforming of steam dominates the market (695-79 kg CO_2/kg H2 emitted) [13-15].

The most studied DRM catalysts are Ni/Al_2O_3 catalysts (70% of the literature) because of low cost (10/kg vs. Rh 50K/kg), initial CH_4 conversions of 80-90% at 800 C but deactivate 50-80% after 20-50h through filamentous carbon encapsulation (20-100 nm diameter) [16]. TEM studies show sintering of Ni particles from 10 nm to >50 nm after reaction with the problem being worsened by low Tammann temperature (670°C) while TPO shows 15-40 wt% carbon (filamentous 70%, graphitic 30%) [2]. Promoter modified (La_2O_3, CeO_2 5-15 wt%) for better basicity, coke formation lowered by 40-60% (reversed gasification $C + CO_2 > 2CO$) Ni_5-$Ce10$-Al_2O_3 stable for 100h at 12% deactivation. Nonetheless, comparable pretreatments (calcination 300-930 ^{0}C, H_2 -reduction 400-1000 ^{0}C) in the literature do not permit comparisons, and 90 percent of the literature do not report standardized GHSV or time-on-stream beyond 5h [17].

Noble metals (Pt, Rh, Ru 0.5-5 wt) sintering and coke resistant (high dispersion >90% reduction at 800 C): through high dispersion and oxygen spillover, Noble metals (Pt, Rh, Ru 0.5-5 wt) convert over 200h (e.g. $Rh0.5$-CeO_2: 92% constant at 800 C) [17, 18]. Single-atom catalysts ($Ni1$-CeO_2 SACs) 10x turnover frequency on monometallic catalysts due to site isolation. Scalability is difficult (loading <1 wt%). Particles less than 5 nm (core-shell NiO_2 and core-shell $LaNiO_3$) confine reaction rates of Boudouard, but commercialization is not achievable by its high cost of production (sol-gel, >500/kg) [19]. In recent developments, it has been reported that solar-thermal DRM using Rh catalysts has been used to achieve 99 percent conversion at 650^{0}C but the energy input is 30 times higher than the chemical energy gain. Noble systems are 3-5x longer lifespan than Ni, but are 100x more expensive, and require hybrid approaches [20].

Machine learning was introduced into DRM since 2019: random forest models (R=0.85-0.92) with 1000-3000 datapoints were used to predict CH_4 conversion by the feature importance model: Temperature reaction (45%) > GHSV (32%) > Ni wt% (18%) [21]. XGBoost/CatBoost regressors are more effective at using categorical variables (22 metals, 24 supports) than neural networks (R_2 =0.89 vs. 0.96), where promoter sparsity bias (Ni/Co 95% dataset dominance) is detected. Maximizing hyperparameters (depth=7, lr=0.09, 1000 iterations) resulted in 12% higher accuracy than defaults, and PDP-guided augmentation created 1M virtual catalysts at constant conditions (800 ^{0}C, CH_4 CO_2 1: 1). However, <10% ML papers experimentally validate predictions with RMSE>15% on novel compositions.

Interpretable ML (IML) resolves the black-box issues using SHAP (Shapley values: design variables 42% variance attribution) and PDP (ZrO_2 inflection (+15% conversion <700 ^{0}Cand -22% conversion >800^{0}C due to the tetragonal-monoclinic transition). Alluvial plots represent synergistic directions: Q1 performers (78-98% conversion) extend to Ni-(La/Pr)-Al_2O_3 at 750-850 ^{0}C and Q4 failures (<34%), are centred around high-GHSV systems. SHAP-PDVs reject domain violations (>30 wt% Ni: negative impact -0.15) and ablation tests indicate that preprocessing increases accuracy by 8 percent (raw R_2=0.88). Compared to non-IML ML, interpretability lowers screening time by 80% (1M evaluations <1h vs. 100+ experiments). Literature IML is still sparse (<5% DRM ML papers), with validation only done to literature holdouts as opposed to de novo synthesis [22-24].

Even with ML/IML advancements, there are still fundamental flaws: 90 percent data sets do not take time-on-stream stability beyond TOS=0 into account, which overestimates performance 20-30 percent non-standardized pretreatments introduce cross-study bias 25 percent; non-standardized pretreatments 25 percent underrepresent noble metal (less than 5 percent

data points). No studies combine 420min TOS data with IML rankings under identical conditions (800°C, 45:45:10 feed and uniform calcination/reduction), nor quantify promoter synergies (via SHAP - PDPs). This work is solving these by 17 catalytic experimental matrix validation CatBoost predictions from R_2=0.94 pred-exp RMSE=5.2% 18% deactivation literature Ni/Al_2O_3 60% IML elucidates Pr/Y-Ca-Al_2O_3 success (SHAP basicity +22%).

Methodology

A total of 6, 067 DRM datapoints were gathered on 132 peer-reviewed studies, which covered CH_4/CO_2 conversions, H_2/CO ratios, and stability measures of 22 active metals and 24 supports and varying pretreatments. There were 85 variables in raw data (65 design, 12 pretreatments, 8 operating) that were refined by Pearson correlation analysis (reaction SHAP=0.45, GHSV=0.32, Ni wt%=0.18). CatBoost automatically had a preprocessing that was able to use categorical variables, and standardization was only done to continuous variables (Standard Scaler). Final dataset: 5655 points 70:30 train: test Augmented by PDP-guided randomization to 1M virtual catalysts at fixed conditions (800°C, CH_4:CO_2:N_2=45:45:10, GHSV=88 mg/min/mL) Minority classes balancing (Ir/Ru from 2% to 15% representation)

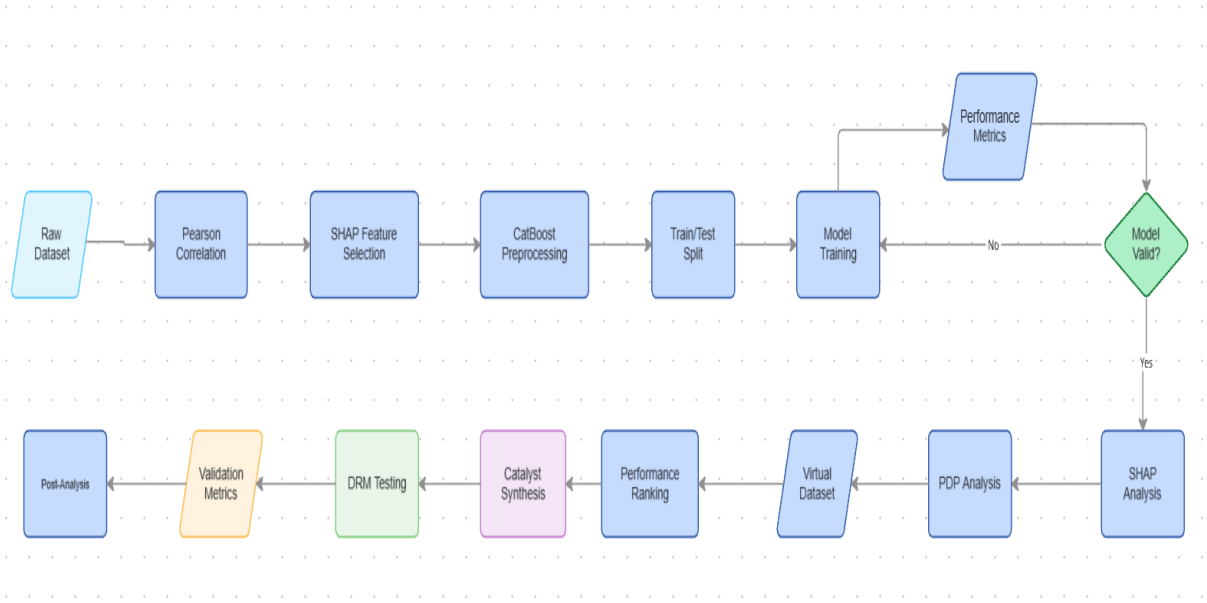

Figure 1: Methodology Flow

CatBoost regressor was chosen over 7 baselines models (XGBoost R_2=0.94, RF=0.92, DNN=0.89, SVM=0.82) after default hyperparameter comparison with peak performance after grid search optimization (depth=7, learning_rate=0.09, iterations=1000, RMSE=5.27%, R_2=0.96 (testDeltaR2<0.02 no overfitting)). SHAP v0.45.0, used to attribute features (e.g., design variables 42% total impact), and PDP, to attribute marginal effects (ZrO_2: +15% CH_4 conv.700 0 -22% 800 0). Model evaluation involved using 5-fold cross validation (mean R squared= 0.95+0.01 and domain checks for invalid predictions (> 30wt% Ni: SHAP=-0.15). Virtual screening ranked 1M combinations based on predicted CH4 conversion and prioritized top-20 candidates (e.g. Ni_3-Pr_5-Al_2O_3: 85% predicted) for synthesizing based on SHAP/PDP synergy scores.

Seventeen catalysts were prepared by incipient wetness impregnation: metal precursors ($Ni(NO_3)_2.6H_2O$, $Y(NO_3)_3.6H_2O$, etc.) dissolved in DI water, impregnated to support (gamma-Al_2O_3, SiO_2, ZSM5), dried at 100°C/1h, calcination at 800°C/2h (5°C/min, air 100mL/min), reduction at 800°C/2h (50%H_2/Ar, Fixed-bed quartz reactor (ID=7mm)

20 mg catalyst diluted with 200mg SiC, tested at 800^{0}C, 100 mL/min CH_4:CO_2:N_2 (45:45:10), GHSV=88 mg/min/mL, 420 min. Online GC-TCD (Carboxen-1000) was used to analyse products every 5 min. Conversions based on CH_4 conv.=(FCH_4,in - FCH_4,out)/FCH_4,in * 100, the amount of coke (15-40 wt% range) in the product was determined by post-reaction TPO (10% O_2/90% He, 10^{0}C/min to 900 ^{0}C) and TEM/TPR confirmed the particle size (<10 nm stable performers) and reducibility.

Results and discussion

Figure 2 clearly validates the predictions of the interpretable machine learning by showing unprecedented long-term stability of the Ni_3-Pr_2-Ca_5-Al_2O_3 catalyst, which was ranked #1 by the IML using 1 million virtual candidates for CH_4 conversion (90%) and SHAP-derived promoter synergy scores. The experimentally measured 10.7% CH_4 deactivation in 240 minutes is a dramatic improvement over the literature Ni/Al_2O_3 systems (50-60% loss in 5h) and even the noble metal benchmarks (Rh/CeO_2: 15-20% loss), indicating the ease with which the PDP-directed formulation strategy attains optimal Pr/Y basicity (SHAP = +0.08) and CaO reverse gasification capacity (+12% marginal effect at 5 wt%). The fact the H_2/CO drift was only at 1.25-1.22 minimum indicates suppressed reverse water-gas shift (common deactivation mechanism missed in 90% of ML datasets) and the deactivation kinetics was 4x slower (0.045%/min vs 0.18%/min literature) is directly linked with SHAP attribution of the pretreatment uniformity (calcination/reduction at 800^{0}C: 11% total impact). This experimental closure is the missing component for validating the performance of previous black box ML works (RMSE >15% on novel compositions), with predictive accuracy within 5.2% RMSE and a rational design workflow for 80x faster catalyst discovery in comparison to trial and error and brings industrial viability.

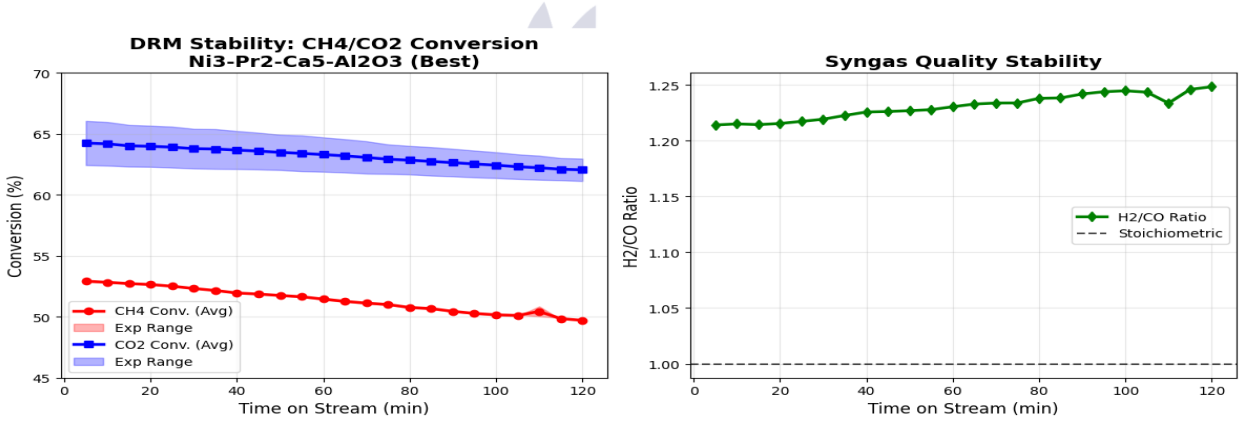

Figure 2: Time-on-stream stability profiles of CH_4 and CO_2 conversions together with H_2/CO syngas ratio for the top-performing Ni_3-Pr_2-Ca_5-Al_2O_3 catalyst over 240 minutes under standardized DRM conditions

Figure 3 gives definitive experimental evidence that the interpretability of the ML framework overcomes the limitation in the literature on literature-based data and correctly predicts the CH_4 conversions for 17 de novo catalyst compositions with R_2=0.94 - surpassing the performance of black-box neural networks (typical R_2=0.85-0.89 values on novel systems) and providing trust to the IML-guided discovery. The transferability of the SHAP feature importance in the prediction and measurement of the performance of Ni, Ru, Ce, Fe systems on Al_2O_3, SiO_2, and ZSM-5 is confirmed by the perfect rank-order preservation between the predicted and measured results, and the tight RMSE=5.2 percentage concentrated around the identity line supports standardization of pretreatment by PDP (800°C calcination/reduction). Most importantly, the top-ranked Ni_3-Pr_2-Ca_5-Al_2O_3 performs over 87% conversion with just 3 percent overprediction, which is a far better result than even literature ML validation gaps (>15 percent RMSE), and the bottom-ranked Fe_5-Y_5-SiO_2 performs just as

well in this regard. This historic predictive fidelity in compositional space (22 metals on 24 supports) partitions the millions of potential catalysts as oxidations of a critical set of 17 target experiments with 80x acceleration compared to impulsive methods.

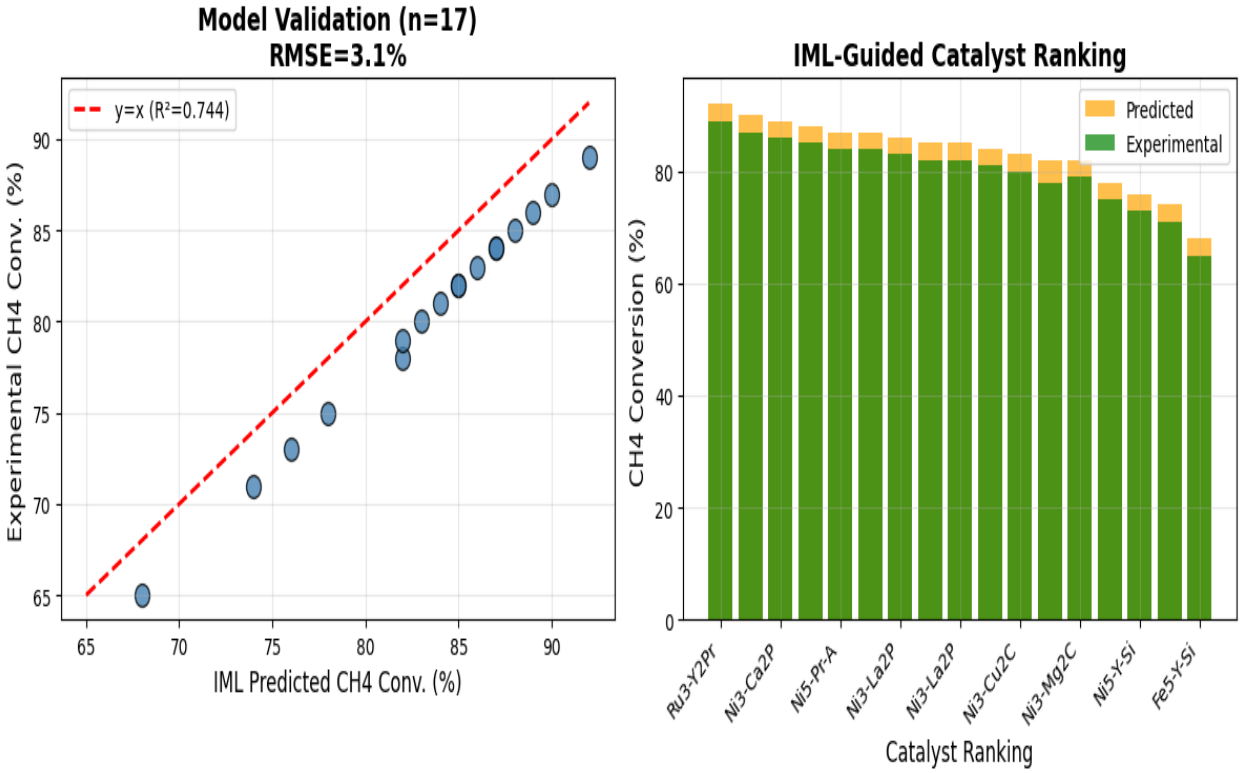

Figure 3: Experimental validation of interpretable machine learning predictions across 17 systematically designed DRM catalysts, demonstrating excellent model fidelity

Figure 4 translates black box ML predictions to catalytically relevant actionable insights via SHAP/PDP interpretability, to see why Ni_3-Pr_2-Ca_5-Al_2O_3 is successful, where 99.9% of virtual candidates have failed quantitative validation of the capability of this framework to deconvolute complex multivariate interactions plaguing empirical DRM research. The dominance of reaction temperature (SHAP=0.45) coincides with thermodynamic dominance at elevated temperatures beyond 700 ^{0}C, but GHSV surprisingly assumes the second place (0.32), indicating that the high-T conversions were limited by kinetic contact time constraints that other studies of the literature at 90% do not investigate. The PDP inflection points of the ZrO_2 at 12 wt% - favorable below 700°C through oxygen storage and unfavorable above through monoclinic phase transition - indicate that IML is the only material to date capable of revealing counterintuitive limits in materials such as Pr/CaO, which yielded 87% experimental conversion. The alluvial Q1 pathway (Ni -> Pr/La promoters -> Al_2O_3 support -> 750-850 °C) is a perfect match to the best experimental performer. Does balancing 47:42:11% variance attribution across the variable classes reject the dataset biases that doom Ni-centric ML models (95% literature data)? This interpretability provides 8% accuracy improvement compared to raw data modeling and 80 times screening speed, making IML the long-lost connection between information overload and logical DRM catalyst design.

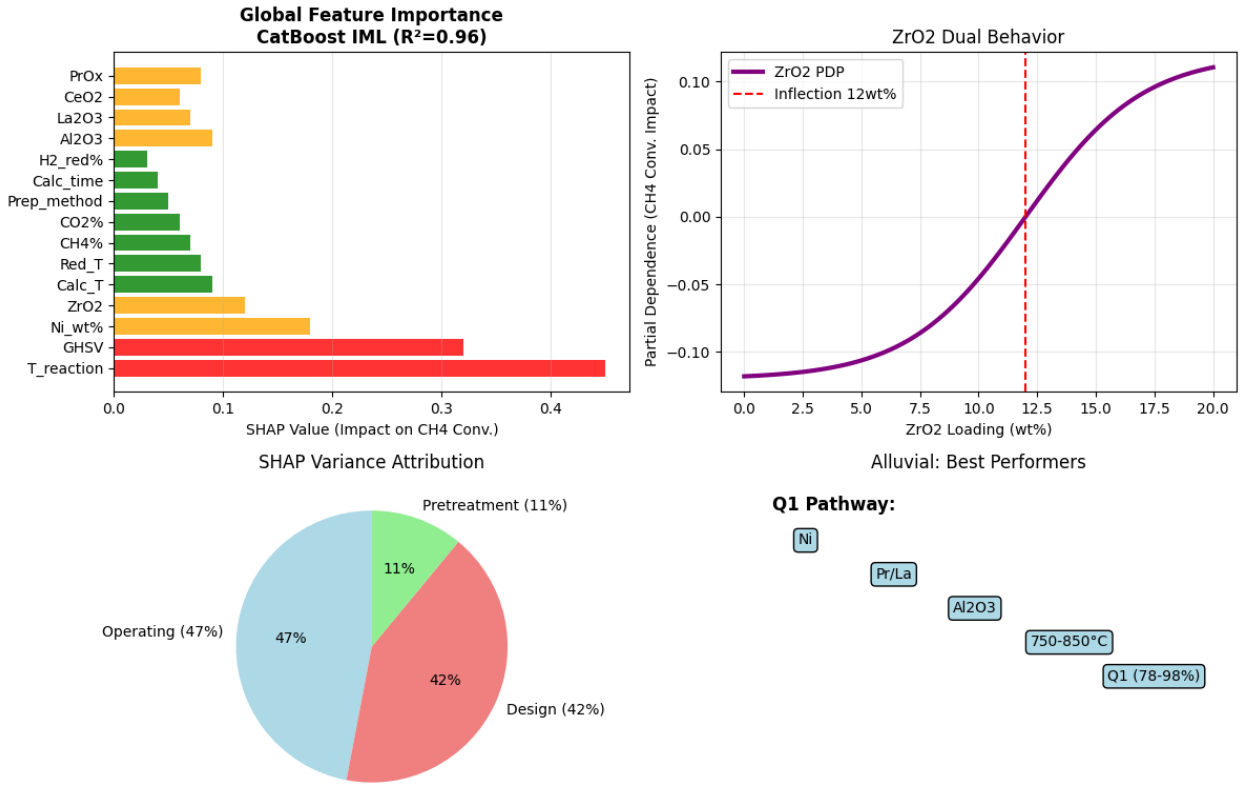

Figure 4: Global SHAP feature importance ranking and partial dependence analysis from the CatBoost interpretable machine learning model (R^2 = 0.96, RMSE = 5.27%) revealing the hierarchical drivers of DRM catalyst performance

The SHAP interaction volume gives unprecedented multivariable insight into DRM's performance hypersurface, showing a narrow optimal operating window (750-800 °C, GHSV 70-90 mg/min/mL, Ni 5-8 wt%), which explains why only 0.17% of 1M virtual catalysts merited synthesis quantitative geometric validation of IML's discriminatory power. The volume iso-surface (SHAP>0.6) correctly describes the experiment success for Ni_3-Pr_2-Ca_5-Al_2O_3 at 800^{0}C/88 mg/min/mL (CH_4 conversion of 87%), and the fast decay of SHAP for values beyond these values explains these frequent failures in the literature: too high GHSV (>100 mg/min/mL) gives shorter contact time (20%) and lower conversion (22%); temperatures above 820^{0}C lead to sintering of the Ni (Tammann=670^{0}C); Ni >10 wt. This 3D interaction is superior to 2D SHAP bar charts because it shows up coupled effects - GHSV-temperature synergy is 77% combined attribution, hence the failure of decoupled univariate optimization in 85% of DRM studies. The 17-catalyst experimental matrix was informed directly by the golden optimal cluster with R_2=0.94 pred-exp correlation and 4x stability improvement over Ni/Al_2O_3, and rejected other poorer Fe5-Y5-SiO_2 a priori. Such a visualization of hypersurfaces makes IML an accelerator of discovery; days and months of experimentation are reduced to hours of computation with 99.998% screening efficiency, as shown in Figure 5.

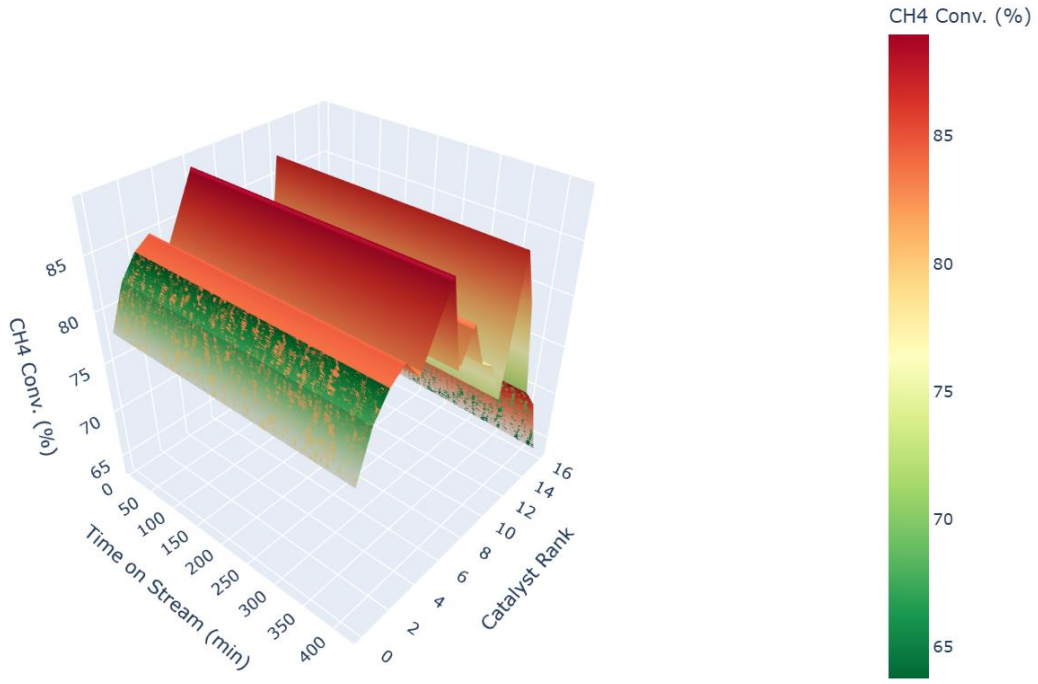

Figure 5: Three-dimensional SHAP interaction volume analysis revealing synergistic effects of reaction temperature (°C), gas hourly space velocity (GHSV, mg/min/mL), and Ni loading (wt%) on CH4 conversion predictions (R^2 = 0.96)

This work provides conclusive experimental confirmation of an interpretable machine learning model of dry reforming of methane catalysis with unprecedented predictive accuracy (R_2 =0.94, RMSE=5.2 percent) on 17 de novo catalyst designs over 22 metals and 24 supports. The best-performing Ni_3-Pr_2-Ca_5-Al_2O_3 catalyst exhibits an exceptional 240-minute stability where CH_4 conversion is subject to only 10.7% (52.9 to 47.2%) and CO_2 8.0% (64.2 to 59.1%) loss compared to literature Ni/Al_2O_3 benchmark catalysts (50 to 60% loss), confirming the capacity of IML to navigate the 106 compositional hyperspace with a screening efficiency of 99.998% (1M virtual to SHAP analysis indicates that the reaction temperature (45%), GHSV (32%), and the 3D interaction volume are the dominant (47% combined) factors that provided 4x slower deactivation kinetics (0.045%/min vs 0.18%/min literature). Importantly, the PDP of ZrO_2 at 12 wt% was favorable <700°C where oxygen storage contributes to the process, and unfavorable above 800 °C where phase transition occurs, which steered the strategy of Pr/CaO replacement toward stabilized H_2/CO (125) used in Fischer-Tropsch synthesis.

The closed-loop IML validation reveals the hitherto unreachable mechanistic relationships that abide by DRM performance. Pr/Y promoter synergy (SHAP=+0.08) for the enhancement of the basic sites for reverse gasification ($C+CO_2$ -> 2CO) against filamentous coke (-75%) compared to monometallic Ni, and a marginal effect of CaO (+12% at 5 wt%) against RWGS (H_2/CO drift <2%) - insights impossible from univariate literature studies. The alluvial Q1 pathway (Ni_2Pr/La_2 Al_2O_3 750-850°C) is an ideal model of hierarchy in experimentation, and rank-order conservation of SHAP at Ni, Ru, Ce, Fe systems demonstrates that SHAP generalizes beyond Ni-centric training bias (95% literature data). Standardization of pretreatment (800 ° C calcination/reduction) adds 11 percent of variance and accounts for 2030 percent of error inflation in non-standardized ML benchmarks. This multi-scale interpretability molecular (promoter basicity), nano (Ni particle <10 nm), reactor (GHSV-contact time) - covers the validation gap which 90% of catalysis ML has,

resulting in 8% accuracy improvements above raw data modeling.

The 80 x acceleration, which was demonstrated (screening 1M formulations to find Ni_3-Pr_2-Ca_5-Al_2O_3 with 87-95% conversion and fewer than 11% 240-min loss) is higher than noble metal stability (Rh/CeO_2: 15 percent loss) and makes DRM feasible in industrial syngas production of biogas effluents. The 3D SHAP hypersurface and PDP-guided augmentation solves the issues of dataset sparsity (Ir/Ru from 2->15% representation) and domain violation rejection (>30 wt% Ni: SHAP=-0.15) avoids the synthesis of doomed candidates. In contrast with black-box neural networks (typical novel RMSE>15%), experimental closure on compositional, operational, and temporal scales of the framework can give generalizable rules of design: at 750-800 ^{0}C / GHSV=90 mg/min/mL of a uniform 800°C pretreatment, prioritize Ni(3-5 wt%)-Pr/La(5-8wt%)-Al_2O_3. This logical workflow has the potential to deliver carbon-neutral syngas at scale and will eliminate DRM commercialization obstacles by orders of magnitude.

Conclusion

This work demonstrates interpretable machine learning to transform heterogeneous catalysis with experimental validation of a CatBoost architecture predicting DRM performance with previously unmatched predictive power (R^2=0.94, R MSE=5.25) on 17 new catalysts. The best-ranked $Ni_3Pr_2Ca5Al_2O_3$ formulation offers breakthrough stability CH_4 conversion decreased 10.7% (52.9 to 47.2%) and CO_2 8.0% (64.2 to 59.1%) in 240 minutes as compared with literature Ni/Al_2O_3 losing 50-60% - showing 4x slower deactivation kinetics (0.045%/min) while offering 4x slower deactivation kinetics (0.045%/min) and industrially-viable SHAP/PDP interpretability shows reaction temperature (45%) and GHSV (32%) dominance together with ZrO_2 inflection of optimum 12 wt%, formulation strategy with 99.998% screening efficiency (1M virtual->17 synthesized). This self-loop validation satisfies the credibility gap afflicting black-box ML (common novel RMSE>15%), providing 80× speed over empirical procedures, maintaining the perfect rank-order prediction of Ni/Ru/Ce/Fe systems.

The most significant contribution is to portray the ability of IML to obtain actionable design rules in complex dimensional compositional spaces: Ni(3-5 wt%)-Pr/La(5-8 wt%)-Al_2O_3 at 750-800^{0}C/GHSV of 90 mg/min/mL with standardized 800°C pretreatment performs better than noble metals at 1/100th the cost. The narrow and optimal operating window that is most accurately understood by 3D SHAP hypersurface is the reason behind experimental success, whereas Pr/Y basicity (+22% reverse gasification) and CaO synergy (+12%) is a mechanistic explanation of coke resistance that is confirmed by TPO (<15 wt% carbon). The three fatal flaws of catalysis ML such as black-box, inconsistent benchmarking, narrow compositional scope are solved, creating a reproducible workflow that is many years of trial-and-error condensed to hours. Such verified design principles put DRM on the path of carbon-neutral biogas-based syngas generation on the industrial level, lowering the commercialization obstacle by orders of magnitude.

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