



An end-to-end framework for reactivity in heterogeneous catalysis

In the format provided by the
authors and unedited

Supplementary Notes

1	CRN blueprint generation algorithm	3
1.1	Molecular templates	4
1.2	Reaction templates	5
2	Adsorbate placement	8
3	GAME-Net-UQ training dataset	10
4	GAME-Net-UQ graph representation	10
5	GAME-Net-UQ uncertainty quantification	13
6	GAME-Net-UQ architecture and training	14
7	GAME-Net-UQ ablation study	16
8	Elementary reaction properties	17
9	Transition state search	19
10	Methanol decomposition case study	20
11	Benchmark against MLIPs	21
12	DFT simulations energy footprint	21

Supplementary Figures

1	CARE blueprint generation algorithm	23
2	GAME-Net-UQ output example	24
3	Comparison between GAME-Net and GAME-Net-UQ	25
4	Transition state graph dataset	26
5	Metals and surfaces in the database of CARE	27
6	DFT transition states frequency validation	28
7	GAME-Net-UQ <i>vs.</i> DFT	28
8	Effect of TAG convolution on transition states	29
9	GAME-Net-UQ performance by system type	29
10	GAME-Net-UQ performance by surface orientation	30
11	Uncertainty quantification in GAME-Net-UQ	31
12	CARE extended chemical space	32
13	Transition state inference with GAME-Net-UQ	33
14	Reaction energy pathways	34
15	C ₁ O ₂ CRN intermediates	35
16	Facet contribution in CO formation rates for methanol decomposition	36
17	CRN evaluation GPU time with GAME-Net-UQ, EquiformerV2, and MACE-MP-0	36
18	DFT <i>vs.</i> GAME-Net-UQ, EquiformerV2, and MACE-MP-0.	37
19	ML-predicted activity for MeOH decomposition case study	38
20	Experimental <i>vs.</i> ML-predicted CO formation rate for MeOH case study	39
21	Reactions and hubs in the electrochemical C ₃ O ₂ CRN	39
22	Energy trends and uncertainty in electrochemical C ₃ O ₂ CRN	40
23	Energy evaluation of C ₆ O ₁ CRN for metals	41

Supplementary Tables

1	Surfaces considered when developing GAME-Net-UQ	42
2	Graph dataset composition for GAME-Net-UQ development	43
3	Training species for GAME-Net-UQ (1)	43
4	Training species for GAME-Net-UQ (2)	44
5	Transition states in the GAME-Net-UQ dataset.	45
6	Energy footprint of DFT simulations	46
7	GAME-Net-UQ performance metrics	46
8	GAME-Net-UQ ablation study	46
9	Energy evaluators in CARE	46
10	DFT <i>vs.</i> GAME-Net-UQ, EquiformerV2-31M, and MACE-MP-0	47
11	Experimental data for methanol decomposition	48
12	Benchmark with MLIPs	48
13	CARE data for methanol decomposition	49
14	GAME-Net-UQ statistics on TS validity	50
15	Top-3 adsorbates from Fischer-Tropsch kinetics on Co, Fe, Ni, and Ru .	50
16	Top-10 products from Fischer-Tropsch kinetics on Co, Fe, Ni, and Ru .	51
17	C-C forming steps from Fischer-Tropsch kinetics on Co and Fe	52
18	C-C forming steps from Fischer-Tropsch kinetics on Ni and Ru	53
19	Overlap GAME-Net-UQ training data <i>vs.</i> applications	54

Supplementary Notes

Note 1: CRN blueprint generation algorithm

The first step for generating a rule-based chemical reaction network (CRN) in CARE is to define the chemical space (CS) and the reactions involved within its boundaries. In our procedure, these boundaries are defined by (i) the carbon and oxygen cutoffs, which represent the maximum number of carbon and oxygen atoms (N_C and N_O , respectively) allowed in a molecule, (ii) by the molecular templates, which define what molecules will be included in the CRN and (iii) by the reaction templates, which determine the type of allowed reactions. Therefore, the completeness of the CRN is defined by the operations permitted through the applied templates.

In heterogeneous catalysis, reactions mostly involve one event at the time, meaning that concerted reactions are scarce [1]. Thus, the defined reaction templates in the algorithm contemplate two main events: bond-breaking/forming reactions and adsorption/desorption reactions [2]. Besides these two fundamental reaction types, CARE includes additional reaction templates, 1,2-H shifts and proton-coupled electron transfer (PCET) reactions for modeling electrochemical systems.

CARE is based on four classes that describe and instantiate the types encapsulating the core building blocks of CRNs: `Intermediate`, `Surface`, `ElementaryReaction` and `ReactionNetwork`, the latter serving as the primary interface.

Starting from N_C and N_O , molecular templates are applied to generate a first pool of saturated molecules defining the chemical space (CS). The CS is then expanded by applying a set of bond-breaking reaction templates that simultaneously generates new species and reactions, obtaining the extended chemical space (eCS). Then, the additional reactions are added. **Supplementary Fig. 1** provides a flowchart of the CRN generation algorithm.

1.1 Molecular templates

Molecular templates allow CARE to build a molecular backbone containing C and O atoms and then hydrogenating them. This spans an initial set of molecules that are the starting point for the CRN generation. These templates are based on SMILES [3] and RDKit to ensure the obtaining of proper structures.

1.1.1 Alkanes Starting from N_C , the algorithm recursively generates all the unique linear and branched alkanes from C_1 up to C_{N_C} by SMILES manipulation. The strategy consists in the addition of "C" strings to the main backbone chain for linear species while for branched "C(C)" expressions are added. The new branched systems are run through the same manipulation process to reach more complex molecules. Using RDKit, SMILES are then canonicalized and converted into `Chem.Mol` objects.

1.1.2 Ethers Molecules containing up to one ether group are generated by manipulation of alkane objects through the following approach: (i) for each alkane, the unique pairs of neighboring carbons are identified, (ii) for each pair, the bond is removed and an oxygen atom is added in between, generating an ether, which is then added to the pool of molecules.

1.1.3 Epoxides Epoxides generation follows a similar strategy as for the ethers: (i) as in the previous section, the unique pairs of neighboring carbon atoms with a lower degree than 4 are identified, (ii) for each pair, an oxygen atom is added in between and two C-O bonds are defined, one for each carbon.

1.1.4 Addition of alcohol groups Addition of $-OH$ groups up to N_O is done for the pool of molecules obtained from the application of the previous templates employing the following strategy: (i) For each molecule in the pool of molecules, carbon atoms with at least one free bonding site (calculated as 4 minus the atom's degree) are identified, (ii) all possible combinations of oxygen addition up to N_O to the suitable carbons of the molecule are generated, (iii) for each combination, $-OH$ groups are added to the

carbon atoms and new alcohol derived species are generated and stored in the pool of molecules.

1.1.5 Addition of reservoir molecules Finally, a small subset of inorganic reservoirs (H_2 , H_2O , CO , CO_2 and O_2) are added *a posteriori* to the pool of molecules through their SMILES representation and converted to objects with RDKit. Oxygen-containing molecules in the reservoir are added accordingly to the defined N_{O} parameter (*i.e.*, if N_{O} has a value of 1, only the molecules with one or none oxygen atoms are added).

1.2 Reaction templates

1.2.1 Bond-breaking reactions CARE employs RDKit to generate the extended chemical space (eCS, or set of species **S**) and bond-breaking reactions. Applying this template to the chemical space (CS) results into the creation of all the possible bond-breaking elementary reactions, and the addition to the CS of their products, including both open- and closed-shell molecules. Specifically, unsaturated molecules like alkenes and alkynes are generated within this template. The implementation of this template goes through the following steps:

- (i). **Template for bond-breaking events:** A set of rules for bond-breaking events are defined as bond types including C-C, C-H, C-O, O-H, O-O, and H-H bonds. Two sets, one for the produced fragments and one for the unique reactions are generated.
- (ii). **Recursive check:** For the given species, the corresponding SMILES representation is checked to verify if it has already been processed avoiding redundancy.
- (iii). **Identification of unique bonds:** Species is then sanitized with RDKit and stereo-chemistry is assigned. Symmetry rules are applied to identify unique bonds and avoid repeating equal events.

- (iv). **Molecule modification:** For each bond type, the specific bond is removed, forming two new fragments.
- (v). **Fragment processing:** The SMILES representation of each fragment is obtained and added to a list. If a fragment has not been processed previously, it is added to a processed fragments dictionary to keep track of all unique fragments generated.
- (vi). **Recursive Bond-Breaking:** The algorithm then recursively calls itself with each fragment as the new species to break further bonds within these smaller structures.
- (vii). **Reaction recording:** For each bond-breaking process, the function constructs a reaction tuple, capturing the transformation from the original species to the resulting fragments and the bond broken. The information regarding the split bond is later used by GAME-Net-UQ to label and distinguish the transition state graphs.

The **eCS** is then labeled using InChIKey [4] and stored in a dictionary. These identifiers have a standard, fixed length, which is a characteristic that can be advantageous for data storage and manipulation. The use of this nomenclature is intended to aid in the organization and processing of large datasets, with a design that may support the effective handling of complex data.

Intermediate instances are then generated for each compound in the chemical space and stored in a dictionary. All systems are labelled as adsorbed species, and a second gas-phase instance is created for species satisfying the closed-shell property. The intermediate identifiers in CARE are the InChIKey plus an extra phase-identifier character, “*” for adsorbed species (i.e., “CURLTUGMZLYLDI-UHFFFAOYSA-N*”) and “g” for gas-phase molecules (i.e., “MHAJPDJPQMIIY-UHFFFAOYSA-Ng”).

ElementaryReaction instances are generated as follows: (i) for each bond-breaking event, the bond-breaking type is retrieved and **Intermediate** instances for reactants and products are accessed through their InChIKey, (ii) the reaction components are defined as a 2-items tuple, one for the reactants and one for the products, (iii) if for

a reactant, two products are generated, an active site is added to the reactants list as a dummy object and (iv) with the components of the reaction and the bond-breaking type defined, the `ElementaryReaction` instance is then generated.

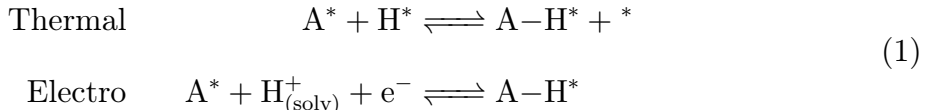
1.2.2 Adsorption reactions CARE includes a reaction template for adsorption/desorption steps. Gas-phase molecules are accessed in the `Intermediate` dictionary and both gas-phase and adsorbed instances are identified. As for the reaction components, the reactant is the gas-phase species plus an active site, and the product is the adsorbed species. Dissociative adsorption for molecular hydrogen and oxygen is included. In this case, the definition of the reaction components is slightly different: the reactant species are defined as before, but for the products the `Intermediate` instances of the individual adsorbed hydrogen or oxygen atoms are added. The reaction type is also labelled as "adsorption" and the `ElementaryReaction` instance is generated.

1.2.3 Rearrangement reactions Expanding the set of templates is feasible. As example, CARE includes [1,2]-H shifts. The template for [1,2]-H shifts is based on SMILES manipulation and the `ElementaryReaction` instances are labelled as "rearrangement". The procedure is as follows: `Intermediate` instances for all adsorbed species are accessed and grouped in a dictionary by molecular formula. For each group with matching formula, using RDKit new subgroups of species are generated based on constitutional isomerism. For each subgroup of isomer families, all possible pairs of species are generated and the algorithm then checks for each pair if a [1,2]-H shift is permitted by SMILES analysis. If the only difference in representation is a neighbouring hydrogen between heavy atoms, an instance of the `ElementaryReaction` is generated.

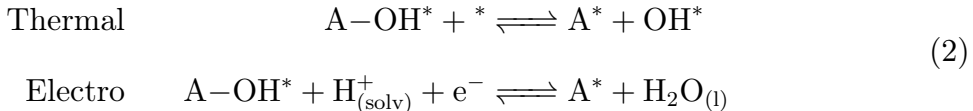
1.2.4 Electrochemical reactions The Computational Hydrogen Electrode (CHE) model [5] enables operating under electrochemical conditions in CARE. To include proton-coupling electron transfer (PCET) steps, a formalism is applied by defining a set of templates for the elementary reactions involving X-H and X-O(H) ($X = C, O$)

bond-breakings, depending on pH, applied potential, and temperature.

The template for these reactions, labeled as “PCET”, requires the definition of two **Intermediate** child-classes for the proton source: **Proton** and **Water**, as well as a specific class for the electron (**Electron**). These classes include information such as the relative charge and phase of the species, labeled as solvent (“*solv*”) or “*electro*” for the electron. For reactions involving X-H (X = C, O) bond-breakings, their **ElementaryReaction** instances are accessed and the adsorbed hydrogen is replaced with a pair of a **Proton** and an **Electron**. A general case showcasing the formalism adaptation is provided below.



Similarly, reactions involving X-O(H) (X = C, O) bond-breakings are updated. The free active sites are replaced by a **Proton**/**Electron** pair and the adsorbed OH is replaced with a **Water** species:



Once all the chemical species, **S**, and elementary reactions, **R**, are generated, these are given to a **ReactionNetwork** instance, which works as an interface for the CRN.

Note 2: Adsorbate placement

For the adsorbate placement to evaluate species with GAME-Net-UQ, a streamlined version of DockOnSurf [6] and ACAT [7] are employed. First, the catalyst slab is adjusted to fit the size of the adsorbate ensuring the minimization of overlapping/lateral interactions. To do so, the maximum length between atoms from the adsorbate is compared with the shortest side of the slab. If the adsorbate length is greater than the slab side, the supercell is then extended laterally.

Second, both the surface and the adsorbate are analyzed to identify their potential anchoring centers. ACAT is used to identify unique anchoring points (top, bridge, hollow, *etc.*) on the surface. Adsorbate anchoring centers are defined as non-saturated atoms and identified through graph-theory with NetworkX. For fully saturated species, their heavy atoms are selected as anchoring centers. For the particular case of carbon monoxide, only the carbon atom is selected.

Adsorption height is then defined, based on heuristics depending on the adsorbate size: 2.0 Å for species containing 10 or more atoms, 1.5 Å for adsorbates containing from 2 to 10 atoms and $x_{\text{site}} + r_{\text{surface atom}}$ for adsorbing single atoms. For the first case, given the size of the adsorbates, the concept of "active sites" is blurred, therefore the procedure is the following: coordinates of all surface anchoring atoms are retained and a mean coordinate is obtained, acting as the anchoring point. For the other cases, mean coordinates are generated for each potential anchoring center and all centers are evaluated separately. Similarly, this procedure is also applied for all adsorbate species containing more than two potential anchoring centers. Then, DockOnSurf is run, generating a large set of adsorbed configurations for each system which is added to the `Intermediate` instances as `ads_configs` attributes. Configurations for monoatomic species are generated through ACAT and only one configuration is generated. It is important to mention that these heuristics rules just serve to generate a proper graph representation and that the exact atomic coordinates are not relevant, as the graph representation in GAME-Net-UQ does not encode bond lengths (**Supplementary Note 4**).

The application of heuristic adsorption lengths serves to the generation of guess configurations that can be later evaluated directly by the desired energy evaluator. These pre-defined adsorbate-surface distances work as a minimum threshold of the adsorption height, if the criteria of adsorbing the adsorbate species to the surface is not met (*e.g.*, the distance is too short leading to non-physically allowed configurations), the distance is iteratively increased by 0.1 Å until reasonable surface-adsorbate configurations are generated.

Note 3: GAME-Net-UQ training dataset

The dataset used to train GAME-Net [8] consisting of closed-shell molecules adsorbed on 14 transition metal surfaces, has been extended to include adsorbed fragments and transition state structures. The new DFT dataset contains 232 gas-phase closed-shell molecules, 2554 surface transition states and 13832 adsorbed intermediates, totaling 17228 entries. The transition states represent bond breaking/forming events. Adsorbed intermediates have been simulated with DFT at low-coverage conditions with no lateral interactions. Adsorbed intermediates consist of the closed-shell molecules from the original dataset and open-shell species. The latter include the smallest ones, like C, H, O, CH, CH₂, up to C₃ fragments. A complete list of the included species can be found in **Supplementary Tables 3–4**.

Besides the most stable surface orientations, the second and third most stable surface orientations for each metal have been considered (face-centered and body-centered cubic: (111), (110), (100); hexagonal closed-packed: (0001), (10 $\bar{1}$ 1), (10 $\bar{1}$ 0)). While CARE targets reaction networks for CHO species, GAME-Net-UQ is trained with adsorbates containing N and S. After cleaning the DFT dataset and discarding isomorphic graphs representing structures converged to the same geometry, the final graph dataset includes 12303 items, 20% transition states and 80% intermediates (**Supplementary Table 2**). The transition state dataset used to train and evaluate GAME-Net UQ consists of 2554 C_xH_yO_z bond-breaking/forming reactions (**Supplementary Fig. 4**, **Supplementary Table 5**). All the metals used in GAME-Net are present except for Fe.

Note 4: GAME-Net-UQ graph representation

GAME-Net-UQ adopts a static graph representation for adsorption structures similar to the one employed in Ref. [8]. This includes the adsorbate (intermediate or transition state), the surface atoms directly in contact with the adsorbate (1-hop nearest neighbours), and their neighbours (2-hop nearest neighbours with respect to the adsorbate,

see **Supplementary Fig. 3**). To detect the graph edges (chemical bonds) from the spatial atomic coordinates, atoms must be adjacent in space and their distance must be smaller than a defined threshold.

Adjacency information is obtained with the Voronoi tessellation algorithm, while the threshold is initially defined as the sum of the covalent radii of the two atoms plus a tolerance value of 0.25 Å. Atomic radii are taken from Cordero *et al.* [9] and the values for the metals are multiplied by a factor of 1.25 to help in the detection of adsorbate-surface edges. Since this strategy leads to a small percentage of graphs without adsorbate-surface edges, the scale factor is increased iteratively by 0.2 (1.25, 1.45, 1.65, *etc.*) until detection of adsorbate-surface edges. For DFT geometries representing bond breaking/forming events, the transition state is correctly detected if the adsorbate graph consists of exactly two disconnected components. An edge between the nearest atoms of the two components is then added and labeled as “1”. Transition state graphs have exactly one edge with attribute “1”. All other edges in the graphs are labeled as “0”.

As the new dataset shows more diversity in terms of closed/open-shell nature and surface orientation, the graph representation originally defined for GAME-Net, which is the simplest possible, has been extended. The graph nodes, besides one-hot encoding the chemical element symbol, now include the normalized generalized coordination number, a descriptor capturing the local environment of the atoms in the catalyst slab. The generalized coordination number gCN of an atom i is defined as:

$$gCN_i = \frac{1}{\max(CN)} \sum_{j \in \mathcal{N}(i)} CN_j \quad (3)$$

where $\mathcal{N}(i)$ is the set of neighbor atoms of i , CN is the coordination number of each neighbor, and $\max(CN)$ is the coordination number of the atoms in the bulk, which have the maximum value (12 for fcc and hcp, and 8 for bcc crystal structures). As the gCN value ranges between 0 and $\max(CN)$, the normalized generalized coordination

number $\overline{gCN}$ is encoded in the surface atoms:

$$\overline{gCN}_i = \frac{gCN_i}{\max(CN)} = \frac{1}{\max(CN)^2} \sum_{j \in \mathcal{N}(i)} CN_j \quad (4)$$

The target of GAME-Net-UQ is the DFT scaled adsorption energy $\tilde{E}_{\text{ads}}$:

$$\tilde{E}_{\text{ads}} = E_{\text{tot}} - E_{\text{slab}} \quad (5)$$

where E_{tot} is the DFT energy of the adsorption system (slab+adsorbed species/transition state) and E_{slab} is the DFT energy of the empty slab.

GAME-Net-UQ is trained to predict not only the energy of stable species but also that of transition states of surface reactions E_{TS} , in order to then derive the activation energy E_{act} . In inference mode, the graph representation of transition states is generated from the graph of the adsorbed reactant A^* in the surface reactions $A^* + * \rightarrow B^* + C^*$ always defined in the bond-breaking direction (**Supplementary Fig. 13**) (concerted steps and electrochemical PCET water elimination processes are not considered).

The TS graph differs by the graph of A^* by two main characteristics: i) the Boolean attribute (1=bond involved in TS, 0=bond not involved) of the edge (bond) linking B^* and C^* fragments (**Supplementary Fig. 3**), and ii) if one of the atoms (nodes) participating in the bond cleavage presents no edge with the surface, the closest reactive surface atom (as the atom with lowest coordination number) is identified. Sequentially, an edge between the atom of the adsorbate and the reactive surface atom is added. Therefore, the activation energy is obtained as:

$$E_{\text{act}} = E_{\text{TS}} - E_{\text{IS}} = E_{\text{TS}} - E_{A^*} \quad (6)$$

Regarding the edge attribute in the graph representation, by default all edges have this attribute set to 0. Only one edge is switched to 1 to label the bond being broken in the transition state graphs.

Note 5: GAME-Net-UQ uncertainty quantification

The inclusion of uncertainty quantification in GAME-Net-UQ required the redefinition of the model output and the change of the training loss function. The scaled DFT adsorption energy $\tilde{E}_{ads}$ is returned by GAME-Net-UQ as a normal probability density function (**Supplementary Fig. 2**):

$$p(\tilde{E}_{ads}) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(\tilde{E}_{ads} - \mu)^2}{2\sigma^2}\right) \quad (7)$$

defined by the mean μ and the standard deviation σ quantifying the model uncertainty. The actual output of GAME-Net-UQ is the tuple (μ, σ) . The original GAME-Net can be thought as its limit case where $\sigma = 0$ for all the inputs (**Supplementary Fig. 3**). Training this model architecture requires the definition of a loss function quantifying accuracy and uncertainty. This means training a model whose output normal distributions are centered as close as possible to the target DFT value, while keeping the variance as small as possible. The negative log likelihood $\mathcal{L}$ is the function of choice for this task:

$$\mathcal{L} = -\log\left(\frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(\tilde{E}_{ads}^{DFT} - \mu)^2}{2\sigma^2}\right)\right) \quad (8)$$

which, after few manipulations, equals:

$$\mathcal{L} = \frac{(\tilde{E}_{ads}^{DFT} - \mu)^2}{2\sigma^2} + \log(\sigma\sqrt{2\pi}) \quad (9)$$

For mini-batch training, $\mathcal{L}$ becomes:

$$\mathcal{L} = \sum_{i=1}^n \frac{(\tilde{E}_{ads,i}^{DFT} - \mu_i)^2}{2\sigma_i^2} + \log(\sqrt{2\pi} \prod_{i=1}^n \sigma_i) \quad (10)$$

Minimizing $\mathcal{L}$ involves finding a balance between accuracy, represented by the numerator in the first term, and uncertainty, present in both terms. $\mathcal{L}$ can be minimized by lowering σ to reduce the second term of the sum, but since σ appears also as

denominator of the first term, it leads to an increase of the first term.

To assess the quality of the uncertainty estimates on the test set, we provide the values of sharpness Sha , variation coefficient c_v , Spearman’s rank correlation coefficient ρ_S , and the calibration curve, following the guidelines from Tran *et al.* [10]. The performance metrics for GAME-Net-UQ are provided in **Supplementary Table 7**.

$$Sha = \sqrt{\frac{\sum_{i=1}^n \sigma_i^2}{n}} \quad (11)$$

$$c_v = \frac{\sqrt{\frac{\sum_{i=1}^n (\sigma_i - \bar{\sigma})^2}{n-1}}}{\bar{\sigma}} \quad (12)$$

with $\bar{\sigma}$ being the mean standard deviation across the n test data points. To quantify the model calibration, the normalized residual $\hat{e}_i$ is first obtained for each test data:

$$\hat{e}_i = \frac{\tilde{E}_{\text{ads},i}^{\text{DFT}} - \mu_i^{\text{model}}}{\sigma_i^{\text{model}}} \quad (13)$$

with $\tilde{E}_{\text{ads},i}^{\text{DFT}}$ being the DFT scaled adsorption energy (our ground truth), μ_i^{model} and σ_i^{model} the predicted mean and standard deviation of graph i . The miscalibration curve is obtained by plotting the distance between the cumulative distribution functions (CDFs) of the normalized residuals and the unit normal distribution (**Supplementary Fig. 11**). The miscalibration area (MCA) is the area between the ideal and observed calibration curve.

Note 6: GAME-Net-UQ architecture and training

The architecture of GAME-Net-UQ implemented in CARE is similar to the one of the original GAME-Net. Input graphs are represented by the node feature vectors, each of them being a 20-dimensional array (14 metals + C, H, O, N, and S, and the normalized generalized coordination number), and by the graph connectivity in coordinate format. To transform the graph representation into the predicted DFT scaled adsorption energy $\tilde{E}_{\text{ads}}$, the following transformations are applied in the listed order:

- (i). Each node feature vector is first transformed *via* one dense layer, increasing the dimensionality of the vector from 20 to 192, $\mathbb{R}^{20} \rightarrow \mathbb{R}^{192}$.
- (ii). Three GraphSAGE [11] convolutions are applied sequentially to all the nodes to capture the information from the neighbors by exploiting the graph connectivity. $\mathbb{R}^{192} \rightarrow \mathbb{R}^{192}$.
- (iii). A topology adaptive graph (TAG) convolutional layer [12] is defined to capture the information encoded in the broken/formed chemical bond in the transition state graphs. $\mathbb{R}^{192} \rightarrow \mathbb{R}^{192}$.
- (iv). The information embedded in the nodes is finally compressed into a graph-level representation with a revised attention pooling layer, [13], $\mathbb{R}^{N \times 192} \rightarrow \mathbb{R}^2$, with N being the number of nodes in the graph. The 2D output vector defines the mean μ and standard deviation σ of the output normal distribution. The latter value passes through a Soft-Max activation function to enforce its positivity.

The activation function used in the node-level layers is the ReLU. Bias terms in all layers are turned off. GAME-Net-UQ contains 558,761 parameters, and compared to the original implementation (286k parameters), this model has an increased layer depth (192 *vs.* 160), an additional topology adaptive graph (TAG) convolutional layer, and the removal of the bias term in the linear transformations in the pooling layer.

The graph dataset ($n=12303$) has been split between train, validation and test sets (60/20/20 %). Due to the reduced presence of gas closed-shell molecules in the dataset compared to adsorption structures, a 10 $\times$ oversampling for these species is applied in the training set, *i.e.*, each gas phase molecule in the training set appears 10 times. The training dataset contains 8547 graphs, while validations and test sets 2460. Target energies $\tilde{E}_{\text{ads}}$ are scaled *via* standardization using training and validation data. The negative log likelihood (NLL) is employed as training loss function using the AMSGrad optimizer [14]. The learning rate (LR) value is steered with the reduce-on-plateau scheduler. The initial LR has been set to 10^{-3} and is reduced exponentially by the scheduler every time there is no improvement after 5 epochs (patience) in the NLL of

the validation set. The minimum LR possible has been set to 10^{-7} , while the decrease factor has been set to 0.7. In each training 250 epochs are performed. During each epoch, the training set is fed to the model in batches of 32 samples, performing a backward propagation and updating the model parameters after each batch.

The present work used only C, O, and H as atoms in the adsorbates, although GAME-Net-UQ is capable of processing also N and S heteroatoms. GAME-Net-UQ can be fine-tuned and extended in terms of materials applicability with external databases provided as ASE database. Documentation is provided in the Github repository of GAME-Net-UQ at https://github.com/LopezGroup-ICIQ/gamenet_uq.

Note 7: GAME-Net-UQ ablation study

An ablation study to GAME-Net-UQ has been performed to observe how the test performance is affected by (i) removing the topology adaptive graph (TAG) convolutional layer from the GNN architecture, (ii) neglecting the generalized coordination number as node feature, and (iii) removing the 2-hop nearest surface neighbours with respect to the adsorbate from the graph representation. The results of this study are shown in **Supplementary Table 8**. The performance has been evaluated on the same test set considered in the manuscript. For each case, we report the global mean absolute error (MAE) and sharpness (*Sha*), and grouped by simulation type (intermediate/transition state). To minimize nondeterminism-introducing factors, the runs have been performed keeping fixed:

- the hardware (GPU) on which the learning processes have been performed.
- the train, validation, and test dataloaders and the number of workers (equal to 1).
- the order on which training data are fed to model by batches.
- the initial weights of the GNN.

For each combination of ablated features, 5 trainings have been performed.

Note 8: Elementary reaction properties

The elementary reaction properties are derived as normal distributions starting from the predictions obtained with GAME-Net-UQ for the intermediates and transition states. Each elementary reaction, regardless of its direction, has four main properties: (i) the direct reaction energy $\vec{\Delta E}$, (ii) activation energy $\vec{E}_{\text{act}}$, (iii) the equilibrium constant $\vec{K}_{\text{eq}}$ and (iv) the kinetic constant $\vec{k}$. Given these values, the properties of the reverse step (i.e., $\overleftarrow{\Delta E}$, $\overleftarrow{E}_{\text{act}}$, $\overrightarrow{k}$) can be derived. An elementary reaction $A^* + B^* \rightarrow C^* + *$ with normally distributed species' energy $E_i \sim \mathcal{N}(\mu, \sigma^2)$ in CARE is characterized as follows:

$$\vec{\Delta E} \sim \mathcal{N} \left(\sum_{i=1}^{n_S} \nu_i \mu(E_i), \sum_{i=1}^{n_S} |\nu_i| \sigma(E_i)^2 \right) \quad (14)$$

$$\vec{E}_{\text{act}} \sim \mathcal{N} \left(\mu(E_{\text{TS}}) - \mu(E_{\text{IS}}), \sigma(E_{\text{TS}})^2 + \sigma(E_{\text{IS}})^2 \right) \quad (15)$$

$$\overleftarrow{\Delta E} = -\vec{\Delta E} \quad (16)$$

$$\overleftarrow{E}_{\text{act}} = \vec{E}_{\text{act}} - \vec{\Delta E} \quad (17)$$

with ν_i being the stoichiometric coefficient of species i in the reaction, and E a generic energy placeholder, in our case it represents the DFT internal energy at 0K, however the same relations hold if additional contributions (entropy, zero-point energy, etc.) are considered. Additionally, E_{IS} in **Eq. 15** refers to the left-hand side of surface reactions in the bond-breaking direction, due to easier manipulation of the transition state graph. As GAME-Net-UQ allows predicting only the transition state energy of surface bond-breaking reactions (see **Supplementary Note 4** for the generation of the transition state graph for GAME-Net-UQ), adsorption, desorption, and rearrangement steps are considered as barrierless (**Supplementary Figure 14**) and $\vec{E}_{\text{act}}$ collapses to:

$$\vec{E}_{\text{act}} \sim \mathcal{N} \left(\max(0, \vec{\Delta E}), \sigma(E_{\text{FS}})^2 + \sigma(E_{\text{IS}})^2 \right) \quad (18)$$

For the machine learning potentials, the nudged elastic band (NEB) method is

implemented in CARE for computing transition states and activation barriers for bond-breaking reactions on the catalytic surfaces. More details on the implementation of the NEB method can be found in **Supplementary Note 9**.

For electro-catalytic reactions, the computational hydrogen electrode formalism [5] is employed. To include pH and applied potential effects on proton-coupled electron transfer (PCET) steps, reaction energies are modified as follows:

$$\vec{\Delta E} = \vec{\Delta E}_T - n_e(U_{\text{SHE}} + 2.303k_B T pH) \quad (19)$$

where $\vec{\Delta E}_T$ is the thermal reaction energy in [eV], n_e the stoichiometric coefficient of the electron involved in the step, and U_{SHE} the applied potential with respect to the Standard Hydrogen Electrode (SHE) in [V]. The equilibrium and kinetic constants are eventually defined as:

$$\vec{K}_{\text{eq}} = \exp\left(-\frac{\vec{\Delta E}}{k_B T}\right) \quad (20)$$

$$\vec{k} = \frac{k_B T}{h} \exp\left(-\frac{\vec{E}_{\text{act}}}{k_B T}\right) \quad (21)$$

$$\overleftarrow{k} = \frac{\vec{k}}{\vec{K}_{\text{eq}}} \quad (22)$$

Transition state theory is used to calculate the forward kinetic coefficients $\vec{k}_f$ (**Eq. 21**) except for the adsorption steps, whose coefficients are evaluated with the Hertz-Knudsen equation [15]. Kinetic coefficients of the reverse reactions are derived by imposing thermodynamic consistency (**Eq. 22**).

As for each adsorbed species multiple configurations can be screened, to choose which value of E_i is used for evaluating the reaction properties ΔE and E_{act} , the user has two options when using GAME-Net-UQ as energy evaluator in CARE: (i) Selecting the configuration with lowest uncertainty, or (ii) the configuration with lowest energy.

DFT databases can be employed with CARE to avoid the evaluation of already

available species. Although this strategy leads to minimal computational savings, the gain is a reduced uncertainty and improved accuracy for the reactions involving these species. As an example, using the DFT energy of adsorbed H^* , the main hub in the C_1O_2 chemical reaction network, positively affects the evaluation of 48% of the reactions in the network by reducing their error and associated uncertainty.

Note 9: Transition state search

CARE provides an interface to evaluate the transition state (TS) energy of surface reactions using either GAME-Net-UQ or ML Interatomic Potentials (MLIPs).

Since GAME-Net-UQ is a direct model, it is limited to constructing the TS graph. In contrast, MLIPs require both the initial state (IS) and final state (FS) structures of the reaction. CARE treats surface reactions in the bond-breaking direction $\text{A}^* + * \rightarrow \text{B}^* + \text{C}^*$. The IS structure is obtained directly from the most stable relaxed representation of A^* . For GAME-Net-UQ, the TS graph is constructed by identifying the edge in the IS graph whose removal produces the graphs of the product fragments $\text{B}^* + \text{C}^*$. This is done by iteratively removing all possible edges in the IS graph and performing a graph isomorphism check against the product graphs (**Supplementary Fig. 13**).

For MLIPs, the FS structure is generated from that of the IS as follows:

- (i). Identify the fragment (B^* or C^*) whose center of mass is farther from the surface.
- (ii). Displace this fragment along a vector parallel to the surface and aligned with the bond between the two atoms connecting B^* and C^* .
- (iii). Relax the resulting structure.
- (iv). If the graph of the obtained relaxed structure represents two dissociated adsorbates ($\text{B}^* + \text{C}^*$), it is accepted as FS. Otherwise, the displacement is increased iteratively until the dissociation condition is satisfied.

TS search is performed with the nudged elastic band (NEB) method [16–18] implemented in ASE [19]. CARE uses the following default NEB settings: five images

between the initial and final states, a spring constant of 0.1 [eV/Å], linear interpolation for the initial guess of intermediate images, and the BFGS optimizer.

The transition state is identified as the highest-energy configuration along the minimum energy path. This energy is then used to compute the activation barrier E_{act} . The geometry and energy of the TS are stored in the `ElementaryReaction` instances.

Note 10: Methanol decomposition case study

C₁O₂ CRNs have been generated for each metal and each facet available in GAME-Net-UQ for the methanol decomposition process. The CRN contains 69 elementary reactions where 28 surface species and 10 gas-phase molecules participate. Microkinetic runs have been performed for each CRN with Julia’s `FBDF()` solver. Settings used in the MKM runs depend on the stiffness of the each CRN, however all simulations required values of absolute (`atol`) and relative (`rtol`) tolerances of at least $\approx 10^{-15}$ and $\approx 10^{-13}$ respectively to converge to steady state. **Supplementary Table 13** contains the energy and rate values obtained for each CRN-MKM run. To convert the CO formation rates from the MKM runs $r_{\text{CO}}^{\text{CARE}}$ to the macroscopic rate and space time yield (STY) presented in **Fig. 3c** and **Supplementary Fig. 16**, a unit conversion has been done for the obtained rate values to match the experimental units through the following equations:

$$r_{\text{CO}}[\mu\text{mol}/\text{g}_{\text{cat}}\text{min}] = r_{\text{CO}}^{\text{CARE}}[1/\text{s}] \times \frac{60[\text{s}/\text{min}] \times 10^6[\mu\text{mol}/\text{mol}] \times D[\%] \times wl[\%]}{MW_{\text{metal}}[\text{g}_{\text{cat}}/\text{mol}]} \quad (23)$$

$$\text{STY}_{\text{CO}}[\text{mol}/\text{mol}_{\text{cat}}\text{h}] = r_{\text{CO}} \times \frac{60[\text{min}/\text{h}] \times MW_{\text{metal}}[\text{g}_{\text{cat}}/\text{mol}]}{D[\%] \times wl[\%] \times 100} \quad (24)$$

where D and wl refer to the metal dispersion and loading, respectively, and MW_{metal} the molecular weight of the active metal.

The CO formation rate for each metal catalyst is obtained by aggregating the contribution of all the hkl surface facets obtained from Ref. [20]. The CO formation rates and adsorption energy in **Fig. 3d** and **Supplementary Fig. 19** have been obtained by weighting the values of each hkl facet by their relative percentage surface

area α_{hkl} :

$$r_{\text{CO}}^{\text{CARE}} = \sum_{hkl} \alpha_{hkl} r_{hkl} \quad (25)$$

$$E_{\text{ads,CO}}^{\text{CARE}} = \sum_{hkl} \alpha_{hkl} E_{\text{ads},hkl} \quad (26)$$

Note 11: Benchmark against MLIPs

The performance of GAME-Net-UQ in reproducing the experimental CO production rates in the methanol decomposition case study has been compared to that of two state-of-the-art Machine Learning Interatomic Potentials (MLIPs) available in CARE: EquiformerV2-31M [21] and MACE-MP-0 (15.8M) [22]. The settings for each model have been reported in **Supplementary Table 12**. The ASE calculator interfaces to these models allow structural relaxation and transition state (TS) search. Therefore, the activation energies E_{act} of the surface reactions have been estimated with the CI-NEB method. The results of this experiment in terms of accuracy are depicted in **Supplementary Figures 19-20**, while the computational time required to evaluate the 63 CRNs with GAME-Net-UQ, EquiformerV2-31M and MACE-MP-0 is reported in **Supplementary Figure 17**. Overall, the comparison of the results obtained with these different energy evaluators demonstrates that the direct transfer of existing MLIPs to reproduce experimental rates or trends in heterogeneous catalysis needs to be more extensively benchmarked and for this, sound experimental databases are needed.

Note 12: DFT simulations energy footprint

The DFT simulations used in this work come from multiple sources in terms of user and cluster/hardware where the simulation has been run. We provide a conservative assessment of the CPU time required by VASP DFT simulations to converge, and the related energy consumption. The following steps have been performed:

- (i). Parse the OUTCAR files for each VASP simulation and collect information about computing cluster and number of cores on which the simulation has been run (written on the top of the file), and elapsed time of the simulation t_{elapsed} (bottom of the file). When N_{runs} DFT runs have been necessary to reach the desired convergence criterion, the OUTCAR files of the previous runs have been saved to collect the total elapsed time:

$$t_{\text{elapsed}} = \sum_{i=1}^{N_{\text{runs}}} t_{\text{elapsed},i} \quad [\text{s}] \quad (27)$$

- (ii). Get the number of CPUs N_{CPU} on which the simulation has been run. Given the number of cores per CPU N_{cores} , the CPU time can be estimated as:

$$\text{CPU time} = t_{\text{elapsed}}[\text{h}] \times N_{\text{cores}} \times N_{\text{CPU}} \quad [\text{CPU h}] \quad (28)$$

- (iii). Retrieve the Thermal Design Power (TDP, in Watt [W]) of the specific CPU. This value defines the theoretical maximum amount of heat generated by a CPU that its cooling system is designed to dissipate. We assume here that CPUs work at 100% capacity.
- (iv). Retrieve the Power Usage Effectiveness (PUE) of the computing cluster. This value provides a measure of energy efficiency for data centers, allowing the inclusion of the cooling requirements in the energy balance.
- (v). Get the estimate of the energy E consumed by a single DFT simulation with the following formula:

$$E = \frac{TDP[\text{W}] \times N_{\text{CPU}} \times t_{\text{elapsed}}[\text{s}] \times PUE}{3.6 \times 10^6 [\text{J/kWh}]} \quad [\text{kWh}] \quad (29)$$

Supplementary Table 6 provides average CPU time and power consumption for DFT relaxations of adsorption structures with spin polarization (ISPIN=2, for Fe, Ni, and Co surfaces), without spin polarization (ISPIN=1), and gas molecules.

Supplementary Figures

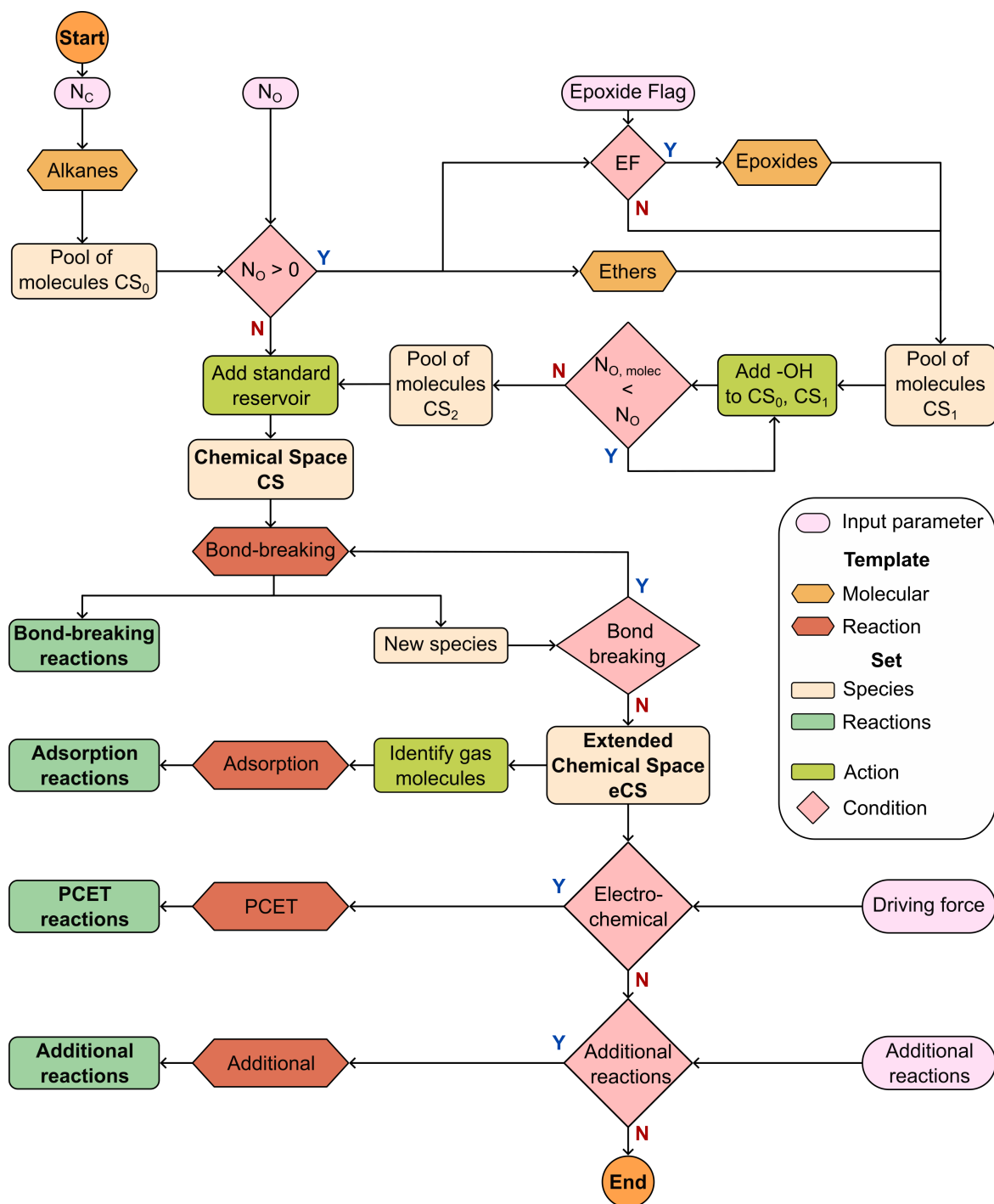


Fig. 1: Algorithm for the generation of the chemical species and reactions in CARE. N_C =Network Carbon Cutoff; N_O =Network Oxygen Cutoff; PCET=Proton-Coupled Electron Transfer.

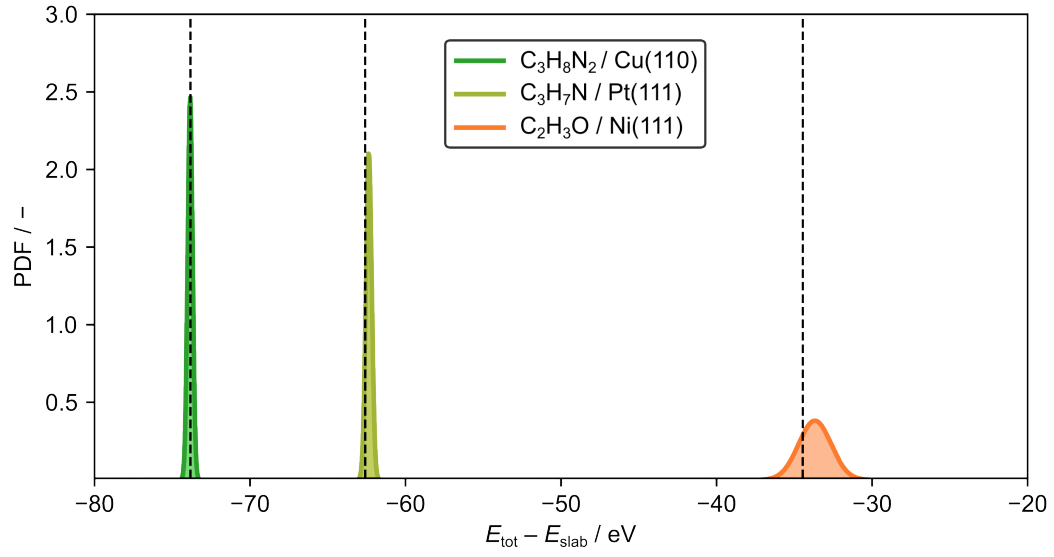


Fig. 2: GAME-Net-UQ example output with three unseen data with increasing uncertainty going from left (dark green) to right (orange). Dashed lines = DFT values. Distributions = GAME-Net-UQ predictions. PDF = Probability Density Function.

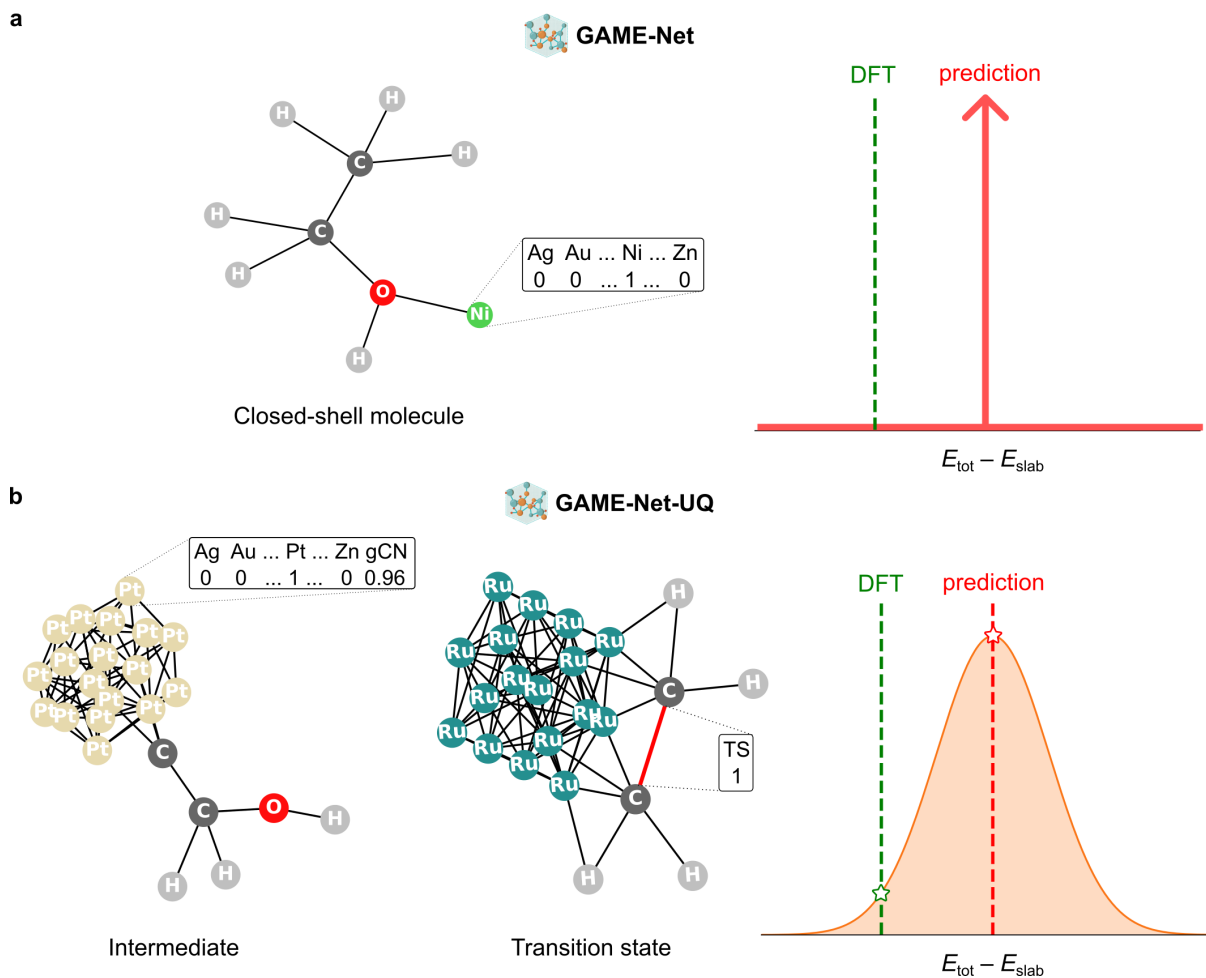


Fig. 3: Comparison between **a** GAME-Net [8] and **b** GAME-Net-UQ in terms of graph representation and output. The red edge in the transition state graph represents the broken/formed chemical bond. gCN = generalized Coordination Number.

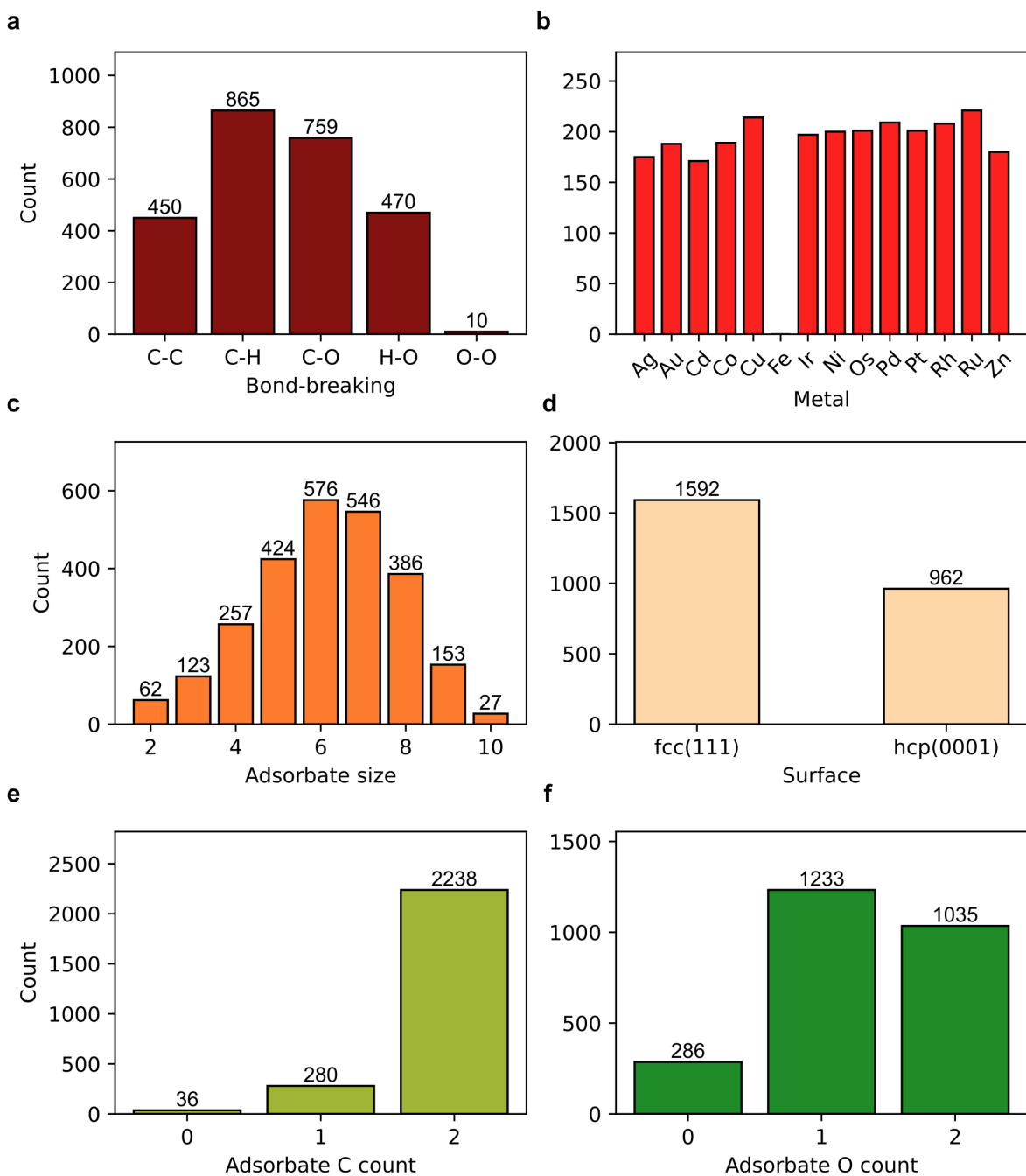


Fig. 4: Overview of the transition state graphs used to train and evaluate GAME-Net-UQ. Count by **a** bond-breaking type, **b** metal, **c** adsorbate size, **d** surface facet, number of **e** C and **f** O atoms in the adsorbate. $n=2554$.

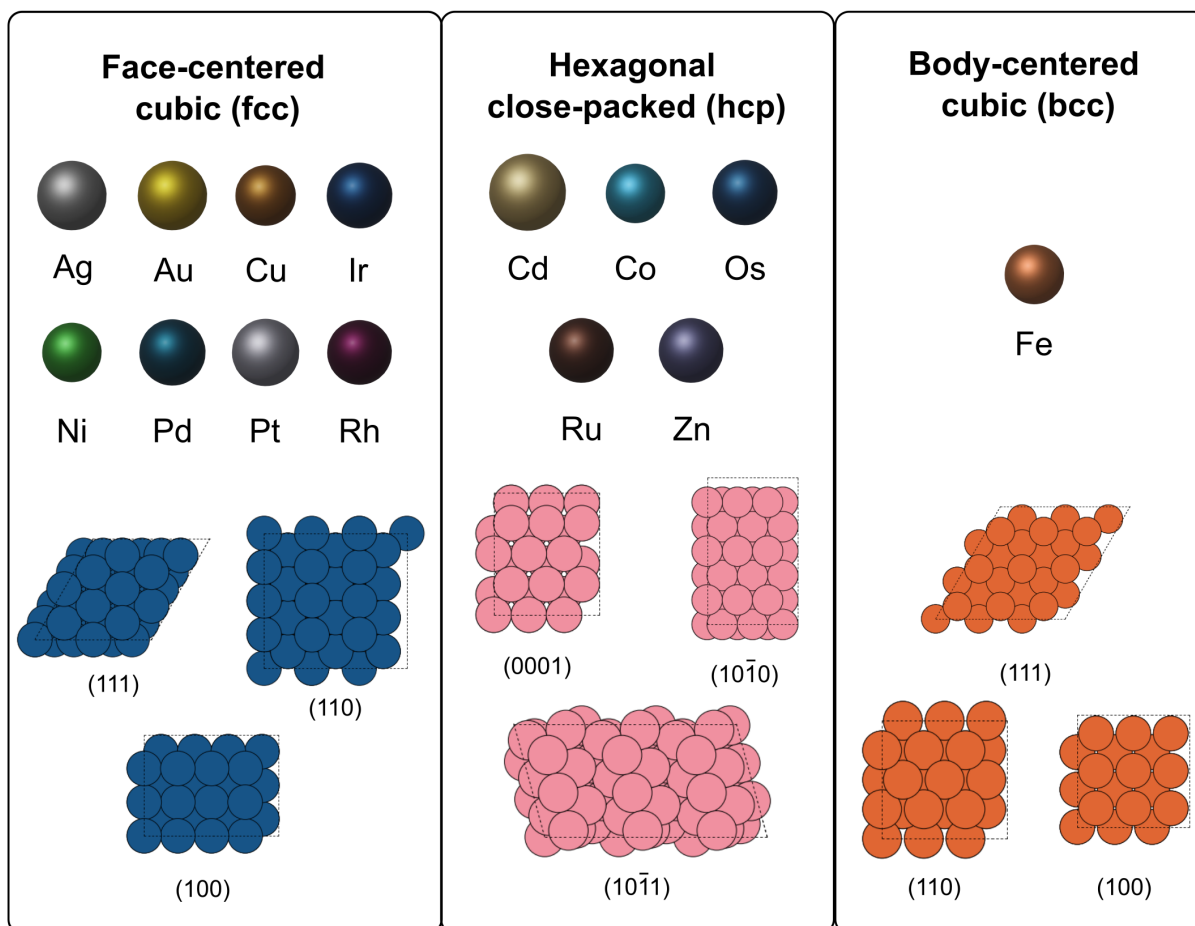


Fig. 5: Metals and surfaces contained in the provided database in CARE grouped by crystal structure. Additional surfaces can be obtained through the API of Materials Project or provided by the user.

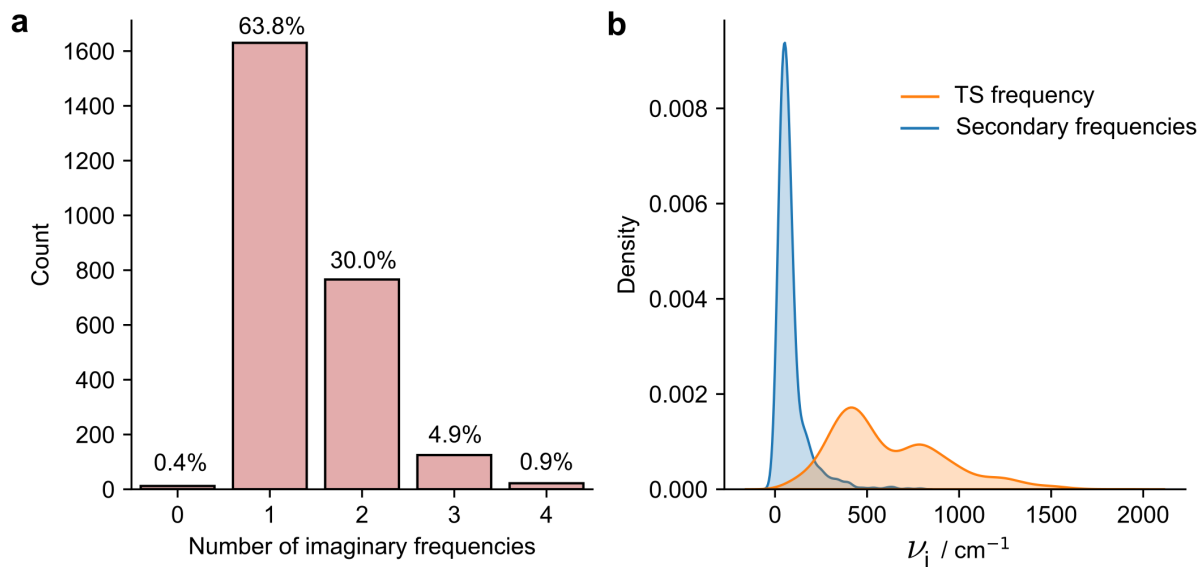


Fig. 6: **a** Histogram of the DFT transition state structures used to develop GAME-Net-UQ grouped by number of imaginary frequencies found *via* frequency analysis. $n=2554$. **b** Distribution of the TS (orange) and secondary (blue) imaginary frequencies.

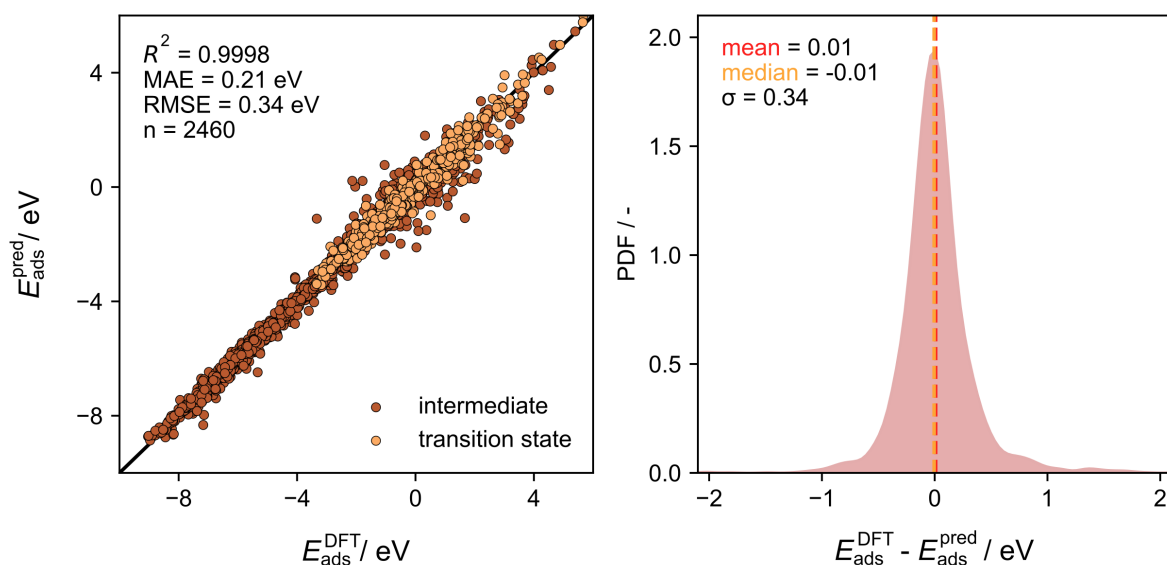


Fig. 7: **a** Predicted *vs.* DFT adsorption energies of the test set grouped by system type and **b** associated error distribution. $n = 2460$, $n_{\text{int}} = 1982$, $n_{\text{ts}} = 478$. PDF = probability density function. std = standard deviation.

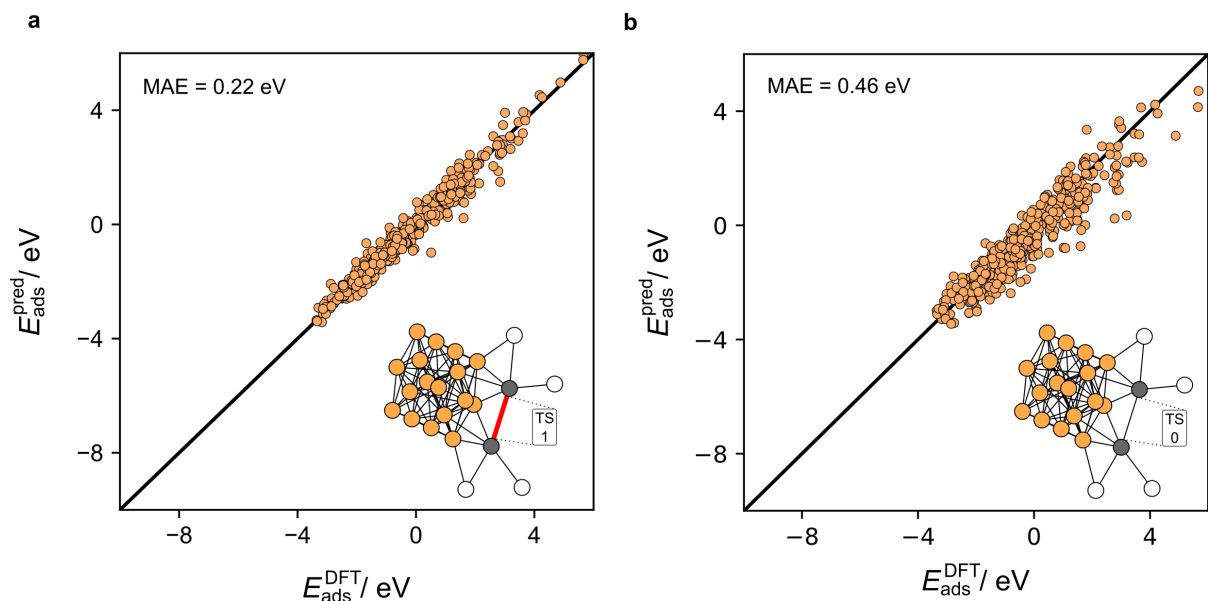


Fig. 8: Predicted *vs.* DFT adsorption energy for the transition state structures in the test set **a** with and **b** without the topology adaptive graph (TAG) convolutional layer used to distinguish transition states in GAME-Net-UQ. Mean absolute error (MAE) in panel **a** refers to the final model implemented in CARE, while the value in **b** stems from the ablation study (Supplementary Note 7). $n=478$.

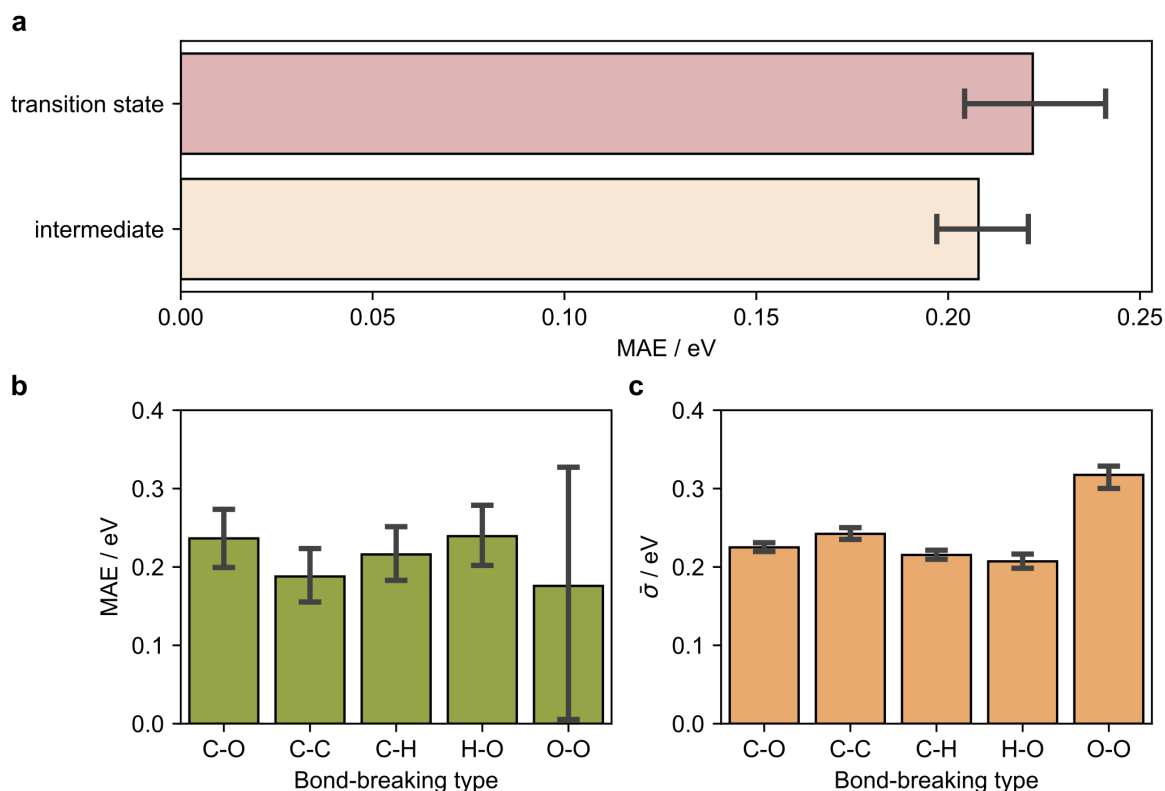


Fig. 9: GAME-Net-UQ test performance by system type. Mean Absolute Error (MAE) bar-plots by **a** system type and **b** bond-breaking involved in the transition state data, together with **c** mean associated uncertainty $\bar{\sigma}$. Error-bars represent the confidence interval at 95% confidence. $n=2460$ (whole test set), $n_{int}=1982$ (intermediates), $n_{ts}=478$ (transition states).

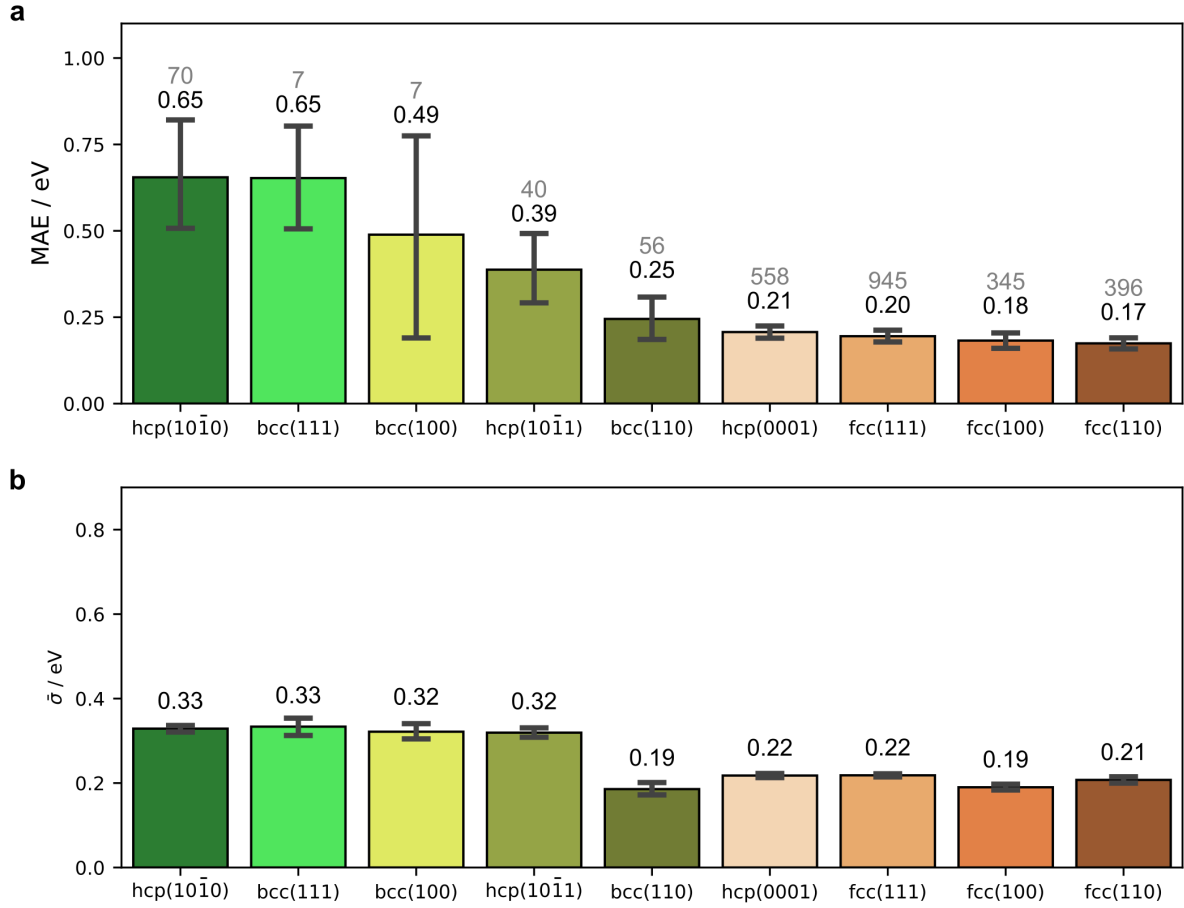


Fig. 10: **a** Mean Absolute Error (MAE) and **b** mean associated uncertainty $\bar{\sigma}$ of the predictions with GAME-Net-UQ on test data grouped by surface orientation. Error-bars represent the confidence interval at 95% confidence. Values on top of the bars in panel **a** are the subgroup size (top) and its MAE (bottom). $n = 2424$ (test set data except gas molecules). fcc=face-centered cubic; hcp=hexagonal close-packed; bcc=body-centered cubic.

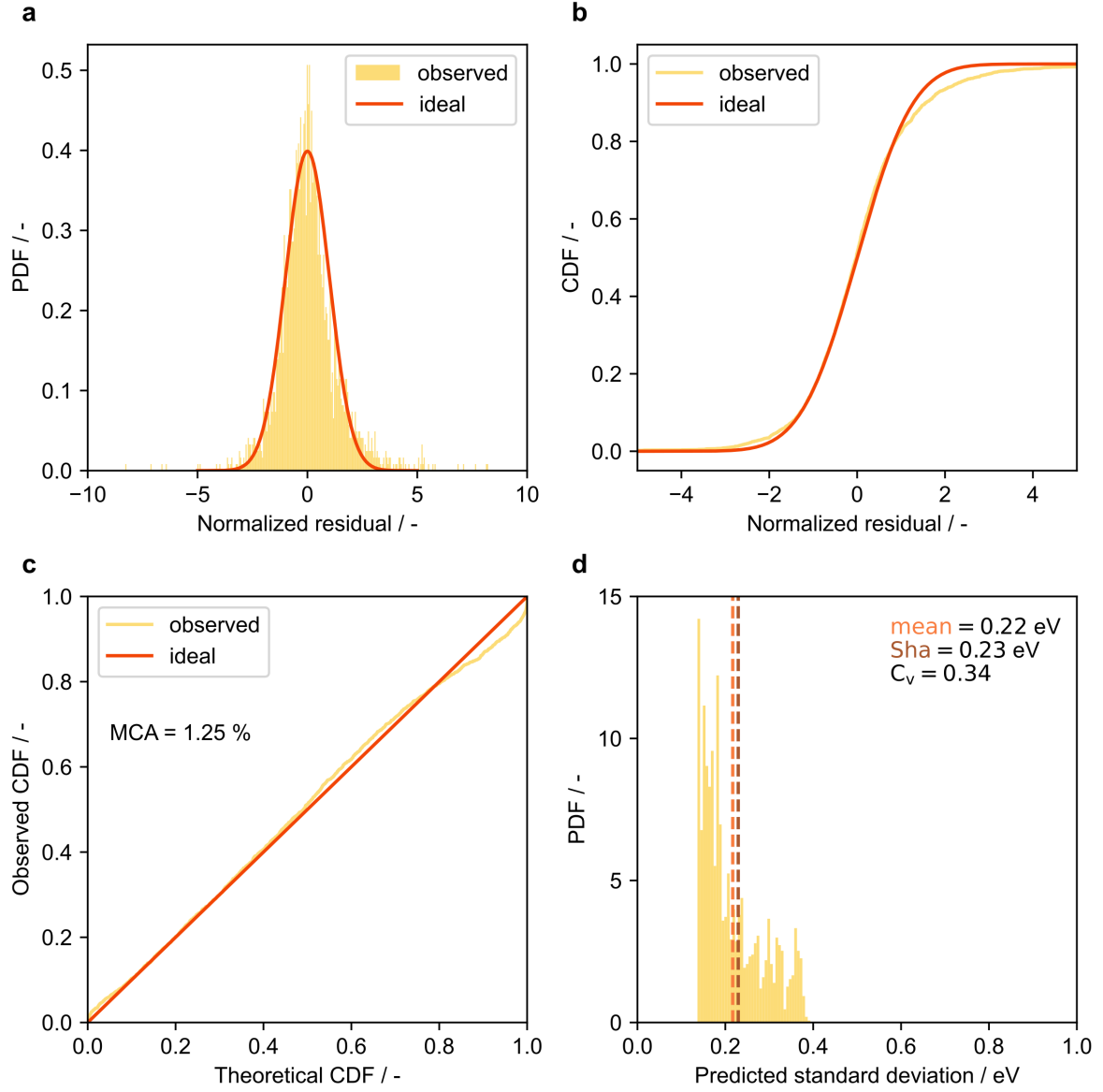


Fig. 11: GAME-Net-UQ uncertainty quantification analysis. **a** Probability density function (PDF) of the normalized residuals on the test set compared to the ideal normal distribution, with **b** corresponding cumulative distribution function (CDF) comparison and **c** derived calibration curve showing the miscalibration area (MCA). **d** Distribution of the predicted standard deviation for the test data, highlighting sharpness (Sha) and mean standard deviation. c_v = Variation coefficient (dimensionless). $n = 2460$. Uncertainty metrics grouped by type of structure can be found in **Supplementary Table 7**.

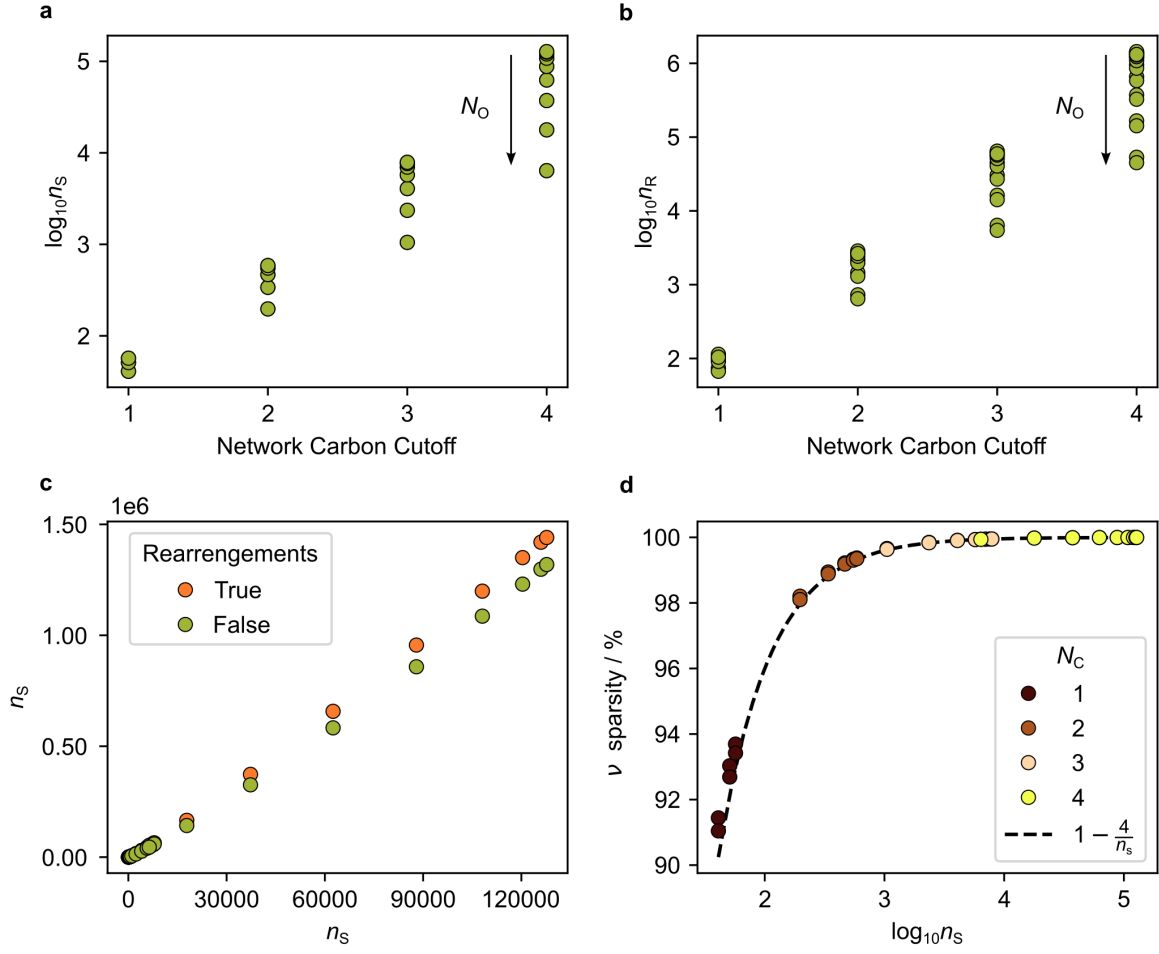


Fig. 12: Quantification of the size of the chemical reaction networks (CRNs) generated with CARE. **a** Number of species n_S and **b** elementary reactions n_R as function of the network carbon and oxygen cutoffs (N_C and N_O), and **c** related scatter-plot highlighting the difference when rearrangement steps are included. **d** Sparsity of the stoichiometric matrix ν as function of n_S . These panels have been constructed by generating the CRN blueprints with $N_C \leq 4$ and all possible values of N_O .

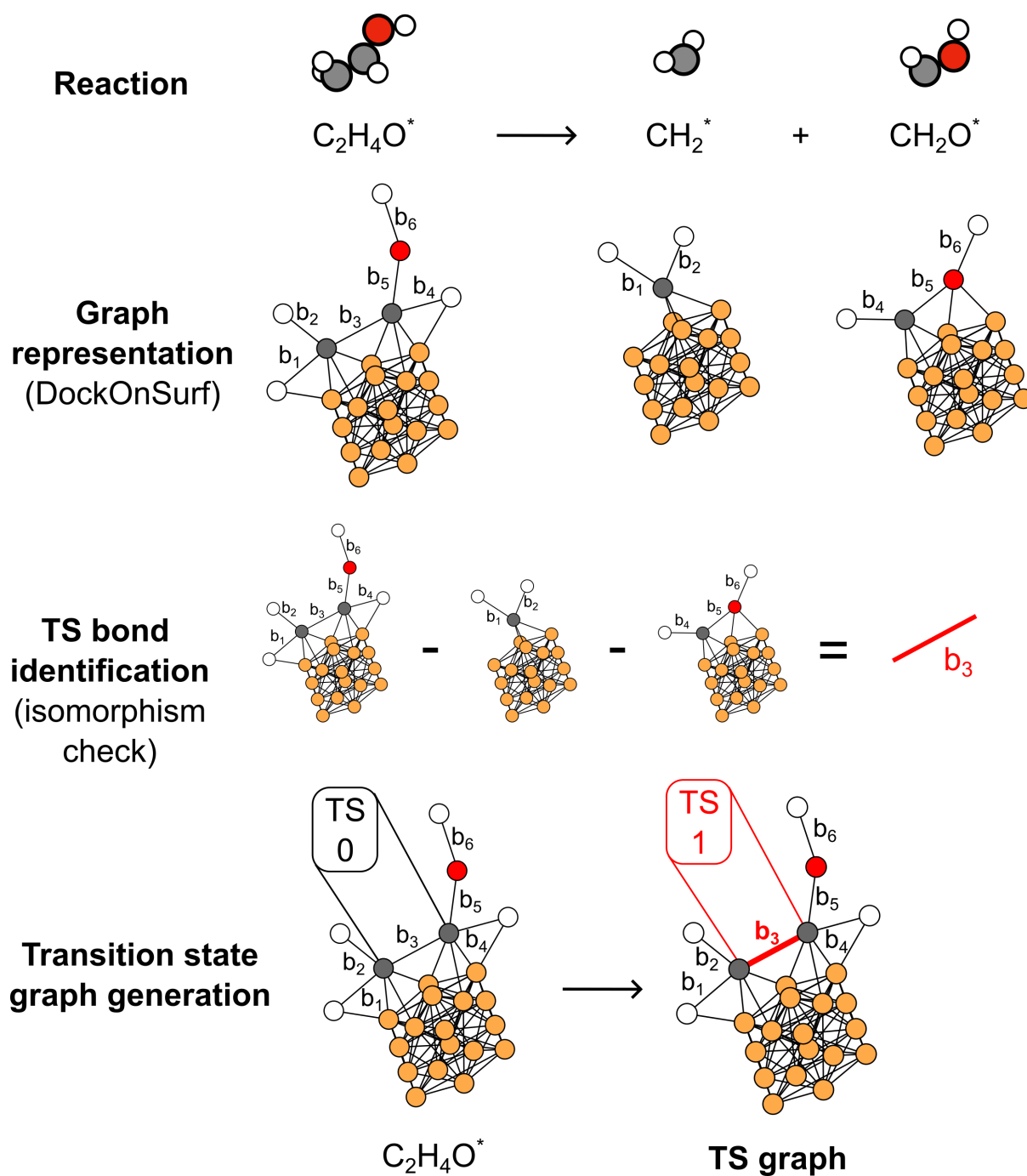


Fig. 13: Scheme of the methodology employed in CARE to generate the graphs representing transition states (TSs) for GAME-Net-UQ (**Supplementary Note 9**).

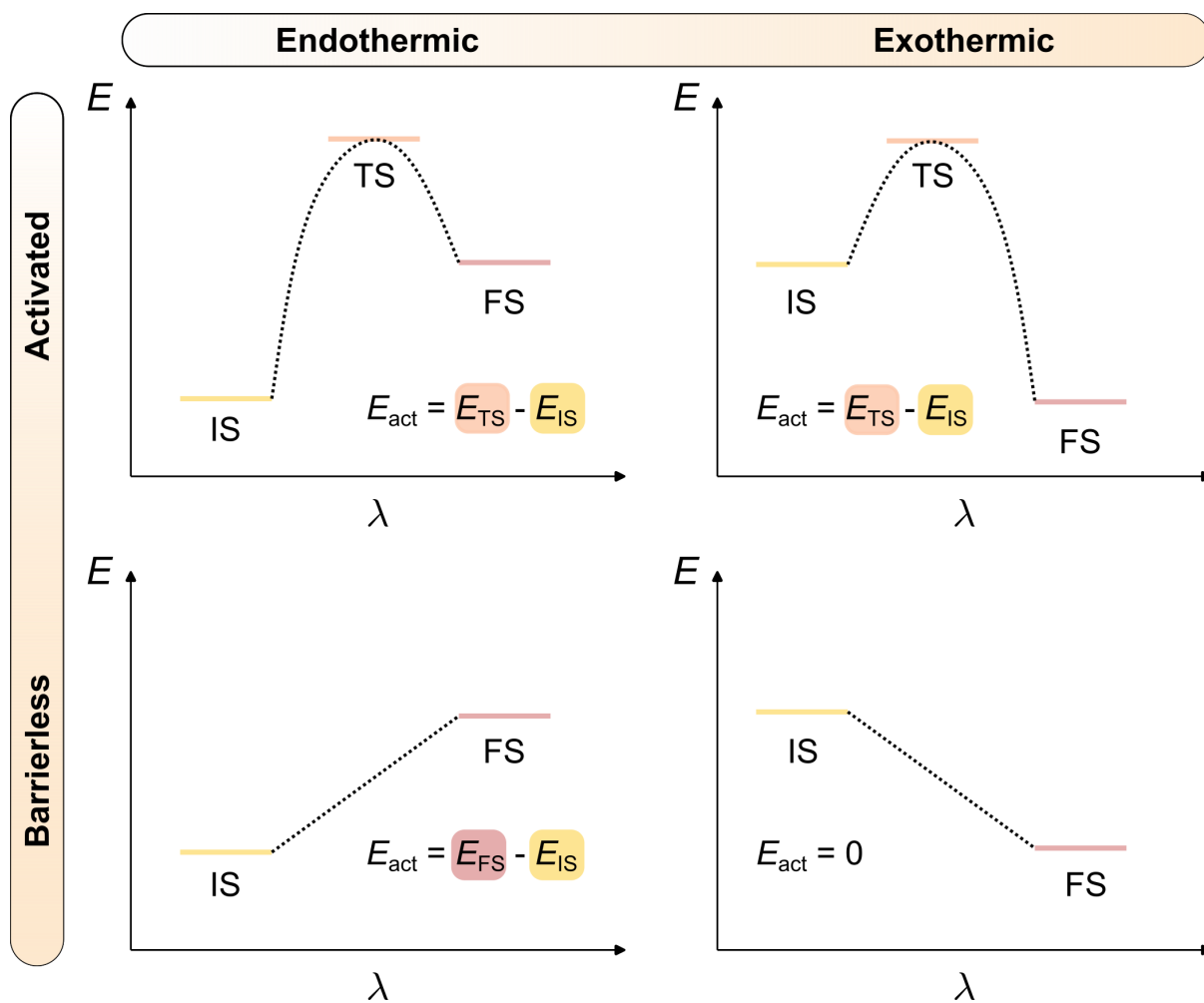


Fig. 14: Overview of the four possible energy paths for elementary reactions as function of activation presence and thermodynamics. E =potential energy, λ =reaction coordinate, IS=Initial State, TS=Transition State, FS=Final State.

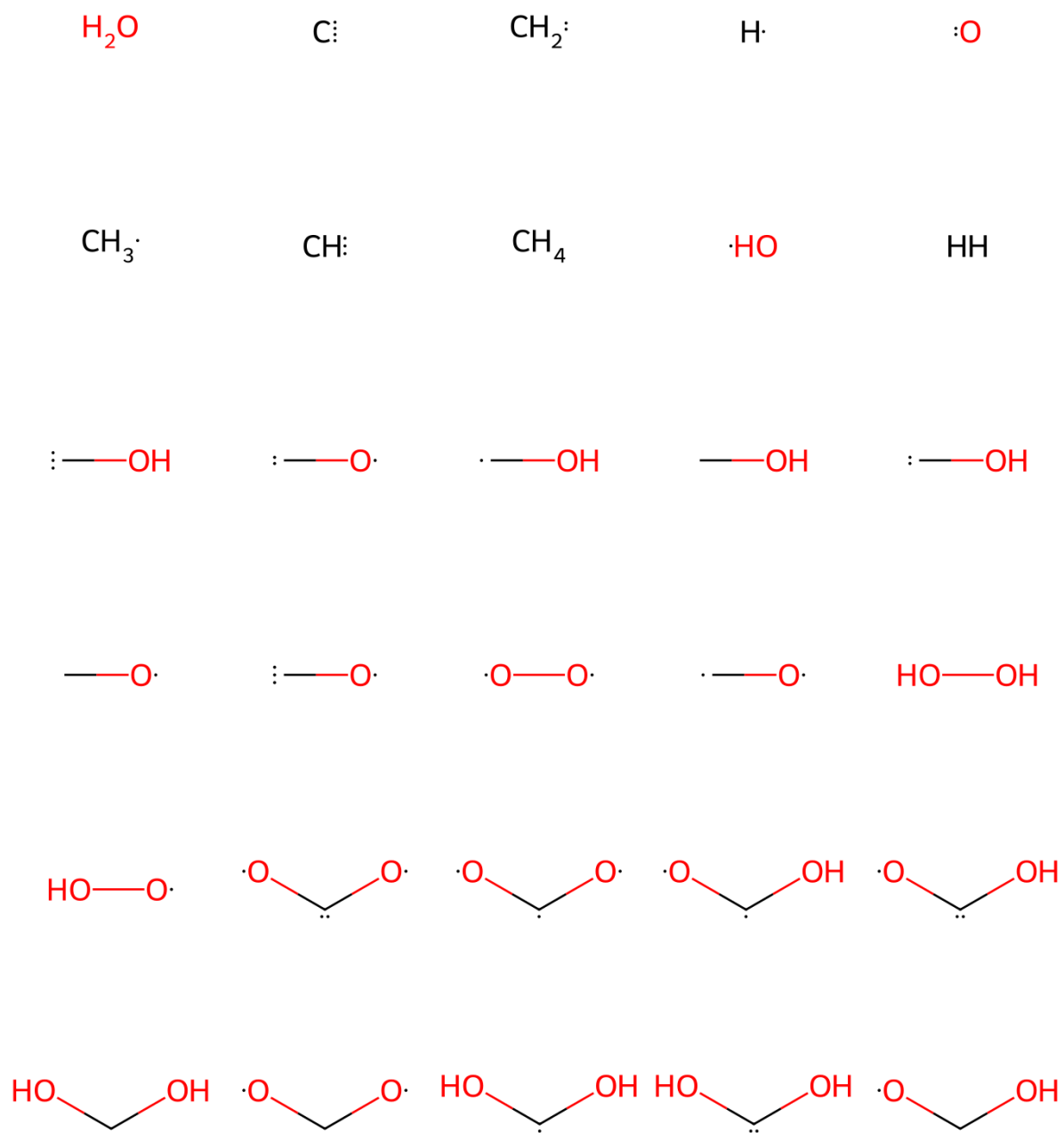


Fig. 15: Intermediates of the C_1O_2 CRN used for the methanol decomposition case study.

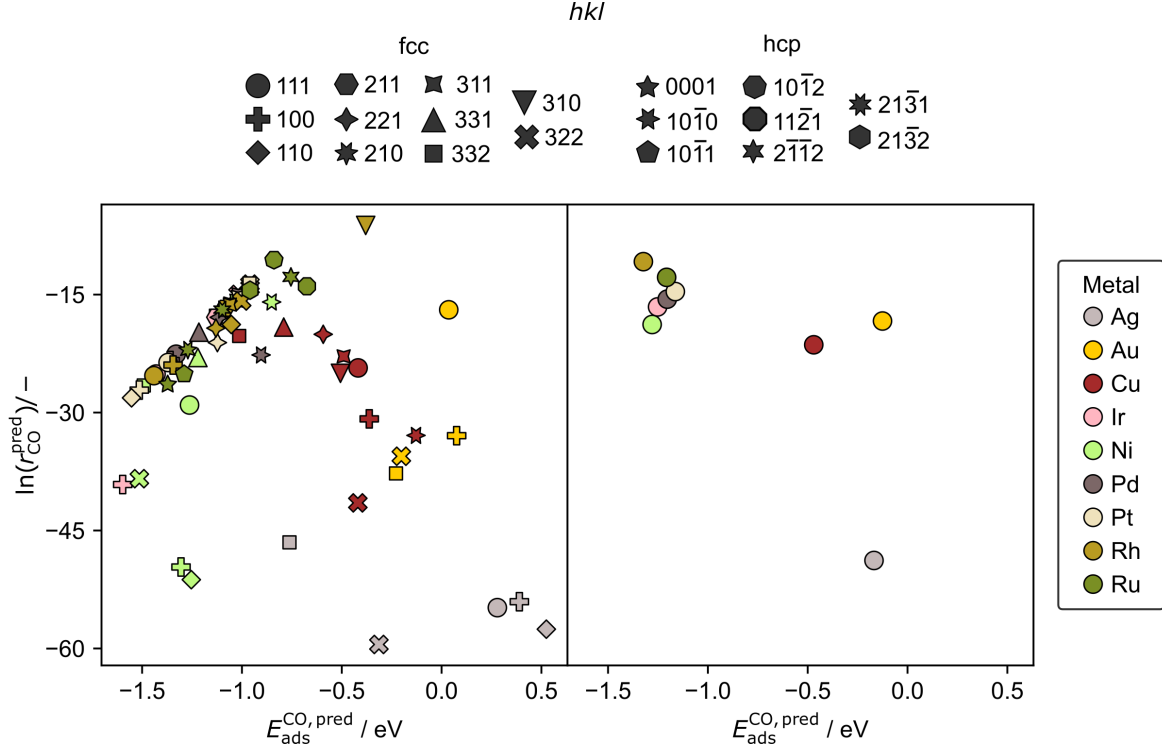


Fig. 16: CO formation rates from the microkinetic simulations for the methanol decomposition case study, as function of the predicted CO adsorption energy for **left** all available hkl surface orientations for each metal and **right** weighted by their fractional area contribution obtained from Crystalium database [20]. $T=473$ K, $P=1$ bar, $y_{\text{MeOH}}=10\%$. fcc=face-centered cubic; hcp=hexagonal close-packed.

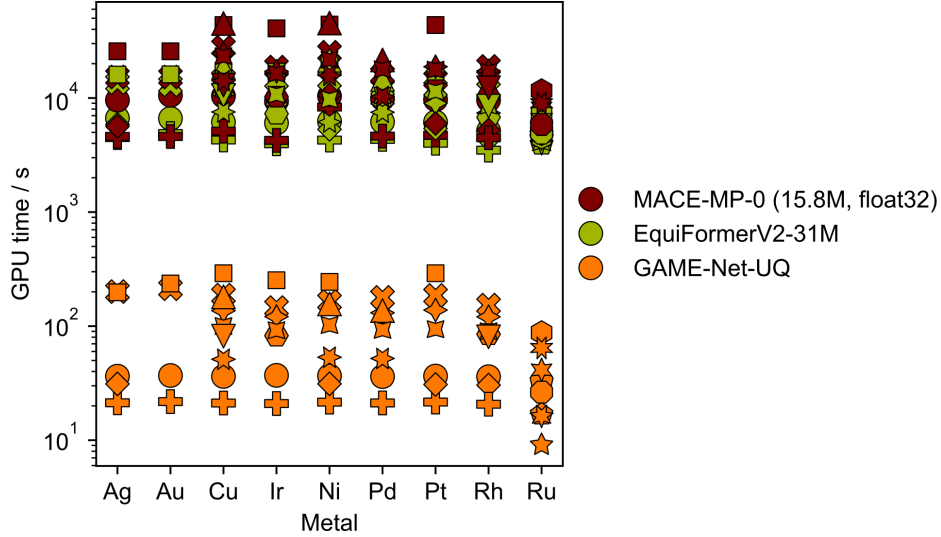


Fig. 17: Total GPU execution time for energy evaluation of intermediates and reactions (ΔE and E_{act}) in the MeOH decomposition CRNs, as function of metal, hkl surface, and ML model.

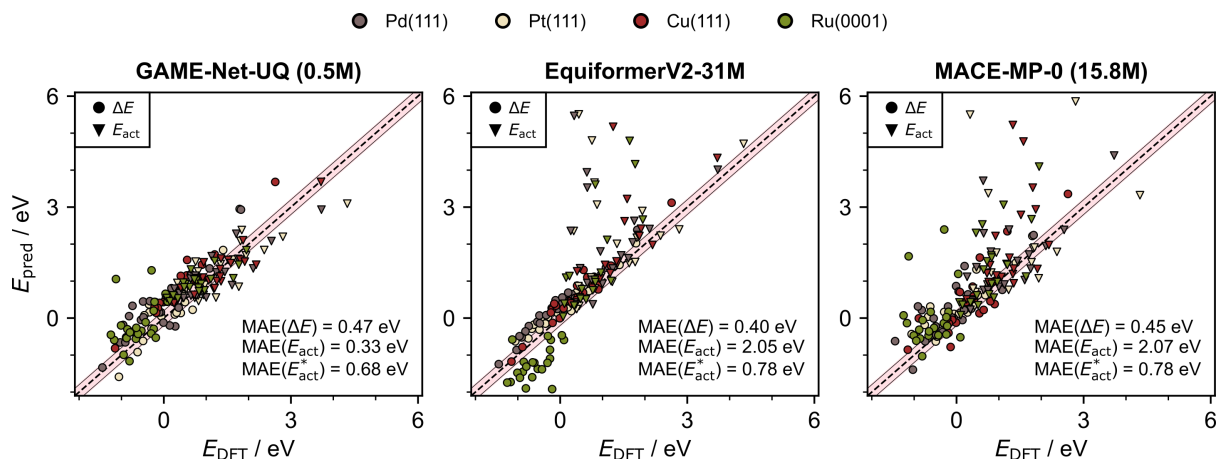


Fig. 18: Parity plots comparing reaction properties obtained with DFT and GAME-Net-UQ, EquiformerV2-31M [21], and MACE-MP-0 [22] for the methanol decomposition case study. Reaction energy ΔE data are shown as circles, while activation energy data E_{act} are displayed as triangles. E_{act}^* represents the activation energy obtained by considering all reactions as barrierless (Supplementary Figure 14). MAE=Mean Absolute Error. The highlighted bands represent the parity line ± 0.2 eV. Values can be found in Supplementary Table 10.

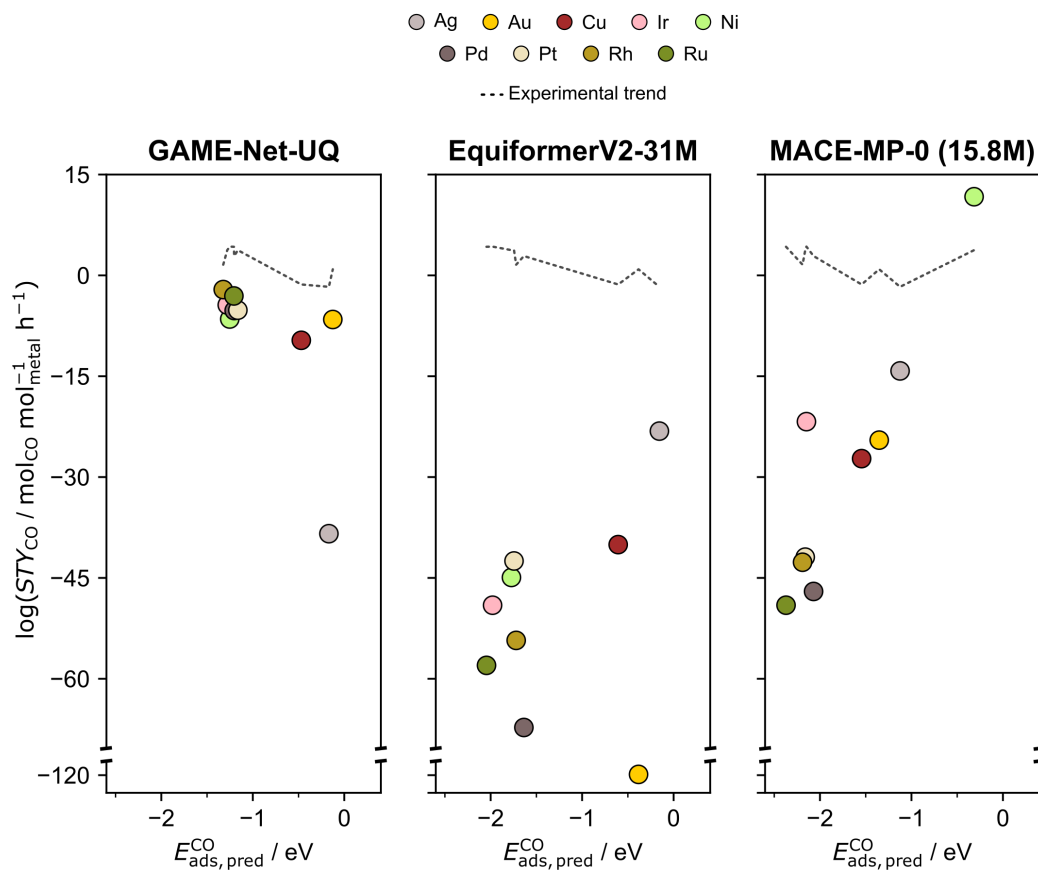


Fig. 19: Benchmark of the microkinetic modeling results obtained from the different MLIPs for the methanol decomposition case study. Logarithmic space-time yield (STY) for the CO formation from microkinetic simulations as function of the CO adsorption energy predicted by each evaluator. Benchmark settings and performance indicators (MAE and RMSE) are reported in **Supplementary Table 12**, while microkinetic results for all ML models can be found in **Supplementary Table 13**.

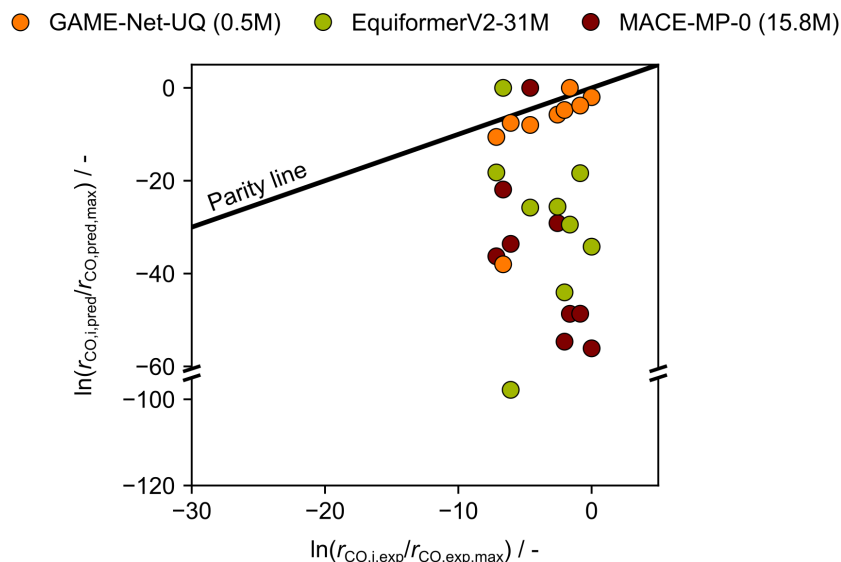


Fig. 20: Parity plot of the experimental *vs.* predicted logarithmic normalized CO production rates with values coming from microkinetic simulations using energy predictions from GAME-Net-UQ, EquiformerV2-31M [21], and MACE-MP-0 [22]. Black line represents the ideal parity line. Benchmark settings and performance metrics (MAE and RMSE) are reported in **Supplementary Table 12**, while microkinetic results for all ML models can be found in **Supplementary Table 13**.

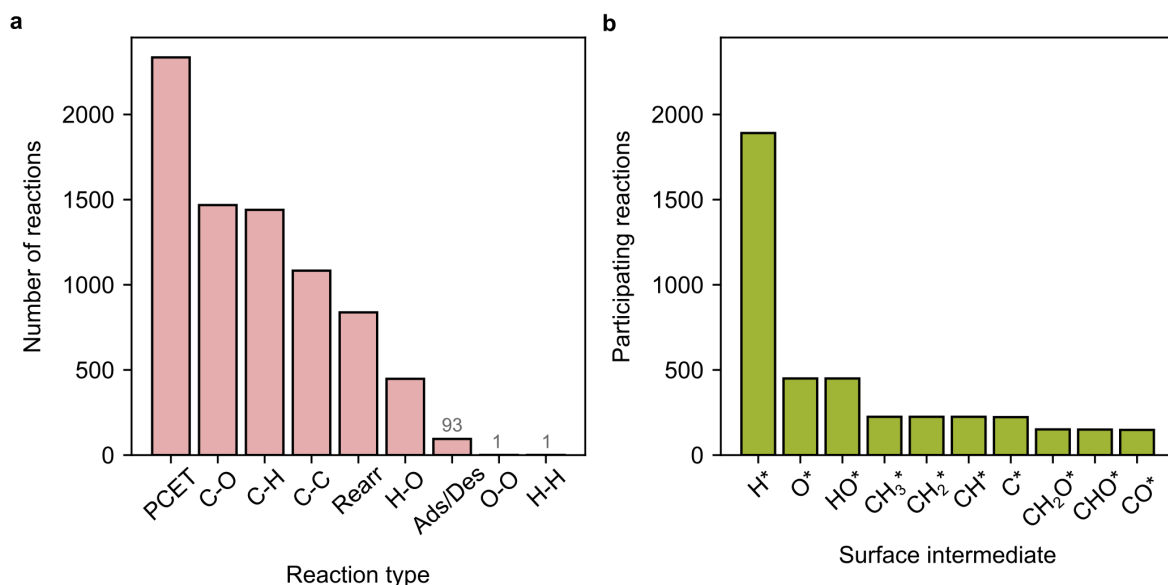


Fig. 21: Barplot of **a** number of elementary reactions by type in the C_3O_2 CRN on Cu(100) and **b** top-10 hub species. PCET=Proton-coupled electron transfer; Rearr=Rearrangement; Ads/Des=Adsorption/Desorption.

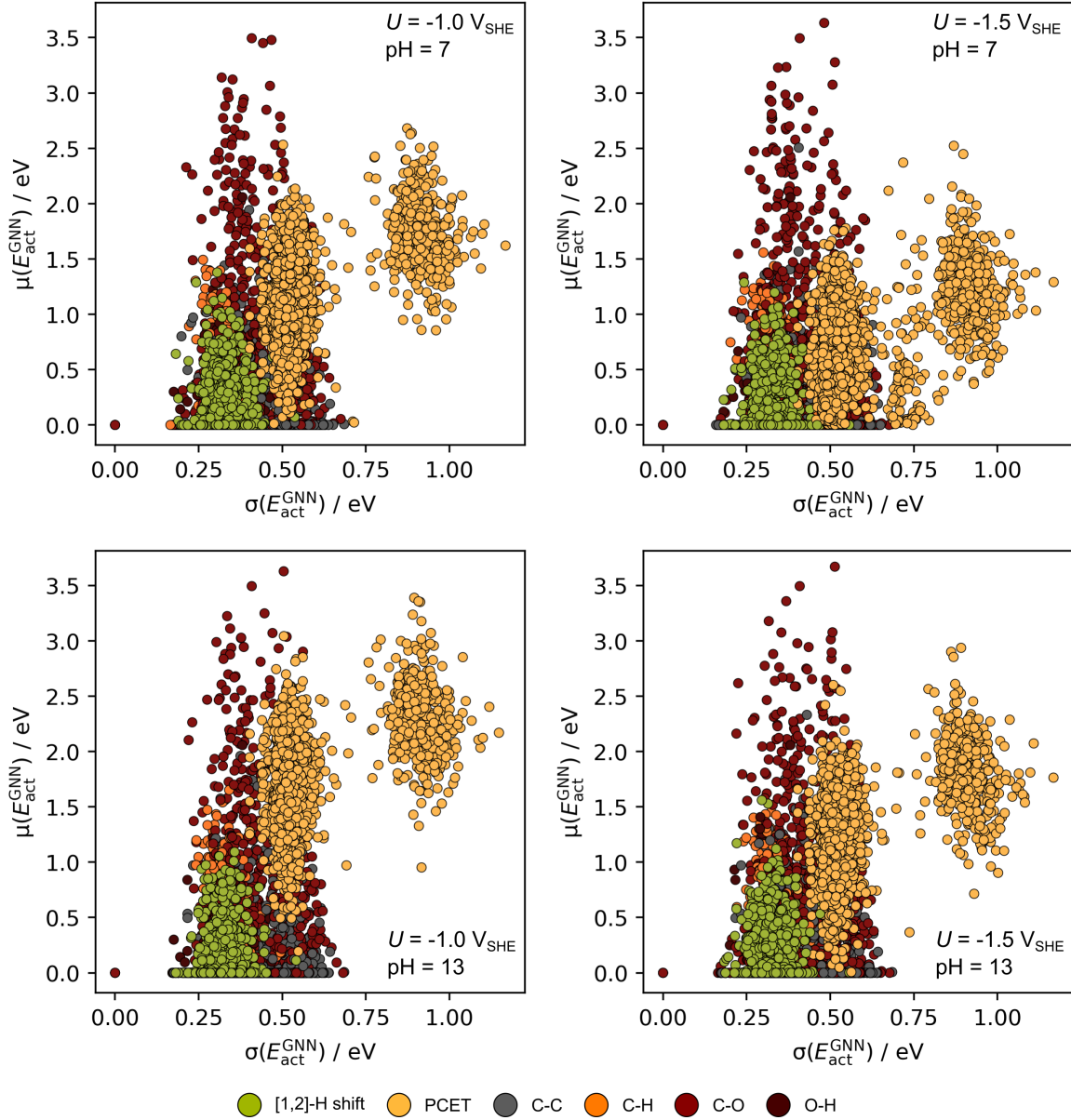


Fig. 22: Scatterplots of the activation energy E_{act} values (μ) and associated uncertainties (σ) obtained with GAME-Net-UQ of the elementary reactions in the electrochemical C_3O_2 network in the bond-forming direction as function of the applied potential and electrolyte pH, coloured by reaction type. Proton-coupled electron transfer (PCET) reactions cluster in two groups. The one with higher uncertainty represents elementary steps where the protonation of an -OH group forms water. $n = 7709$. SHE=Standard Hydrogen Electrode.

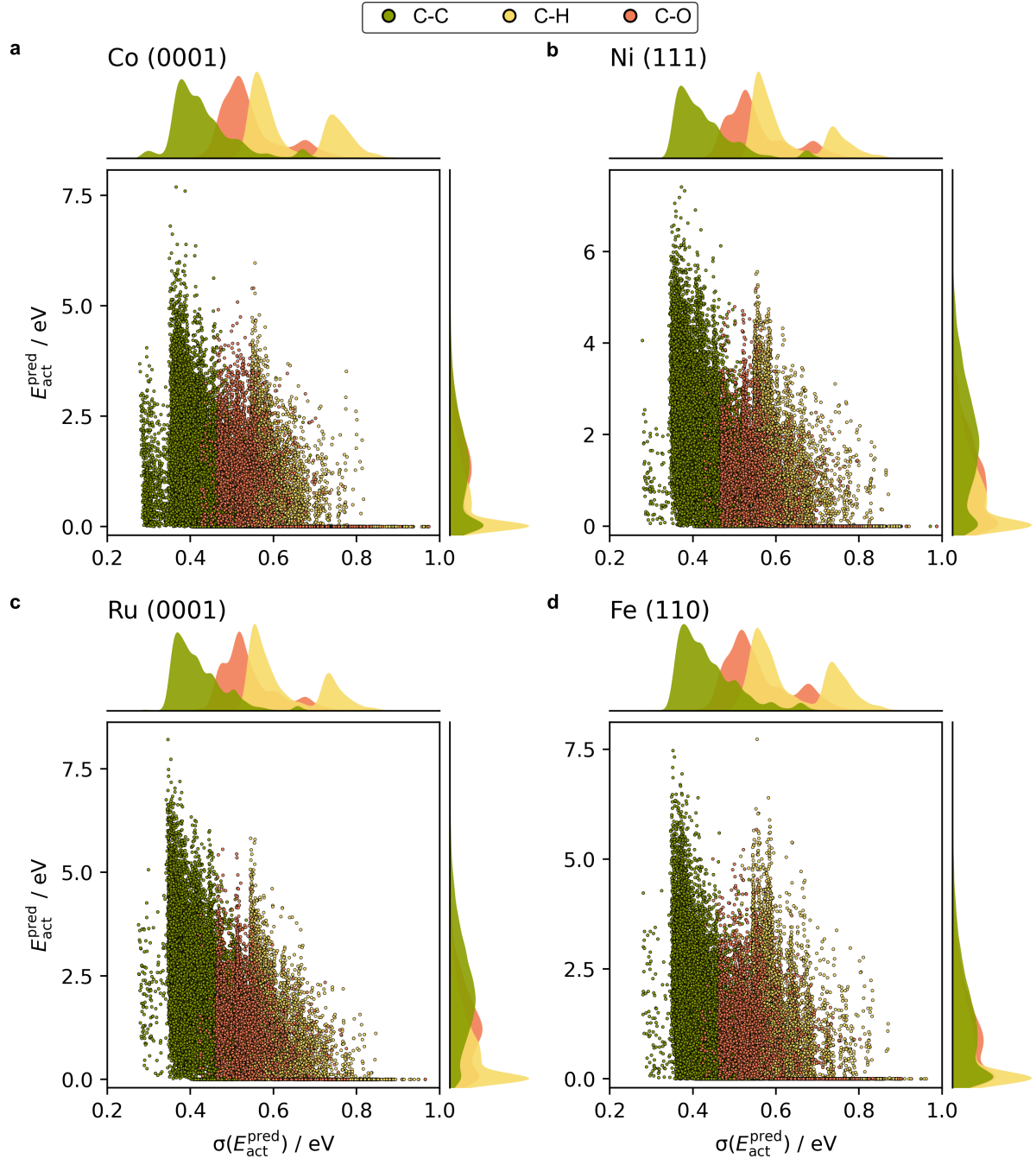


Fig. 23: Scatter-plot of the activation energies predicted by GAME-Net-UQ $E_{\text{act}}^{\text{pred}}$ and associated uncertainties σ grouped by reaction type, for the C-C, C-H and C-O dissociation reactions in the C_6O_1 reaction network representative of the Fischer-Tropsch process, on **a** Co(0001), **b** Ni(111), **c** Ru(0001), and **d** Fe(110). $n = 357, 224$.

Supplementary Tables

Table 1: Surfaces considered when developing GAME-Net-UQ. Slab height and vacuum reported in [$\text{\AA}$], area in [$\text{\AA}$]. fcc=face-centered cubic; hcp=hexagonal close-packed; bcc=body-centered cubic.

Metal	Structure	hkl	Layers	Area	Slab height	Atoms	Vacuum
Ag	fcc	111	4	89.35	7.11	48	12.89
		100	4	103.17	6.16	48	13.84
		110	6	145.91	7.22	72	12.78
Au	fcc	111	4	89.75	7.17	48	12.83
		100	4	103.64	6.18	48	13.82
		110	10	146.57	7.26	72	12.74
Cd	hcp	0001	4	95.47	8.59	48	11.41
		10 $\bar{1}$ 0	8	158.72	8.75	72	14.93
		10 $\bar{1}$ 1	8	173.68	8.78	72	15.35
Co	hcp	0001	4	64.50	6.05	48	13.95
		10 $\bar{1}$ 0	8	90.45	7.13	72	15.08
		10 $\bar{1}$ 1	8	102.65	7.12	72	15.18
Cu	fcc	111	4	68.60	6.21	48	13.79
		100	4	79.22	5.39	48	14.61
		110	8	112.03	6.32	72	13.68
Fe	bcc	110	4	67.90	6.05	48	13.95
		111	8	124.79	5.69	72	14.31
		100	6	72.04	7.19	54	14.89
Ir	fcc	111	4	77.91	6.61	48	13.39
		100	4	89.96	5.72	48	14.28
		110	7	127.22	6.75	72	13.25
Ni	fcc	111	4	64.32	6.04	48	13.96
		100	4	74.27	5.21	48	14.79
		110	7	105.03	6.10	72	13.90
Os	hcp	0001	4	78.02	6.47	48	13.53
		10 $\bar{1}$ 0	8	107.71	7.84	72	15.11
		10 $\bar{1}$ 1	8	122.90	7.80	72	15.21
Pd	fcc	111	4	80.61	6.80	48	13.20
		100	4	93.08	5.83	48	14.17
		110	7	131.63	6.90	72	13.10
Pt	fcc	111	4	81.82	6.85	48	13.15
		100	4	94.47	5.84	48	14.16
		110	8	133.60	6.91	72	13.09
Rh	fcc	111	4	75.97	6.52	48	13.48
		100	4	87.72	5.65	48	14.35
		110	8	124.06	6.72	72	13.28
Ru	hcp	0001	4	76.44	6.34	48	13.66
		10 $\bar{1}$ 0	8	104.26	7.71	72	15.11
		10 $\bar{1}$ 1	8	118.94	7.69	72	15.20
Zn	hcp	0001	4	76.61	7.14	48	12.86
		10 $\bar{1}$ 0	8	121.86	7.62	72	14.98
		10 $\bar{1}$ 1	8	133.31	7.51	72	15.48

Table 2: Composition of the graph dataset used to train and evaluate GAME-Net-UQ. Details on data splitting for training and evaluation can be found in **Supplementary Note 3**.

Category	<i>n</i>	%	Metals	Facets
Adsorbed species	9565	77.7	Ag, Au, Cu, Fe, Ir, Ni, Pd, Pt, Rh Cd, Co, Os, Ru, Zn	111, 110, 100 0001, 10 $\bar{1}$ 1, 10 $\bar{1}$ 0
Gas species	184	1.5	-	-
Transition states	2554	20.8	Ag, Au, Cu, Ir, Ni, Pd, Pt, Rh; Cd, Co, Os, Ru, Zn	111 0001
Total	12303	100	14	6

Table 3: Species in the GAME-Net-UQ training dataset (1/2).

InChIKey	Formula	InChIKey	Formula
AECGUCXUTHPJTJ-UHFFFAOYSA-N	CH3NO	JTNXQVCPQMQLHK-UHFFFAOYSA-N	C3H6S
SFKDSPYJDFPUMH-UHFFFAOYSA-N	C2H5NO	SUVIGLJNEAMWEG-UHFFFAOYSA-N	C3H8S
DGJKFPUBZQKYDG-UHFFFAOYSA-N	C2H5NO	KJRCEJOSASVSRA-UHFFFAOYSA-N	C3H8S
UITRKXIKGGFNRRZ-UHFFFAOYSA-N	C3H7NO	WXEHBUMAEPOYKP-UHFFFAOYSA-N	C3H8S
XQJNRLAOMOWOQA-UHFFFAOYSA-N	C3H7NO	QYYAEQQRMTZUPJ-UHFFFAOYSA-N	C4H8S
ZMXDDKWL CZADIW-UHFFFAOYSA-N	C3H7NO	OMNLPKGAQSUGE-UHFFFAOYSA-N	C4H8S
JFJVFPQRELNGCW-UHFFFAOYSA-N	C3H7NO	WQAQPCDUOCURKW-UHFFFAOYSA-N	C4H10S
CHOPEEJXLNOHPU-UHFFFAOYSA-N	C4H9NO	LOCHFZBWPCLPAN-SCSAIBSYSAN	C4H10S
ASBZENBKXGHKNE-UHFFFAOYSA-N	C4H9NO	LOCHFZBWPCLPAN-BYPYZUCNSAN	C4H10S
IANOTGWVCWAYKW-UHFFFAOYSA-N	C4H9NO	ZOASGOXWEHUTKZ-UHFFFAOYSA-N	C4H10S
FJLHLD BEZKTSOK-UHFFFAOYSA-N	C4H9NO	CQSQMIXIROIYTLO-UHFFFAOYSA-N	C4H8S
RPWDRUCHXZVYGM-UHFFFAOYSA-N	C4H9NO	WMXCDAVJEZZYLT-UHFFFAOYSA-N	C4H10S
WEAKFKMYBMVALU-UHFFFAOYSA-N	C4H9NO	LJSQFQKUNVCTIA-UHFFFAOYSA-N	C4H10S
FXHOIIRPVKKKFG-UHFFFAOYSA-N	C4H9NO	OWARERHZICHRON-UHFFFAOYSA-N	CH2O2
XWBHWICPYKYZBS-UHFFFAOYSA-N	C4H9NO	LBCQFPBSELSYJB-UHFFFAOYSA-N	CH2O3
OXDAYEMKZMQHGM-UHFFFAOYSA-N	CH4N2	MODBTQMWBRTGTDU-UHFFFAOYSA-N	C2H4O2
GPZREJXFUUUZH-UHFFFAOYSA-N	C2H6N2	TZIHFWKZFHZASV-UHFFFAOYSA-N	C2H4O2
CYKRHAOKXOSDJO-UHFFFAOYSA-N	C2H6N2	FOOFOXSSIXQYRV-UHFFFAOYSA-N	C2H4O3
JGBMTIKRTQWECX-UHFFFAOYSA-N	C2H6N2	LTEZBCNAMGAAES-UHFFFAOYSA-N	C3H6O2
GQSMXRFRDWRRSO-UHFFFAOYSA-N	C3H8N2	WBJINCZRORDGAQ-UHFFFAOYSA-N	C3H6O2
NHPHQOKECZBKU-UHFFFAOYSA-N	C3H8N2	KXKVLQRXCPEJIC-UHFFFAOYSA-N	C3H6O2
WVRKMAFAXWIPEL-UHFFFAOYSA-N	C3H8N2	JTQSZMCPUPZSKZ-UHFFFAOYSA-N	C3H6O3
XYEMSMVHYCFQCX-UHFFFAOYSA-N	C3H8N2	IEJIGPNLZYLLBP-UHFFFAOYSA-N	C3H6O3
DSWNRHCOGVRDOE-UHFFFAOYSA-N	C3H8N2	NRMXCYOYDXLOK-UHFFFAOYSA-N	C4H8O2
IWPDYHTXXUWAOG-UHFFFAOYSA-N	C3H8N2	FEAPOKONUDZADA-UHFFFAOYSA-N	C4H8O2
OKOGENLSQNAZNG-UHFFFAOYSA-N	C3H8N2	KFNHILCVOLYIR-UHFFFAOYSA-N	C4H8O2
BBDVJSZQELZGRA-UHFFFAOYSA-N	C4H10N2	RMOUBSOVHSONPZ-UHFFFAOYSA-N	C4H8O2
DGJGQEXKDZKKJC-UHFFFAOYSA-N	C4H10N2	XEKOWRVHYACXOJ-UHFFFAOYSA-N	C4H8O2
CEZZGEGVYIGHKV-UHFFFAOYSA-N	C4H10N2	RJUFJBKOKNCXHH-UHFFFAOYSA-N	C4H8O2
YBGDWIUBUSZYDE-UHFFFAOYSA-N	C4H10N2	JPGVXXHSEFTIDC-UHFFFAOYSA-N	C4H8O3
IIMUSGFHNCKDFC-UHFFFAOYSA-N	C4H10N2	WHQZWHMMXFTTLH-UHFFFAOYSA-N	C4H8O3
LXPOLDQCPQHY-UHFFFAOYSA-N	C4H10N2	JBTWLSYZIRCDFO-UHFFFAOYSA-N	C4H8O3
PBCGKWSGSBCQIO-UHFFFAOYSA-N	C4H10N2	SQDFHQTJAWCFIB-UHFFFAOYSA-N	CH3NO
QXHV FOLQIFMHND-UHFFFAOYSA-N	C4H10N2	FZENGLVLUJGJX-UHFFFAOYSA-N	C2H5NO
PAXRVGTBDATMF-UHFFFAOYSA-N	C4H10N2	IFDZZSXEPSSHNC-UHFFFAOYSA-N	C3H7NO
IGQMRXQIBZWPQ-UHFFFAOYSA-N	C4H10N2	PXAJQJMDXJWFB-UHFFFAOYSA-N	C3H7NO
KXRGLKFLUYWCPK-UHFFFAOYSA-N	C4H10N2	KGGVGTQEGGOZRN-UHFFFAOYSA-N	C4H9NO
DMNYJSMNRDYEOT-UHFFFAOYSA-N	C4H10N2	SYJPAKDNFZLSMV-UHFFFAOYSA-N	C4H9NO
BJYBAILYGNMNGZ-UHFFFAOYSA-N	C4H10N2	WHIVNJATOVLBWB-UHFFFAOYSA-N	C4H9NO
LYMPVLRYGOKDLY-UHFFFAOYSA-N	C4H10N2	XLYOFNOQVPJJNP-UHFFFAOYSA-N	H2O
RPJOONIASVZOKB-UHFFFAOYSA-N	C4H10N2	UGFAIRIUMAVXCW-UHFFFAOYSA-N	CO
LEJDDBPWNWNHCT-UHFFFAOYSA-N	C4H10N2	VNWKTOKETHTGBQD-UHFFFAOYSA-N	CH4
IAYPJSHDLNXXLG-UHFFFAOYSA-N	C4H4S	OKKJLVBELUTLKV-UHFFFAOYSA-N	CH4O
HGVFOSNZNHQXIW-UHFFFAOYSA-N	CH3NO2	CURLTUGMZLYLDI-UHFFFAOYSA-N	CO2
VBTYNRHZDSWJCL-UHFFFAOYSA-N	C2H5NO2	SMIUKHFIQXZIP-UHFFFAOYSA-N	H2
NXUOLOBRZBCFJE-UHFFFAOYSA-N	C2H5NO2	RWSOTUBLDIXVET-UHFFFAOYSA-N	H2S
HRBJQYCIEOOCPU-UHFFFAOYSA-N	C3H7NO2	QGZKDVFNQNGYKY-UHFFFAOYSA-N	H3N
WMXFSBZDJRGXFF-UHFFFAOYSA-N	C3H7NO2	MYMOFIZGZYHOMD-UHFFFAOYSA-N	O2
JTRNLKDJASRMNC-UHFFFAOYSA-N	C3H7NO2	RAHZWNYVWXNFOC-UHFFFAOYSA-N	O2S
LCDHCQSTRWYJQT-UHFFFAOYSA-N	C3H7NO2	SNRUBQQJIBEYMU-UHFFFAOYSA-N	C12H26
GLHFUWDZRHCLYD-UHFFFAOYSA-N	C4H9NO2	WIRCLRWWKJJKHO-ZNMIVQPWSAN	C15H32
JHNFHGOYCHADHO-UHFFFAOYSA-N	C4H9NO2	WIRCLRWWKJJKHO-ILXRZTDVSA-N	C15H32
HRKAQXFFKCYSHV-UHFFFAOYSA-N	C4H9NO2	OKTJSMMVPCPJKN-UHFFFAOYSA-N	C
ZCEKZAREXZJYHV-UHFFFAOYSA-N	C4H9NO2	HZVOZRGWRWCICA-UHFFFAOYSA-N	CH2
UVTCDTIWDJBSBP-UHFFFAOYSA-N	C4H9NO2	WCYWMZMISLQXQU-UHFFFAOYSA-N	CH3
UWUUBGJUIUNVEJ-UHFFFAOYSA-N	C4H9NO2	VRILPUYDFBWXCH-UHFFFAOYSA-N	CH
BXYNEAWHAAGRQE-UHFFFAOYSA-N	C4H9NO2	YZCKVEUIGOORGS-UHFFFAOYSA-N	H
SELXJABLPLKXOY-UHFFFAOYSA-N	C4H9NO2	QJGQUHNMIGDVP-M-UHFFFAOYSA-N	N
WSFSSNUMVMOOMR-UHFFFAOYSA-N	CH2O	MDFNEOEWEAXZRQ-UHFFFAOYSA-N	H2N
HSFWRNGVRCDJHI-UHFFFAOYSA-N	C2H2	PDCKRJYJMCOFU-UHFFFAOYSA-N	HN
VGGSQFUCUMXWEO-UHFFFAOYSA-N	C2H4	QVGXLLKOCUKJST-UHFFFAOYSA-N	O
IKHGUXGNTLKF-UHFFFAOYSA-N	C2H4O	TUJKJAMUKRIRHC-UHFFFAOYSA-N	HO

Table 4: Species in the GAME-Net-UQ training dataset (2/2).

InChIKey	Formula	InChIKey	Formula
OTMSDBZUPAUEDD-UHFFFAOYSA-N	C2H6	NINIDFKCEFEMDL-UHFFFAOYSA-N	S
LCGLNKUTAGEVQW-UHFFFAOYSA-N	C2H6O	PXQLVRUNWNTZOS-UHFFFAOYSA-N	HS
LFQSCWFLJHTTHZ-UHFFFAOYSA-N	C2H6O	CFHIDWOYWUOIHU-UHFFFAOYSA-N	CHO
MWWATHDPGQKSAR-UHFFFAOYSA-N	C3H4	GRVDJDISBSALJP-UHFFFAOYSA-N	CH3O
QQONPFPTGQHPMA-UHFFFAOYSA-N	C3H6	CBOIHMHRHGLHBPB-UHFFFAOYSA-N	CH3O
CSCPACGZOOCCGX-UHFFFAOYSA-N	C3H6O	UUOIGPDTLYBNGY-UHFFFAOYSA-N	CH2O
ATUOYWHBWRKTHZ-UHFFFAOYSA-N	C3H8	ADJRDRYKXYFKM-UHFFFAOYSA-N	CHO
XOBKSJDNFUFZPF-UHFFFAOYSA-N	C3H8O	RGKSXKVIDIQWGT-UHFFFAOYSA-N	C2H3O
BDERNNFJNOPAEC-UHFFFAOYSA-N	C3H8O	VOLGAXAGEUPBDM-UHFFFAOYSA-N	C2H5O
KFZMGEQAYNKOFG-UHFFFAOYSA-N	C3H8O	KBQZXXMNJQRFEK-UHFFFAOYSA-N	C2H3O
XNMQEEKYCVKGBD-UHFFFAOYSA-N	C4H6	XMHKKYSQISNWMY-UHFFFAOYSA-N	C2HO
KDKYADYSIPSCCQ-UHFFFAOYSA-N	C4H6	KQXYVMHIWIXTEY-UHFFFAOYSA-N	C2H2O
VXNZUUAINFGBPY-UHFFFAOYSA-N	C4H8	FATAVLOOLIRUNA-UHFFFAOYSA-N	C2H3O
IAQRGUVFOMOMEM-UHFFFAOYSA-N	C4H8	YOHSDXODKVUPAKH-UHFFFAOYSA-N	C2HO
VQTUBCKSQIDNK-UHFFFAOYSA-N	C4H8	CCGKOQOJPYTBH-UHFFFAOYSA-N	C2H2O
ZWEHNKRNPVVGH-UHFFFAOYSA-N	C4H8O	IMROMDMJAWUWLK-UHFFFAOYSA-N	C2H4O
IJDNQMDRQITEOD-UHFFFAOYSA-N	C4H10	QFXZANXYUCUTQH-UHFFFAOYSA-N	C2H2O
NNPMTNADJDCUHE-UHFFFAOYSA-N	C4H10	AXSRCFOSKHEJGQ-UHFFFAOYSA-N	C2H2O
LRHPLDYGVMQRHN-UHFFFAOYSA-N	C4H10O	QNCISOQOWBORKW-UHFFFAOYSA-N	C2H3O
BTANRVKQWNVYAZ-SCSAIBSYSAN	C4H10O	TUCNEACPLKLKNU-UHFFFAOYSA-N	C2H3O
DKGAVHZHDRPRBM-UHFFFAOYSA-N	C4H10O	DCQVFLDXBPQJHX-UHFFFAOYSA-N	C2H4O
BTANRVKQWNVYAZ-BYPYZUCNSAN	C4H10O	JVKQHDUTAFISFX-UHFFFAOYSA-N	C2H4O
NBBJYMSMWHIQUU-UHFFFAOYSA-N	C3H6O	ODVIVHCWPKHAQ-UHFFFAOYSA-N	C2H4O
ZTQSAQDEMFDMZ-UHFFFAOYSA-N	C4H8O	VILAVOFMLJHSJA-UHFFFAOYSA-N	C2O
AMIMRNSIRUDHCM-UHFFFAOYSA-N	C4H8O	QEQJAPYSVNHJF-UHFFFAOYSA-N	C2HO
RTZKZFJDLAIYFH-UHFFFAOYSA-N	C4H10O	UBXLAXBHBLQHZ-UHFFFAOYSA-N	C2H2O
VNKYQGIUYNRMU-UHFFFAOYSA-N	C4H10O	HVFGPIFTSJAKK-UHFFFAOYSA-N	C2H5O
WDWDWGRYHDPDS-UHFFFAOYSA-N	CH3N	GAWIXWVDTYZZAW-UHFFFAOYSA-N	C2H5O
BAVYZALUXZFZLV-UHFFFAOYSA-N	CH5N	UHAMPRMMGCGNGCH-UHFFFAOYSA-N	C2H3O
MPAYEWNVIPXRDP-UHFFFAOYSA-N	C2H5N	SIFIIWPGTLBBLK-UHFFFAOYSA-N	C2H5O2
NVSPDPNXBIEFPW-UHFFFAOYSA-N	C2H5N	NIDSBKZSDFQVDP-UHFFFAOYSA-N	C2H2O2
QUSNBJAOMFDB-UHFFFAOYSA-N	C2H7N	GXBYUAHVCCRBRG-UHFFFAOYSA-N	C2H3O2
ROSDSFDQJNGOL-UHFFFAOYSA-N	C2H7N	PJJVOLCMZFXCJL-UHFFFAOYSA-N	C2H4O2
WJKYOQDIQYJXSD-UHFFFAOYSA-N	C3H7N	IGWGBFUBFQGEQP-UHFFFAOYSA-N	C2H3O2
FKOYAFITBICFAO-UHFFFAOYSA-N	C3H7N	QKCFVDWBORTSIN-UHFFFAOYSA-N	C2H5O2
OHACMJAMDKRMWS-UHFFFAOYSA-N	C3H7N	YIDVHFDVPYGGSK-UHFFFAOYSA-N	C2H3O2
XDAGXZXKTKRFMT-UHFFFAOYSA-N	C3H7N	IKFPMWBBLSRQRA-UHFFFAOYSA-N	C2HO2
WGYKZJWCGVVSQN-UHFFFAOYSA-N	C3H9N	LEQAOMBKQFMDFZ-UHFFFAOYSA-N	C2H2O2
JJWLVOIRVHMVIS-UHFFFAOYSA-N	C3H9N	LYCAIKOWRPUZTN-UHFFFAOYSA-N	C2H6O2
LIWAQLJGPBVORC-UHFFFAOYSA-N	C3H9N	ZUQAPLKKNQJAU-UHFFFAOYSA-N	C2H2O2
GETQZCLCWQTVFV-UHFFFAOYSA-N	C3H9N	MURXBMUMXOJVCU-UHFFFAOYSA-N	C2H4O2
UOFFEDRAIFYOBS-UHFFFAOYSA-N	C4H9N	JMCREDBJJPRTPV-UHFFFAOYSA-N	C2H4O2
MWIRQVRILGQXFA-UHFFFAOYSA-N	C4H9N	ZDMBQXQILMMBHM-UHFFFAOYSA-N	C2HO2
IGPVHVVXJXDFSII-UHFFFAOYSA-N	C4H9N	WGCNASOHLSPBMP-UHFFFAOYSA-N	C2H4O2
MGKTYORWGSVLRD-UHFFFAOYSA-N	C4H9N	JSBBMMHTNIUVTC-UHFFFAOYSA-N	C2H3O2
ARYPXRGDKSAXBV-UHFFFAOYSA-N	C4H9N	OOSBGBJKELAIIG-UHFFFAOYSA-N	C2H3O2
JKHYHXLSTVHIA-UHFFFAOYSA-N	C4H9N	WVKOKNQPFEMSR-UHFFFAOYSA-N	C2H2O2
BMMKPGVCTLTADE-UHFFFAOYSA-N	C4H9N	PDWPSYGMFMFGKP-UHFFFAOYSA-N	C2H2O2
GPASKEFEXOCRNP-UHFFFAOYSA-N	C4H9N	NTLDRJWXTJMITR-UHFFFAOYSA-N	C2H4O2
HQABUPZFAYXKJW-UHFFFAOYSA-N	C4H11N	JRCLCXASVCEQC-UHFFFAOYSA-N	CHO2
BHRZNVHARXXAHW-SCSAIBSYSAN	C4H11N	XEHVFKKSDRMODY-UHFFFAOYSA-N	C2H
YBRBMKDOPFTVDT-UHFFFAOYSA-N	C4H11N	SNVLJLYUUXKWOJ-UHFFFAOYSA-N	C2H2
HPNMFZURITQLUMO-UHFFFAOYSA-N	C4H11N	QUPDWYMUPZLYJZ-UHFFFAOYSA-N	C2H5
GVWISOJSERXQBM-UHFFFAOYSA-N	C4H11N	UUFQTNFCRMXOAE-UHFFFAOYSA-N	C2H4
DAZXVJBIRMWXPJ-UHFFFAOYSA-N	C4H11N	ADLIKHOGUAKPM-UHFFFAOYSA-N	C2H3
BHRZNVHARXXAHW-BYPYZUCNSAN	C4H11N	LBVWYGGGGJURHQ-UHFFFAOYSA-N	C2
DBTDEFJAFBUGPP-UHFFFAOYSA-N	CH2S	ORGHESHFPYLAO-UHFFFAOYSA-N	C2H3
LSDPWHWYPCBBB-UHFFFAOYSA-N	CH4S	MPKDVKDMYXRMIU-UHFFFAOYSA-N	CHO2
QJFUMFCCMJLIE-UHFFFAOYSA-N	C2H4S	FONOSWYYBCBQGN-UHFFFAOYSA-N	C2O2
DNJIEGIFACGWOD-UHFFFAOYSA-N	C2H6S	NKXMGCUADHKCDW-UHFFFAOYSA-N	C3H4
QMMFVYPAHWMCMS-UHFFFAOYSA-N	C2H6S	NOZVYUMSEIFOSW-UHFFFAOYSA-N	C3H3
UXBLKPIXRLLBH-UHFFFAOYSA-N	C3H6S	QNVRIHYSUZMSGM-LURJTMIESAN	C6H14O

Table 5: Elementary reactions for which the transition state is included in the dataset for GAME-Net-UQ. The same reaction string can represent more than one reaction.

$\text{CO} \longrightarrow \text{C} + \text{O}$	$\text{HO} \longrightarrow \text{O} + \text{H}$
$\text{C}_2 \longrightarrow \text{C} + \text{C}$	$\text{CH} \longrightarrow \text{C} + \text{H}$
$\text{O}_2 \longrightarrow \text{O} + \text{O}$	$\text{C}_2\text{H} \longrightarrow \text{CH} + \text{C}$
$\text{H}_2\text{O} \longrightarrow \text{HO} + \text{H}$	$\text{C}_2\text{O} \longrightarrow \text{C}_2 + \text{O}$
$\text{CHO} \longrightarrow \text{C} + \text{HO}$	$\text{C}_2\text{H} \longrightarrow \text{C}_2 + \text{H}$
$\text{CO}_2 \longrightarrow \text{CO} + \text{O}$	$\text{C}_2\text{O} \longrightarrow \text{CO} + \text{C}$
$\text{CHO} \longrightarrow \text{CH} + \text{O}$	$\text{CHO} \longrightarrow \text{CO} + \text{H}$
$\text{CH}_2 \longrightarrow \text{CH} + \text{H}$	$\text{CH}_4 \longrightarrow \text{CH}_3 + \text{H}$
$\text{CH}_3 \longrightarrow \text{CH}_2 + \text{H}$	$\text{CHO}_2 \longrightarrow \text{CO}_2 + \text{H}$
$\text{C}_2\text{O}_2 \longrightarrow \text{C}_2\text{O} + \text{O}$	$\text{C}_2\text{HO} \longrightarrow \text{CO} + \text{CH}$
$\text{C}_2\text{HO} \longrightarrow \text{C}_2\text{O} + \text{H}$	$\text{CH}_2\text{O} \longrightarrow \text{CH} + \text{HO}$
$\text{CH}_3\text{O} \longrightarrow \text{CH}_3 + \text{O}$	$\text{CHO}_2 \longrightarrow \text{CO} + \text{HO}$
$\text{C}_2\text{O}_2 \longrightarrow \text{CO} + \text{CO}$	$\text{C}_2\text{H}_3 \longrightarrow \text{CH}_3 + \text{C}$
$\text{CH}_2\text{O} \longrightarrow \text{CH}_2 + \text{O}$	$\text{C}_2\text{HO} \longrightarrow \text{C}_2\text{H} + \text{O}$
$\text{CH}_2\text{O} \longrightarrow \text{CHO} + \text{H}$	$\text{C}_2\text{H}_2 \longrightarrow \text{CH}_2 + \text{C}$
$\text{C}_2\text{HO} \longrightarrow \text{CHO} + \text{C}$	$\text{C}_2\text{HO} \longrightarrow \text{C}_2 + \text{HO}$
$\text{C}_2\text{H}_2 \longrightarrow \text{CH} + \text{CH}$	$\text{C}_2\text{H}_2 \longrightarrow \text{C}_2\text{H} + \text{H}$
$\text{C}_2\text{H}_4 \longrightarrow \text{C}_2\text{H}_3 + \text{H}$	$\text{C}_2\text{H}_3 \longrightarrow \text{C}_2\text{H}_2 + \text{H}$
$\text{C}_2\text{H}_5 \longrightarrow \text{C}_2\text{H}_4 + \text{H}$	$\text{C}_2\text{H}_3 \longrightarrow \text{CH}_2 + \text{CH}$
$\text{C}_2\text{H}_6 \longrightarrow \text{C}_2\text{H}_5 + \text{H}$	$\text{CH}_3\text{O} \longrightarrow \text{CH}_2 + \text{HO}$
$\text{C}_2\text{H}_4 \longrightarrow \text{CH}_3 + \text{CH}$	$\text{CH}_4\text{O} \longrightarrow \text{CH}_3 + \text{HO}$
$\text{CH}_4\text{O} \longrightarrow \text{CH}_3\text{O} + \text{H}$	$\text{CH}_3\text{O} \longrightarrow \text{CH}_2\text{O} + \text{H}$
$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{CHO} + \text{CH}$	$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{C}_2\text{H}_2 + \text{O}$
$\text{C}_2\text{H}_4 \longrightarrow \text{CH}_2 + \text{CH}_2$	$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{C}_2\text{H}_3 + \text{O}$
$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{CH}_3\text{O} + \text{C}$	$\text{C}_2\text{HO}_2 \longrightarrow \text{C}_2\text{O}_2 + \text{H}$
$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{C}_2\text{HO} + \text{H}$	$\text{C}_2\text{H}_6 \longrightarrow \text{CH}_3 + \text{CH}_3$
$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{CO} + \text{CH}_3$	$\text{C}_2\text{H}_5 \longrightarrow \text{CH}_3 + \text{CH}_2$
$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{C}_2\text{H} + \text{HO}$	$\text{C}_2\text{HO}_2 \longrightarrow \text{CHO} + \text{CO}$
$\text{C}_2\text{HO}_2 \longrightarrow \text{C}_2\text{HO} + \text{O}$	$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{CH}_2\text{O} + \text{C}$
$\text{C}_2\text{H}_5\text{O} \longrightarrow \text{C}_2\text{H}_5 + \text{O}$	$\text{C}_2\text{HO}_2 \longrightarrow \text{C}_2\text{O} + \text{HO}$
$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{C}_2\text{H}_4 + \text{O}$	$\text{C}_2\text{H}_2\text{O} \longrightarrow \text{CO} + \text{CH}_2$
$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{C}_2\text{H}_2 + \text{HO}$	$\text{C}_2\text{H}_6\text{O} \longrightarrow \text{C}_2\text{H}_5\text{O} + \text{H}$
$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{CH}_3\text{O} + \text{CH}$	$\text{C}_2\text{H}_6\text{O} \longrightarrow \text{C}_2\text{H}_5 + \text{HO}$
$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{C}_2\text{H}_3\text{O} + \text{H}$	$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{C}_2\text{H}_3 + \text{HO}$
$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{CHO} + \text{CH}_2$	$\text{C}_2\text{H}_5\text{O} \longrightarrow \text{C}_2\text{H}_4\text{O} + \text{H}$
$\text{C}_2\text{H}_5\text{O} \longrightarrow \text{C}_2\text{H}_4 + \text{HO}$	$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{CH}_2\text{O} + \text{CH}$
$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{CHO} + \text{CH}_3$	$\text{C}_2\text{H}_3\text{O} \longrightarrow \text{C}_2\text{H}_2\text{O} + \text{H}$
$\text{C}_2\text{H}_4\text{O}_2 \longrightarrow \text{C}_2\text{H}_4\text{O} + \text{O}$	$\text{C}_2\text{H}_5\text{O} \longrightarrow \text{CH}_3\text{O} + \text{CH}_2$
$\text{C}_2\text{H}_5\text{O} \longrightarrow \text{CH}_2\text{O} + \text{CH}_3$	$\text{C}_2\text{H}_3\text{O}_2 \longrightarrow \text{CH}_3\text{O} + \text{CO}$
$\text{C}_2\text{H}_5\text{O}_2 \longrightarrow \text{C}_2\text{H}_5\text{O} + \text{O}$	$\text{C}_2\text{H}_2\text{O}_2 \longrightarrow \text{C}_2\text{H}_2\text{O} + \text{O}$
$\text{C}_2\text{H}_2\text{O}_2 \longrightarrow \text{CH}_2\text{O} + \text{CO}$	$\text{C}_2\text{H}_3\text{O}_2 \longrightarrow \text{C}_2\text{H}_3\text{O} + \text{O}$
$\text{C}_2\text{H}_2\text{O}_2 \longrightarrow \text{C}_2\text{HO} + \text{HO}$	$\text{C}_2\text{H}_2\text{O}_2 \longrightarrow \text{C}_2\text{HO}_2 + \text{H}$
$\text{C}_2\text{H}_4\text{O} \longrightarrow \text{CH}_2\text{O} + \text{CH}_2$	$\text{C}_2\text{H}_2\text{O}_2 \longrightarrow \text{CHO} + \text{CHO}$
$\text{C}_2\text{H}_3\text{O}_2 \longrightarrow \text{CH}_2\text{O} + \text{CHO}$	$\text{C}_2\text{H}_5\text{O}_2 \longrightarrow \text{C}_2\text{H}_4\text{O} + \text{HO}$
$\text{C}_2\text{H}_5\text{O}_2 \longrightarrow \text{C}_2\text{H}_4\text{O}_2 + \text{H}$	$\text{C}_2\text{H}_4\text{O}_2 \longrightarrow \text{C}_2\text{H}_3\text{O}_2 + \text{H}$
$\text{C}_2\text{H}_6\text{O}_2 \longrightarrow \text{C}_2\text{H}_5\text{O} + \text{HO}$	$\text{C}_2\text{H}_3\text{O}_2 \longrightarrow \text{C}_2\text{H}_2\text{O} + \text{HO}$
$\text{C}_2\text{H}_6\text{O}_2 \longrightarrow \text{C}_2\text{H}_5\text{O}_2 + \text{H}$	$\text{C}_2\text{H}_4\text{O}_2 \longrightarrow \text{CH}_3\text{O} + \text{CHO}$
$\text{C}_2\text{H}_3\text{O}_2 \longrightarrow \text{C}_2\text{H}_2\text{O}_2 + \text{H}$	$\text{C}_2\text{H}_4\text{O}_2 \longrightarrow \text{C}_2\text{H}_3\text{O} + \text{HO}$
$\text{C}_2\text{H}_5\text{O}_2 \longrightarrow \text{CH}_3\text{O} + \text{CH}_2\text{O}$	$\text{C}_2\text{H}_4\text{O}_2 \longrightarrow \text{CH}_2\text{O} + \text{CH}_2\text{O}$

Table 6: Energy footprint of density functional theory (DFT) relaxations used to train GAME-Net-UQ. Details on the estimations can be found in **Supplementary Note 12**.

Surface	Avg. CPU hours	Avg. consumption [kWh]
Ferromagnetic (Fe, Ni, Co)	307.03	2.54
Diamagnetic (all others)	135.86	1.13
Absent (gas)	6.82	0.08

Table 7: Performance metrics on the test set for GAME-Net-UQ. MAE=Mean Absolute Error. RMSE=Root Mean Squared Error. MDAE=Median Absolute Error. Sha=Sharpness. C_v =Variation Coefficient. MCA=Miscalibration Area. ρ_S =Spearman’s rank correlation coefficient.

Category	Metric	Unit	All	Intermediates	Transition states
Accuracy	MAE	eV	0.211	0.208	0.222
	RMSE	eV	0.339	0.348	0.301
	MDAE	eV	0.132	0.123	0.171
Uncertainty	Sha	eV	0.229	0.230	0.225
	C_v	-	0.345	0.375	0.178
Calibration	MCA	%	1.25	1.29	1.79
	ρ_S	-	0.818	0.781	0.974
n	-	-	2460	1982	478

Table 8: Ablation study on GAME-Net-UQ (**Supplementary Note 7**) analyzing the effect on the test set ($n=2460$) of TAG convolution, generalized coordination number (gCN), and 2-hop nearest surface neighbours (2-hop). Values are reported in [eV] as mean $\pm$ standard deviation across 5 runs. For each metric, the best (lowest) value is reported in bold, while the worst (highest) after removing only one feature is reported in red. TS=Transition State. MAE=Mean Absolute Error. Sha=Sharpness.

Features			All		Intermediates		Transition States	
TAG	gCN	2-hop	MAE	Sha	MAE	Sha	MAE	Sha
✓	✓	✓	0.20±0.00	0.25±0.01	0.19±0.01	0.25±0.01	0.23±0.00	0.25±0.02
✓	✓	×	0.27±0.09	0.58±0.67	0.26±0.09	0.60±0.75	0.31±0.06	0.39±0.21
×	✓	✓	0.28±0.01	0.45±0.02	0.24±0.01	0.41±0.02	0.46±0.02	0.60±0.02
✓	×	✓	0.24±0.02	0.29±0.03	0.24±0.02	0.28±0.03	0.28±0.02	0.30±0.03
✓	×	×	0.26±0.05	0.40±0.26	0.25±0.05	0.42±0.29	0.28±0.04	0.30±0.07
×	✓	×	0.27±0.00	0.37±0.02	0.22±0.00	0.29±0.02	0.48±0.01	0.60±0.04
×	×	✓	0.36±0.03	0.46±0.03	0.32±0.03	0.44±0.04	0.55±0.03	0.52±0.02
×	×	×	0.31±0.02	0.38±0.03	0.27±0.02	0.36±0.05	0.50±0.01	0.45±0.02

Table 9: Energy evaluators in CARE. The definition of the energy targets can be found in **Eqs. 5**. MLIP=Machine Learning Interatomic Potential.

Family	Type	Elements	Target	# models	Repo version
GAME-Net-UQ	Direct	19	E_{ads}	1	0.1.0
MACE [22]	MLIP	89	E_{tot}	9	0.3.12
Fairchem-v1 [23]	MLIP	56	E_{ads}	67	1.4.0
Orb-v2 [24]	MLIP	89	E_{tot}	5	0.4.2
PET-MAD [25]	MLIP	85	E_{tot}	4	1.4.3
SevenNet [26]	MLIP	89	E_{tot}	5	0.11.2

Table 10: Reaction (ΔE) and activation (E_{act}) energies in [eV] obtained with DFT, GAME-Net-UQ, EquiformerV2-31M and MACE-MP-0 (large), for a subset of reactions in the methanol decomposition network. DFT data come from Supplementary Table 6 of Ref. [2]. Corresponding parity plots can be found in **Supplementary Figure 18**.

Surface	Reaction	ΔE				E_{act}			
		DFT	GAME-Net	Equiformer	MACE	DFT	GAME-Net	Equiformer	MACE
Cu(111)	$\text{CH}^* + * \rightarrow \text{H}^* + \text{C}^*$	1.20	1.72	1.42	2.35	1.87	2.10	2.22	2.94
	$\text{CH}_2^* + * \rightarrow \text{CH}^* + \text{H}^*$	0.38	0.74	0.49	0.85	0.94	1.51	1.07	1.15
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CH}^* + \text{HO}^*$	-0.22	0.30	0.15	-0.01	0.40	0.81	1.26	10.57
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CHO}^* + \text{H}^*$	0.08	0.44	0.26	0.70	0.79	1.08	9.32	1.43
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	0.23	0.43	0.45	0.44	0.72	0.63	11.15	1.21
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.18	0.14	-0.13	-0.14	0.76	1.06	0.82	0.97
	$\text{CH}_2\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_2^*$	0.40	1.14	0.88	0.59	1.35	1.61	12.40	1.34
	$\text{CH}_3^* + * \rightarrow \text{H}^* + \text{CH}_2^*$	0.75	0.99	0.96	0.83	1.33	1.32	1.53	1.19
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	0.41	0.72	0.57	0.46	1.18	0.79	1.21	1.46
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.01	0.43	-0.00	0.92	1.10	1.10	1.07	2.22
	$\text{CH}_3\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_2^*$	-0.19	0.28	0.23	-0.40	1.06	1.02	1.44	1.11
	$\text{CH}_3\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_3^*$	0.54	1.15	0.69	0.13	1.80	1.58	23.09	3.53
	$\text{CH}_4^* + * \rightarrow \text{H}^* + \text{CH}_3^*$	0.64	1.19	1.10	0.22	1.58	1.48	3.22	4.77
	$\text{CH}_4\text{O}^* + * \rightarrow \text{CH}_3\text{O}^* + \text{H}^*$	-0.10	0.41	0.34	-0.05	1.20	0.81	6.72	20.13
	$\text{CH}_4\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_3\text{O}^*$	0.80	0.98	1.12	0.46	1.33	1.12	12.27	5.22
	$\text{CH}_4\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_3^*$	-0.14	0.27	0.39	-0.78	1.91	1.52	2.42	1.33
	$\text{CHO}^* + * \rightarrow \text{CH}^* + \text{O}^*$	0.90	1.44	0.92	1.00	2.18	1.44	1.98	1.98
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-0.89	-0.52	-0.78	-0.01	0.24	0.40	0.39	0.50
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-1.15	-0.82	-1.18	-0.86	1.35	1.00	1.48	0.94
	$\text{CHO}^* + * \rightarrow \text{HO}^* + \text{C}^*$	0.55	1.57	1.30	1.64	1.52	1.57	2.62	2.79
	$\text{CO}^* + * \rightarrow \text{O}^* + \text{C}^*$	2.63	3.68	3.12	3.36	3.71	3.68	4.33	23.73
Pd(111)	$\text{CH}^* + * \rightarrow \text{H}^* + \text{C}^*$	0.18	0.81	0.45	0.21	1.61	0.89	1.62	1.03
	$\text{CH}_2^* + * \rightarrow \text{CH}^* + \text{H}^*$	-0.82	0.05	-0.36	0.22	0.34	0.64	0.42	0.50
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CH}^* + \text{HO}^*$	-0.01	-0.12	0.39	0.87	1.24	1.24	1.34	2.39
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CHO}^* + \text{H}^*$	-0.91	-0.66	-0.52	0.32	0.47	0.55	1.35	1.37
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.96	-0.19	-0.49	-0.47	0.37	0.67	0.37	0.47
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.48	0.02	-0.14	-0.46	0.77	0.61	0.37	0.91
	$\text{CH}_2\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_2^*$	0.05	0.55	0.56	0.72	1.71	2.28	2.05	1.72
	$\text{CH}_3^* + * \rightarrow \text{H}^* + \text{CH}_2^*$	-0.20	0.43	0.27	0.31	0.74	1.15	1.03	0.75
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.54	-0.05	-0.10	-0.01	0.64	0.61	0.95	0.76
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.07	0.16	0.25	0.01	0.81	0.61	0.98	1.80
	$\text{CH}_3\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_2^*$	0.27	-0.22	0.65	0.64	1.44	1.25	1.69	1.54
	$\text{CH}_3\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_3^*$	-0.49	0.45	0.07	-0.22	1.13	1.67	10.90	0.91
	$\text{CH}_4^* + * \rightarrow \text{H}^* + \text{CH}_3^*$	-0.11	0.15	0.48	0.01	0.63	1.06	3.94	3.71
	$\text{CH}_4\text{O}^* + * \rightarrow \text{CH}_3\text{O}^* + \text{H}^*$	0.35	0.28	0.54	0.25	0.95	0.74	1.80	13.56
	$\text{CH}_4\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_3\text{O}^*$	-0.33	0.46	0.08	-0.38	0.64	1.14	3.53	2.29
	$\text{CH}_4\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_3^*$	0.14	-0.20	0.46	-0.05	1.75	1.60	2.63	1.33
	$\text{CHO}^* + * \rightarrow \text{CH}^* + \text{O}^*$	0.19	0.79	0.69	1.41	2.54	2.08	20.27	2.38
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-1.45	-1.34	-1.24	-0.62	0.33	0.50	5.47	0.23
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-1.03	-0.66	-0.86	-1.40	1.00	0.56	0.93	0.85
	$\text{CHO}^* + * \rightarrow \text{HO}^* + \text{C}^*$	1.08	1.34	1.36	0.76	2.11	1.34	2.41	1.95
	$\text{CO}^* + * \rightarrow \text{O}^* + \text{C}^*$	1.82	2.93	2.38	2.24	3.72	2.93	4.01	4.39
Pt(111)	$\text{CH}^* + * \rightarrow \text{H}^* + \text{C}^*$	0.47	0.77	0.84	1.32	1.77	0.85	1.51	1.51
	$\text{CH}_2^* + * \rightarrow \text{CH}^* + \text{H}^*$	-0.67	-0.36	-0.29	0.04	0.44	0.59	5.51	7.42
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CH}^* + \text{HO}^*$	0.30	0.15	0.42	0.49	1.34	1.46	2.01	8.82
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CHO}^* + \text{H}^*$	-0.62	-0.92	-0.18	0.31	0.82	0.46	0.80	1.23
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.75	-0.37	-0.55	-0.18	0.39	0.80	0.24	0.80
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.06	0.05	0.14	0.11	0.45	0.54	0.33	0.70
	$\text{CH}_2\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_2^*$	0.18	0.58	0.29	0.25	1.84	2.38	2.25	1.91
	$\text{CH}_3^* + * \rightarrow \text{H}^* + \text{CH}_2^*$	0.06	0.50	0.39	0.12	0.97	0.90	1.12	1.00
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.29	-0.28	0.03	0.17	0.73	0.55	1.10	0.91
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	0.40	0.14	0.71	0.46	0.94	0.63	1.06	1.77
	$\text{CH}_3\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_2^*$	0.68	0.22	0.73	0.62	1.63	1.58	1.49	6.63
	$\text{CH}_3\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_3^*$	-0.34	0.16	-0.15	-0.37	1.81	2.08	2.17	1.88
	$\text{CH}_4^* + * \rightarrow \text{H}^* + \text{CH}_3^*$	-0.18	0.14	0.24	0.10	0.75	1.53	4.80	3.37
	$\text{CH}_4\text{O}^* + * \rightarrow \text{CH}_3\text{O}^* + \text{H}^*$	0.50	0.43	0.61	0.62	0.87	0.81	3.07	17.93
	$\text{CH}_4\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_3\text{O}^*$	-0.35	0.37	-0.15	-0.34	0.81	1.26	3.66	9.50
	$\text{CH}_4\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_3^*$	0.26	0.10	0.19	0.15	1.94	1.70	2.90	1.08
	$\text{CHO}^* + * \rightarrow \text{CH}^* + \text{O}^*$	0.27	0.59	0.55	0.47	2.81	2.20	2.40	5.86
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-1.06	-1.59	-0.63	-0.43	0.43	0.64	0.86	0.32
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-0.49	-0.62	-0.31	-0.63	1.23	0.55	0.98	0.76
	$\text{CHO}^* + * \rightarrow \text{HO}^* + \text{C}^*$	1.40	1.84	1.44	1.50	2.37	1.84	2.23	1.79
	$\text{CO}^* + * \rightarrow \text{O}^* + \text{C}^*$	1.79	2.95	2.02	2.22	4.33	3.09	4.71	3.32
Ru(0001)	$\text{CH}^* + * \rightarrow \text{H}^* + \text{C}^*$	0.04	0.66	-0.46	1.19	0.82	0.82	1.04	1.69
	$\text{CH}_2^* + * \rightarrow \text{CH}^* + \text{H}^*$	-0.53	-0.34	-1.37	-0.26	0.07	0.54	0.04	0.42
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CH}^* + \text{HO}^*$	-1.17	-0.59	-1.73	-0.14	0.46	0.96	0.56	2.30
	$\text{CH}_2\text{O}^* + * \rightarrow \text{CHO}^* + \text{H}^*$	-0.95	-0.99	-1.61	-0.61	0.14	0.45	1.09	0.36
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.65	-0.26	-1.20	-0.36	0.22	0.61	1.14	16.38
	$\text{CH}_2\text{O}^* + * \rightarrow \text{H}^* + \text{CHO}^*$	-0.80	-0.39	-1.31	-0.06	0.61	0.84	0.50	2.53
	$\text{CH}_2\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_2^*$	-1.25	-0.45	-1.74	0.07	0.83	0.72	0.97	1.63
	$\text{CH}_3^* + * \rightarrow \text{H}^* + \text{CH}_2^*$	-0.24	0.20	-0.86	0.03	0.48	0.83	0.52	0.61
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.24	-0.29	-1.10	-0.44	0.33	0.53	0.33	0.43
	$\text{CH}_3\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_2\text{O}^*$	-0.30	-0.42	-1.21	-0.14	0.51	0.92	0.78	0.35
	$\text{CH}_3\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_2^*$	-0.88	-0.54	-1.45	-0.31	0.77	1.01	0.75	0.64
	$\text{CH}_3\text{O}^* + * \rightarrow \text{O}^* + \text{CH}_3^*$	-1.01	-0.20	-1.48	-0.47	1.64	1.08	4.79	1.12
	$\text{CH}_4^* + * \rightarrow \text{H}^* + \text{CH}_3^*$	-0.43	-0.11	-1.38	-0.46	0.72	1.04	7.48	1.53
	$\text{CH}_4\text{O}^* + * \rightarrow \text{CH}_3\text{O}^* + \text{H}^*$	-0.61	-0.43	-1.21	0.17	0.83	0.74	3.61	0.93
	$\text{CH}_4\text{O}^* + * \rightarrow \text{H}^* + \text{CH}_3\text{O}^*$	-0.22	0.44	-0.59	-0.21	0.64	1.31	7.40	1.31
	$\text{CH}_4\text{O}^* + * \rightarrow \text{HO}^* + \text{CH}_3^*$	-0.86	-0.30	-1.18	-0.55	1.77	1.44	4.16	2.67
	$\text{CHO}^* + * \rightarrow \text{CH}^* + \text{O}^*$	-0.19	-0.53	-1.92	0.17	1.20	1.54	1.00	1.44
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-0.81	-1.16	-1.90	-1.03	0.10	0.54	0.39	0.08
	$\text{CHO}^* + * \rightarrow \text{CO}^* + \text{H}^*$	-0.66	-0.56	-1.60	-0.47	0.96	0.75	1.23	0.97
	$\text{CHO}^* + * \rightarrow \text{HO}^* + \text{C}^*$	-1.13	1.06	-0.58	1.67	1.11	1.06	2.11	3.06
	$\text{CO}^* + * \rightarrow \text{O}^* + \text{C}^*$	-0.29	1.29	-0.48	2.39	1.95	1.84	2.66	4.09

Table 11: Experimental data from CatTestHub [27] under the same operating conditions: Support=Carbon, $T=473$ [K], $P_{\text{Total}}=102.73$ [kPa], Inlet $P_{\text{MeOH}}=10.27$ [kPa], Inlet inert molar fraction=90.01 [%], Inlet MeOH molar fraction=9.99 [%]. wl=weight loading [%] MW=Molecular Weight [g mol⁻¹], D=Dispersion [%], STY_{CO}=Space Time Yield [mol_{CO} mol_{metal}⁻¹ h⁻¹], S_{CO}=Selectivity CO [%].

Metal	wl	MW	D	STY _{CO}	S _{CO}
Ag	5	107.87	11.1	0.32	57
Au	5	196.97	2.7	3.85	64
Cu	5	63.55	2.9	0.54	48
Ir	1	192.22	15.1	92.39	78
Ni	5	58.69	0.2	45.96	93
Pd	5	106.42	11.5	18.30	97
Pt	5	195.08	28.5	43.84	97
Rh	5	102.91	59.7	5.19	95
Ru	5	101.07	21.0	73.43	96

Table 12: Settings and performance metrics for the comparison between GAME-Net-UQ, EquiformerV2-31M [21], and MACE-MP-0 [22] for evaluating the networks for the MeOH case study. “Configurations” refer to the number of screened configurations for the same adsorbate/surface pair. fmax=Force threshold for relaxation stopping criterion; nsteps=Maximum number of relaxation steps. TS=Transition State; CI-NEB=Climbing Image-Nudged Elastic Band; UQ=Uncertainty Quantification; MAE=Mean Absolute Error. RMSE=Root Mean Squared Error. $n = 9$. Target of the benchmark for which MAE and RMSE are computed is the experimental CO logarithmic space-time yield (STY) from CatTestHub [27]. Total time refers to time required to evaluate the energies of all 63 CRNs included in the MeOH case study. Total time distribution and predictions from microkinetic simulations for the studied metals are depicted in **Supplementary Figures 19-20**.

	GAME-Net-UQ	EquiformerV2	MACE-MP-0
Method	Direct	Relaxation	Relaxation
Configurations	3	3	3
fmax [eV/Å]	-	0.05	0.05
nsteps	-	250	250
Model size	0.5M	31M	15.8M
dtype	float32	float32	float32
TS evaluation	Direct	CI-NEB	CI-NEB
NEB images	-	5	5
NEB steps	-	50	50
UQ	✓	×	×
MAE _{log(STY)}	11.05	57.5	32.3
RMSE _{log(STY)}	14.41	63.0	35.9
Total GPU time [h]	0.78	158.9	273.7

Table 13: Predicted CO adsorption energy and formation rates, the most abundant surface intermediate (MASI) and its coverage θ for the different metal catalysts and models employed in the MeOH decomposition case study (**Supplementary Note 10**). Energies are in [eV], rates in [molec s⁻¹ active site⁻¹], and coverages are unitless.

Metal	hkl	Area [%]	GAME-Net-UQ (0.5M)			EquiformerV2-31M			MACE-MP-0 (15.8M)		
			$E_{\text{ads}}^{\text{CO}}$	r_{CO}	MASI (θ)	$E_{\text{ads}}^{\text{CO}}$	r_{CO}	MASI (θ)	$E_{\text{ads}}^{\text{CO}}$	r_{CO}	MASI (θ)
Ag	100	18	+0.39	3.35e-24	CH ₄ O* (1.00)	-0.14	4.72e-61	CH ₄ O* (1.00)	-1.04	5.31e-24	CH ₃ O* (1.00)
	110	2	+0.52	1.02e-25	CH ₄ O* (1.00)	-0.21	4.49e-18	* (0.73)	-1.17	1.71e-17	CH* (0.98)
	111	8	+0.28	1.58e-24	CH ₄ O* (1.00)	-0.04	1.16e-75	CH ₃ O* (1.00)	-0.96	2.63e-20	CH* (1.00)
	322	62	-0.31	1.46e-26	CH ₄ O* (1.00)	-0.18	4.34e-21	* (1.00)	-1.16	3.35e-11	* (0.99)
	332	10	-0.76	6.34e-21	CH ₄ O* (1.00)	-0.13	2.69e-14	* (0.93)	-1.19	1.22e-13	CH* (0.63)
Au	100	9	+0.08	4.94e-15	CH ₄ O* (1.00)	-0.28	8.22e-57	CH ₃ O* (1.00)	-1.38	2.53e-25	CH ₄ O* (0.48)
	111	24	+0.04	4.42e-08	CH ₄ O* (1.00)	-0.13	6.63e-58	CH ₃ O* (0.97)	-1.18	7.12e-16	CH* (1.00)
	322	46	-0.20	3.61e-16	CH ₄ O* (1.00)	-0.47	1.57e-71	CH ₃ O* (1.00)	-1.54	6.47e-23	CH ₃ O* (0.98)
	332	21	-0.23	4.07e-17	CH ₄ O* (1.00)	-0.53	1.81e-69	CH ₃ O* (1.00)	-1.13	9.83e-28	CH* (1.00)
Cu	100	19	-0.36	4.19e-14	CH ₄ O* (1.00)	-0.56	7.38e-57	CH ₄ O* (0.98)	-1.80	1.69e-23	CH* (1.00)
	111	45	-0.42	2.74e-11	CH ₄ O* (0.95)	-0.53	2.64e-37	CH ₄ O* (0.95)	-1.43	3.85e-22	CH* (1.00)
	210	3	-0.13	5.11e-15	CH ₄ O* (1.00)	-0.82	6.10e-59	CH ₄ O* (1.00)	-1.70	3.47e-29	CH* (1.00)
	221	6	-0.59	1.96e-09	CH ₃ O* (0.71)	-0.61	1.92e-40	CH ₄ O* (1.00)	-1.57	6.98e-24	CH ₃ * (1.00)
	310	1	-0.51	1.37e-11	CH ₄ O* (0.99)	-0.79	2.76e-51	CH ₄ O* (1.00)	-1.76	8.29e-35	CH ₄ O* (0.96)
	311	10	-0.49	1.18e-10	CH ₄ O* (0.94)	-0.76	1.82e-35	CH ₄ O* (1.00)	-1.52	2.47e-23	CH* (1.00)
	322	5	-0.42	9.60e-19	CH ₄ O* (0.80)	-0.71	5.03e-36	CH ₄ * (0.83)	-1.47	2.23e-16	CH ₃ * (0.84)
	331	6	-0.79	4.91e-09	CH ₄ O* (0.96)	-0.78	5.45e-22	* (1.00)	-1.60	4.37e-27	HO* (1.00)
Ir	332	5	-1.01	1.59e-09	CH ₂ O* (0.95)	-0.67	4.85e-35	CH ₄ O* (1.00)	-1.47	1.20e-17	CH* (1.00)
	100	8	-1.60	9.94e-18	CH* (0.50)	-1.73	4.12e-35	CH ₄ O* (0.54)	-2.31	1.86e-13	CH ₃ * (0.94)
	111	36	-1.43	1.25e-11	CO* (0.94)	-1.78	5.06e-42	CH ₄ * (0.72)	-1.83	3.56e-21	CH ₄ O* (0.52)
	211	5	-1.13	1.72e-08	CO* (0.84)	-2.19	2.74e-43	CH ₄ O* (0.98)	-1.74	1.10e-15	CH ₂ * (0.66)
	221	10	-1.11	3.13e-08	CO* (1.00)	-2.12	4.39e-27	CH ₄ * (0.83)	-2.30	5.01e-27	CH ₃ * (0.94)
	311	6	-1.06	9.81e-08	CO* (0.80)	-2.14	3.37e-25	C* (0.99)	-2.28	2.13e-16	CH ₂ * (1.00)
	322	16	-1.02	3.12e-07	CO* (0.99)	-2.16	7.74e-53	CH ₄ * (0.84)	-2.56	8.78e-26	CH ₂ * (0.91)
	332	19	-1.13	2.08e-08	CO* (1.00)	-2.13	8.84e-37	CH ₄ O* (1.00)	-2.31	3.73e-35	CH ₂ * (1.00)
Ni	100	19	-1.31	2.77e-22	CH ₃ * (1.00)	-1.72	9.72e-47	CH ₄ O* (1.00)	-0.28	1.22e-17	CH ₃ O* (1.00)
	110	1	-1.25	5.51e-23	CH ₃ * (1.00)	-1.83	1.72e-50	CH ₄ O* (1.00)	-0.51	3.05e-15	CH ₄ O* (1.00)
	111	60	-1.26	2.44e-13	CH ₃ O* (0.95)	-1.79	3.34e-47	CH ₄ O* (0.90)	-0.32	7.42e-22	CH ₄ O* (0.66)
	210	1	-0.85	1.19e-07	CH ₄ O* (0.93)	-1.76	5.13e-45	CH ₄ O* (1.00)	-0.40	3.32e-17	CH ₄ O* (0.71)
	311	4	-1.03	1.41e-07	CO* (0.55)	-1.86	1.35e-46	CH ₄ O* (1.00)	-0.49	1.23e-16	CH ₃ O* (0.53)
	322	6	-1.51	2.08e-17	CH ₄ O* (0.86)	-1.77	1.03e-32	CH ₄ * (0.59)	-0.26	2.71e-18	CH* (0.99)
	331	5	-1.22	1.02e-10	CH ₄ O* (0.95)	-1.78	3.41e-25	C* (1.00)	-0.36	2.86e-18	CH ₄ O* (1.00)
Pd	332	4	-1.49	2.93e-12	CO* (0.99)	-1.71	5.93e-50	CH ₄ O* (1.00)	-0.18	1.68e+00	CH ₂ * (0.57)
	100	16	-1.36	7.00e-11	CO* (0.93)	-1.68	7.79e-45	CH ₄ * (0.56)	-2.08	3.44e-28	CH ₃ * (0.65)
	111	39	-1.33	1.56e-10	CO* (0.99)	-1.63	3.44e-34	CH ₄ O* (1.00)	-1.93	3.13e-25	CH ₃ * (0.98)
	210	3	-0.90	1.42e-10	CH ₄ O* (0.98)	-1.62	6.45e-42	CH ₄ O* (1.00)	-2.23	1.87e-43	CH ₄ O* (1.00)
	221	5	-1.11	1.69e-08	CO* (0.54)	-1.62	1.24e-33	CH ₄ O* (1.00)	-2.15	1.30e-29	CH ₃ * (0.76)
	311	7	-1.02	3.21e-07	CO* (0.98)	-1.69	2.57e-35	CH ₄ O* (0.50)	-2.16	2.93e-31	CH ₂ * (0.94)
	322	20	-0.96	7.38e-07	CO* (0.53)	-1.59	4.93e-36	CH ₄ * (0.61)	-2.19	3.04e-33	C* (1.00)
Pt	331	10	-1.22	2.57e-09	CO* (1.00)	-1.65	1.32e-38	CH ₄ O* (1.00)	-2.22	3.60e-33	C* (1.00)
	100	5	-1.51	1.63e-12	CO* (0.95)	-1.85	5.19e-22	* (1.00)	-2.32	8.10e-27	CH* (0.87)
	110	5	-1.55	6.07e-13	CO* (0.92)	-1.93	3.03e-43	CH ₄ O* (0.83)	-2.37	1.23e-32	CH* (0.89)
	111	27	-1.37	5.42e-11	CO* (0.93)	-1.44	8.57e-30	CH ₄ O* (1.00)	-1.87	2.93e-27	* (0.86)
	221	18	-1.12	6.82e-10	CH ₄ O* (0.97)	-1.82	2.51e-45	CH ₄ * (0.83)	-2.28	1.74e-28	CH ₄ O* (0.63)
	311	9	-1.02	2.29e-07	CO* (0.73)	-1.88	2.44e-23	CH* (0.51)	-2.05	5.54e-22	CH* (0.95)
	322	25	-0.96	1.15e-06	CO* (0.85)	-1.87	2.88e-44	CH ₄ * (0.83)	-2.30	9.23e-39	CH ₄ O* (0.78)
Rh	332	11	-0.96	1.27e-06	CO* (0.99)	-1.81	5.96e-37	CH ₄ O* (1.00)	-2.30	3.05e-29	CH ₄ O* (0.88)
	100	14	-1.35	3.93e-11	CH ₃ * (0.56)	-1.79	1.86e-38	CH ₄ O* (1.00)	-2.29	3.45e-28	CH ₂ * (1.00)
	110	5	-1.05	6.80e-09	CH ₄ O* (0.89)	-1.76	9.89e-36	CH ₄ O* (1.00)	-2.33	6.44e-22	CH ₂ * (0.77)
	111	59	-1.44	1.02e-11	CO* (0.99)	-1.68	5.00e-33	CH ₄ * (0.75)	-2.09	6.37e-30	CH ₃ * (0.97)
	211	4	-1.09	5.35e-08	CO* (0.85)	-1.75	1.62e-48	CH ₄ * (1.00)	-2.42	6.13e-26	CHO* (0.94)
	221	8	-1.13	4.30e-09	CH ₄ O* (0.40)	-1.76	5.46e-27	CH ₄ O* (0.65)	-2.33	4.46e-33	CH ₃ * (0.52)
	310	1	-0.38	2.01e-03	CH ₄ O* (0.96)	-1.81	3.92e-42	CH ₄ O* (1.00)	-2.46	1.48e-22	CO* (0.99)
Ru	311	7	-1.06	8.54e-08	CO* (0.74)	-1.81	1.77e-41	CH ₄ O* (1.00)	-2.38	2.16e-22	CH ₃ O ₂ * (0.55)
	322	2	-1.00	1.35e-07	CH ₃ O* (0.53)	-1.84	7.81e-39	CH ₄ * (0.85)	-2.39	1.13e-25	CH ₂ * (0.51)
	1010	21	-1.27	2.77e-10	CH ₃ O* (0.58)	-2.18	2.73e-45	CH ₄ O* (1.00)	-2.29	1.22e-25	CHO* (1.00)
	1011	43	-1.29	1.23e-11	CH ₃ O* (0.97)	-2.08	3.12e-41	CH ₄ * (0.77)	-2.39	2.43e-27	CH* (1.00)
	1012	10	-0.84	2.54e-05	CO* (0.98)	-1.99	1.39e-34	CH ₃ O* (0.97)	-2.49	2.08e-26	CH ₃ * (0.82)
	1121	1.34	-0.67	8.79e-07	CH ₄ O* (0.95)	-1.77	6.36e-33	C* (1.00)	-2.35	4.83e-29	CH* (0.99)
	2131	1.33	-1.10	4.55e-08	CO* (1.00)	-1.84	1.62e-47	CH ₄ O* (0.99)	-2.47	1.70e-38	CH ₂ * (1.00)
	2132	1.33	-0.96	5.34e-07	CO* (0.39)	-1.75	6.82e-44	CH ₄ O* (1.00)	-2.37	2.38e-29	CH* (1.00)
	2112	6	-0.75	2.89e-06	CH ₄ O* (0.74)	-1.74	5.17e-29	C* (1.00)	-2.34	5.67e-29	CH ₂ * (0.99)
	0001	16	-1.37	3.35e-12	CH ₃ O* (0.44)	-1.98	4.06e-30	CH ₃ * (0.97)	-2.36	3.75e-28	CH ₃ * (0.94)

Table 14: Statistics on the success rate of GAME-Net-UQ in predicting valid transition state (TS) energies higher than the initial (IS) and final (FS) state energies $E_{\text{TS}} > (E_{\text{IS}}, E_{\text{FS}})$ for the 36580 surface reactions in the Fischer-Tropsch reaction networks used to run the microkinetic simulations. All reactions are considered in the bond-breaking direction.

	Co(0001)	Fe(110)	Ni(111)	Ru(0001)	Total
n	36580	36580	36580	36580	146320
Valid TS	24524	31095	32160	33736	121515
Invalid TS	12056	5485	4420	2844	24805
Invalid TS ($\Delta E < 0$)	3573	513	304	113	4503
Invalid TS ($\Delta E > 0$)	8483	4972	4116	2731	20302
Success rate [%]	67.04	85.01	87.92	92.23	83.05
Failure rate [%]	32.96	14.99	12.08	7.77	16.95

Table 15: Top-3 intermediates by surface coverage θ obtained from the kinetic simulations of the Fischer-Tropsch process at $T = 523$ K, $P = 10$ bar and $\text{H}_2/\text{CO}=2$.

Surface	Rank	Species	θ [-]
Co(0001)	1	H	0.655
	2	CO	0.341
	3	CH ₃ O	0.003
Fe(110)	1	H	0.676
	2	CH ₃ O	0.302
	3	CO	0.015
Ni(111)	1	CO	0.988
	2	H	0.009
	3	CHO	0.001
Ru(0001)	1	CHO	0.892
	2	CO	0.102
	3	C ₂ H ₃ O	0.004

Table 16: Top-10 gas products by formation rate obtained from the kinetic simulations of the Fischer-Tropsch process at $T = 523$ K, $P = 10$ bar and $H_2/CO=2$.

Surface	Rank	Formula	Formation Rate [s^{-1}]
Co(0001)	1	CH ₄ O	1.271×10^{-6}
	2	CH ₂ O	7.179×10^{-11}
	3	H ₂ O	2.618×10^{-12}
	4	C ₂ H ₆ O	2.617×10^{-12}
	5	C ₂ H ₄ O	6.926×10^{-16}
	6	C ₂ H ₄ O	1.918×10^{-16}
	7	CH ₄	7.345×10^{-18}
	8	C ₆ H ₁₄ O	1.271×10^{-19}
	9	C ₃ H ₈ O	1.276×10^{-20}
	10	C ₆ H ₁₂ O	1.115×10^{-20}
Fe(110)	1	CH ₄ O	2.893×10^{-6}
	2	H ₂ O	2.411×10^{-9}
	3	C ₂ H ₆ O	2.280×10^{-9}
	4	CH ₂ O	1.302×10^{-12}
	5	CO ₂	5.957×10^{-13}
	6	C ₂ H ₄ O	2.902×10^{-14}
	7	C ₂ H ₄ O	3.966×10^{-15}
	8	C ₃ H ₈ O	2.669×10^{-15}
	9	CH ₄	2.064×10^{-18}
	10	C ₂ H ₂ O	5.888×10^{-19}
Ni(111)	1	CH ₄ O	4.071×10^{-12}
	2	CH ₂ O	3.612×10^{-15}
	3	H ₂ O	8.396×10^{-17}
	4	C ₂ H ₆ O	7.410×10^{-17}
	5	C ₂ H ₂ O	2.364×10^{-19}
	6	C ₂ H ₄ O	4.500×10^{-20}
	7	C ₂ H ₄ O	8.305×10^{-21}
	8	CO ₂	8.224×10^{-21}
	9	C ₃ H ₆ O	7.139×10^{-23}
	10	C ₂ H ₄	4.720×10^{-23}
Ru(0001)	1	CH ₄ O	9.568×10^{-13}
	2	H ₂ O	1.519×10^{-15}
	3	C ₂ H ₆ O	2.376×10^{-16}
	4	CH ₂ O	1.791×10^{-16}
	5	C ₃ H ₈ O	5.222×10^{-17}
	6	CO ₂	6.125×10^{-19}
	7	C ₂ H ₄ O	2.414×10^{-19}
	8	C ₂ H ₄ O	4.744×10^{-20}
	9	C ₄ H ₁₀ O	4.471×10^{-21}
	10	C ₃ H ₆ O	4.033×10^{-21}

Table 17: Top-3 C-C forming reactions by net rate obtained from the kinetic simulations of the Fischer-Tropsch process for Co(0001) and Fe(110) at $T = 523$ K, $P = 10$ bar and $H_2/CO=2$.

Surface	Type	Rank	Reaction	Rate [s^{-1}]
Co(0001)	C ₂	1	$CH_2^* + CH_3O^* \longrightarrow C_2H_5O^* + ^*$	2.601×10^{-12}
		2	$CH_2^* + CHO^* \longrightarrow C_2H_3O^* + ^*$	1.449×10^{-14}
		3	$CH_2^* + CHO^* \longrightarrow C_2H_3O^* + ^*$	1.761×10^{-15}
	C ₃	1	$C_2HO^* + CH_2^* \longrightarrow C_3H_3O^* + ^*$	3.703×10^{-21}
		2	$C_2H_3^* + CH_3O^* \longrightarrow C_3H_6O^* + ^*$	4.746×10^{-22}
		3	$C_2H_3O^* + CH^* \longrightarrow C_3H_4O^* + ^*$	3.048×10^{-22}
	C ₄	1	$C_3H_3^* + CH_3O^* \longrightarrow C_4H_6O^* + ^*$	5.293×10^{-22}
		2	$C_3H_3^* + CHO^* \longrightarrow C_4H_4O^* + ^*$	2.133×10^{-22}
		3	$C_3H_4^* + CH_3O^* \longrightarrow C_4H_7O^* + ^*$	1.482×10^{-22}
	C ₅	1	$C_2H_5O^* + C_3H_6^* \longrightarrow C_5H_{11}O^* + ^*$	1.130×10^{-21}
		2	$C_2H_5O^* + C_3H_5^* \longrightarrow C_5H_{10}O^* + ^*$	9.602×10^{-22}
		3	$C_2H_3O^* + C_3H_2^* \longrightarrow C_5H_5O^* + ^*$	6.386×10^{-22}
	C ₆	1	$C_2H_3O^* + C_4H_6^* \longrightarrow C_6H_9O^* + ^*$	7.006×10^{-21}
		2	$C_5H_7O^* + CH_2^* \longrightarrow C_6H_9O^* + ^*$	8.699×10^{-22}
		3	$C_5H_8O^* + CH_2^* \longrightarrow C_6H_{10}O^* + ^*$	5.918×10^{-22}
Fe(110)	C ₂	1	$CH_2^* + CHO^* \longrightarrow C_2H_3O^* + ^*$	2.277×10^{-9}
		2	$CH_2^* + CH_2O^* \longrightarrow C_2H_4O^* + ^*$	1.189×10^{-10}
		3	$C^* + CH_3O^* \longrightarrow C_2H_3O^* + ^*$	1.133×10^{-11}
	C ₃	1	$C_2H_3^* + CH_2O^* \longrightarrow C_3H_5O^* + ^*$	7.568×10^{-14}
		2	$C_2H_3^* + CH_3O^* \longrightarrow C_3H_6O^* + ^*$	5.189×10^{-14}
		3	$C_2H_4^* + CHO^* \longrightarrow C_3H_5O^* + ^*$	3.461×10^{-14}
	C ₄	1	$C_2H_3O^* + C_2H_4^* \longrightarrow C_4H_7O^* + ^*$	5.398×10^{-17}
		2	$C_3H_4^* + CO^* \longrightarrow C_4H_4O^* + ^*$	6.473×10^{-20}
		3	$C_3H_5^* + CO^* \longrightarrow C_4H_5O^* + ^*$	8.987×10^{-21}
	C ₅	1	$C_2H_4^* + C_3H_7O^* \longrightarrow C_5H_{11}O^* + ^*$	1.191×10^{-17}
		2	$C_2H_4^* + C_3H_7O^* \longrightarrow C_5H_{11}O^* + ^*$	4.420×10^{-19}
		3	$C_2H_4^* + C_3H_6O^* \longrightarrow C_5H_{10}O^* + ^*$	8.807×10^{-21}
	C ₆	1	$C_3H_4^* + C_3H_6O^* \longrightarrow C_6H_{10}O^* + ^*$	3.495×10^{-18}
		2	$C_2H_4^* + C_4H_8O^* \longrightarrow C_6H_{12}O^* + ^*$	7.283×10^{-19}
		3	$C_3H_5^* + C_3H_6O^* \longrightarrow C_6H_{11}O^* + ^*$	6.560×10^{-19}

Table 18: Top-3 C-C forming reactions by net rate obtained from the kinetic simulations of the Fischer-Tropsch process for Ni(111) and Ru(0001) at $T = 523$ K, $P = 10$ bar and $H_2/CO=2$.

Surface	Type	Rank	Reaction	Rate [s^{-1}]
Ni(111)	C ₂	1	$C^* + CHO^* \longrightarrow C_2HO^* + *$	8.088×10^{-17}
		2	$C^* + CHO^* \longrightarrow C_2HO^* + *$	2.231×10^{-18}
		3	$C^* + CH_2O^* \longrightarrow C_2H_2O^* + *$	3.858×10^{-19}
	C ₃	1	$C_2H_4^* + CH_2O^* \longrightarrow C_3H_6O^* + *$	2.341×10^{-21}
		2	$C_2H_2^* + CO^* \longrightarrow C_3H_2O^* + *$	1.470×10^{-21}
		3	$C_2H_4^* + CHO^* \longrightarrow C_3H_5O^* + *$	7.096×10^{-22}
	C ₄	1	$C_3H_3^* + CHO^* \longrightarrow C_4H_4O^* + *$	8.498×10^{-27}
		2	$C_3H_4^* + CHO^* \longrightarrow C_4H_5O^* + *$	4.387×10^{-27}
		3	$C_3H_4^* + CHO^* \longrightarrow C_4H_5O^* + *$	2.323×10^{-27}
	C ₅	1	$C^* + C_4H_8O^* \longrightarrow C_5H_8O^* + *$	1.335×10^{-25}
		2	$C^* + C_4H_6O^* \longrightarrow C_5H_6O^* + *$	1.458×10^{-28}
		3	$C_4H_6^* + CHO^* \longrightarrow C_5H_7O^* + *$	1.128×10^{-29}
	C ₆	1	$C_5H_7O^* + CH_3^* \longrightarrow C_6H_{10}O^* + *$	2.374×10^{-23}
		2	$C_2H_2^* + C_4H_8O^* \longrightarrow C_6H_{10}O^* + *$	2.886×10^{-24}
		3	$C_5H_7O^* + CH_3^* \longrightarrow C_6H_{10}O^* + *$	2.486×10^{-24}
Ru(0001)	C ₂	1	$C^* + CH_3O^* \longrightarrow C_2H_3O^* + *$	9.507×10^{-16}
		2	$C^* + CH_2O^* \longrightarrow C_2H_2O^* + *$	3.412×10^{-16}
		3	$C^* + CHO^* \longrightarrow C_2HO^* + *$	4.529×10^{-18}
	C ₃	1	$C^* + C_2H_5O^* \longrightarrow C_3H_5O^* + *$	2.076×10^{-16}
		2	$C_2H_2^* + CH_3O^* \longrightarrow C_3H_5O^* + *$	1.007×10^{-17}
		3	$C_2H_2^* + CO^* \longrightarrow C_3H_2O^* + *$	3.150×10^{-18}
	C ₄	1	$C_3H_4^* + CHO^* \longrightarrow C_4H_5O^* + *$	4.032×10^{-19}
		2	$C_3H_4^* + CH_2O^* \longrightarrow C_4H_6O^* + *$	2.292×10^{-19}
		3	$C_3H_4^* + CH_3O^* \longrightarrow C_4H_7O^* + *$	1.568×10^{-19}
	C ₅	1	$C^* + C_4H_8O^* \longrightarrow C_5H_8O^* + *$	2.191×10^{-22}
		2	$C^* + C_4H_8O^* \longrightarrow C_5H_8O^* + *$	9.859×10^{-23}
		3	$C_2H_5O^* + C_3H_4^* \longrightarrow C_5H_9O^* + *$	4.479×10^{-23}
	C ₆	1	$C_3H_4^* + C_3H_7O^* \longrightarrow C_6H_{11}O^* + *$	5.275×10^{-21}
		2	$C_3H_4^* + C_3H_5O^* \longrightarrow C_6H_9O^* + *$	1.304×10^{-21}
		3	$C_2H_2^* + C_4H_8O^* \longrightarrow C_6H_{10}O^* + *$	1.537×10^{-22}

Table 19: Overlap between transition states seen by GAME-Net-UQ in the training dataset and those present in the reaction networks discussed in this work. Note that in all cases the transition states in the CRNs are directly predicted by GAME-Net-UQ.

Application	Metal	<i>hkl</i>	Transition states in training data	Dissociation steps	Overlap [%]
MeOH decomposition	Ag	111	16	50	32
		322	0	50	0
		332	0	50	0
		100	0	50	0
		110	0	50	0
	Au	111	12	50	24
		322	0	50	0
		332	0	50	0
		100	0	50	0
	Cu	111	17	50	34
		322	0	50	0
		332	0	50	0
		100	0	50	0
		211	0	50	0
		331	0	50	0
		311	0	50	0
		310	0	50	0
		110	0	50	0
	Ir	111	14	50	28
		322	0	50	0
		332	0	50	0
		221	0	50	0
		211	0	50	0
		311	0	50	0
	Ni	111	14	50	28
		322	0	50	0
		332	0	50	0
		100	0	50	0
		331	0	50	0
		110	0	50	0
		311	0	50	0
	Pd	210	0	50	0
		111	16	50	32
		322	0	50	0
		221	0	50	0
		331	0	50	0
		100	0	50	0
		311	0	50	0
	Pt	110	0	50	0
		111	18	50	36
		322	0	50	0
		332	0	50	0
		221	0	50	0
		100	0	50	0
		110	0	50	0
	Rh	311	0	50	0
		310	0	50	0
		111	18	50	36
		322	0	50	0
		221	0	50	0
		211	0	50	0
		110	0	50	0
	Ru	100	0	50	0
		311	0	50	0
		310	0	50	0
		0001	18	50	36
		10 $\bar{1}$ 1	0	50	0
		10 $\bar{1}$ 0	0	50	0
		10 $\bar{1}$ 2	0	50	0
		2 $\bar{1}$ 12	0	50	0
eC ₃ O ₂ RR	Cu	2132	0	50	0
		2131	0	50	0
		1121	0	50	0
		100	0	4439	0
		0001	71	357222	0.02
Fischer-Tropsch	Ru	111	65	357222	0.02
		110	0	357222	0
		0001	79	357222	0.02
		0001	79	357222	0.02

References

1. García-Melchor, M., Bellarosa, L. & López, N. Unique Reaction Path in Heterogeneous Catalysis: The Concerted Semi-Hydrogenation of Propyne to Propene on CeO₂. *ACS Catalysis* **4**, 4015–4020 (2014).
2. García-Muelas, R., Li, Q. & López, N. Density Functional Theory Comparison of Methanol Decomposition and Reverse Reactions on Metal Surfaces. *ACS Catalysis* **5**, 1027–1036 (2015).
3. Weininger, D. Smiles, a Chemical Language and Information System. 1. Introduction To Methodology and Encoding Rules. *Journal of Chemical Information and Modeling* **28**, 31–36 (1988).
4. Heller, S. R., McNaught, A., Pletnev, I., Stein, S. & Tchekhovskoi, D. InChI, the IUPAC International Chemical Identifier. *Journal of Cheminformatics* **7**, 23 (2015).
5. Nørskov, J. K., Rossmeisl, J., Logadottir, A., Lindqvist, L., Kitchin, J. R., Bligaard, T. & Jónsson, H. Origin of the Overpotential for Oxygen Reduction at a Fuel-Cell Cathode. *The Journal of Physical Chemistry B* **108**, 17886–17892 (2004).
6. Martí, C., Blanck, S., Staub, R., Loehlé, S., Michel, C. & Steinmann, S. N. Dock-OnSurf: A Python Code for the High-Throughput Screening of Flexible Molecules Adsorbed on Surfaces. *Journal of Chemical Information and Modeling* **61**, 3386–3396 (2021).
7. Han, S., Lysgaard, S., Vegge, T. & Hansen, H. A. Rapid mapping of alloy surface phase diagrams via Bayesian evolutionary multitasking. *npj Computational Materials* **9**, 139 (2023).
8. Pablo-García, S., Morandi, S., Vargas-Hernández, R. A., Jorner, K., Ivković, Ž., López, N. & Aspuru-Guzik, A. Fast evaluation of the adsorption energy of organic

- molecules on metals via graph neural networks. *Nature Computational Science* **3**, 433–442 (2023).
9. Cordero, B., Gómez, V., Platero-Prats, A. E., Revés, M., Echeverría, J., Cremades, E., Barragán, F. & Alvarez, S. Covalent Radii Revisited. *Dalton Transactions*, 2832 (2008).
 10. Tran, R., Lan, J., Shuaibi, M., Wood, B. M., Goyal, S., Das, A., Heras-Domingo, J., Kolluru, A., Rizvi, A., Shoghi, N., Sriram, A., Therrien, F., Abed, J., Voznyy, O., Sargent, E. H., Ulissi, Z. & Zitnick, C. L. The Open Catalyst 2022 (OC22) Dataset and Challenges for Oxide Electrocatalysts. *ACS Catalysis* **13**, 3066–3084 (2023).
 11. Hamilton, W. L., Ying, R. & Leskovec, J. *Inductive Representation Learning on Large Graphs* 2018. arXiv: [1706.02216 \[cs.SI\]](#).
 12. Du, J., Zhang, S., Wu, G., Moura, J. M. F. & Kar, S. *Topology Adaptive Graph Convolutional Networks* 2018.
 13. Baek, J., Kang, M. & Hwang, S. J. *Accurate Learning of Graph Representations with Graph Multiset Pooling* 2021.
 14. Reddi, S. J., Kale, S. & Kumar, S. On the Convergence of Adam and Beyond. *arXiv* (2019).
 15. Chorkendorff, I. & Niemantsverdriet, J. *Concepts of modern catalysis and kinetics* (Wiley-VCH Verlag, 2003).
 16. Jónsson, H., Mills, G. & Jacobsen, K. W. in *Classical and Quantum Dynamics in Condensed Phase Systems* (eds Berne, B. J., Cicotti, G. & Coker, D. F.) 385–404 (World Scientific, 1998).
 17. Henkelman, G., Uberuaga, B. P. & Jónsson, H. A climbing image nudged elastic band method for finding saddle points and minimum energy paths. *The Journal of Chemical Physics* **113**, 9901–9904 (2000).

18. Jónsson, H. Improved Tangent Estimate in the Nudged Elastic Band Method for Finding Minimum Energy Paths and Saddle Points. *J. Chem. Phys.* **113**, 9978–9985 (2000).
19. Larsen, A. H. *et al.* The atomic simulation environment—a Python library for working with atoms. *Journal of Physics: Condensed Matter* **29**, 273002 (2017).
20. Tran, R., Xu, Z., Radhakrishnan, B., Winston, D., Sun, W., Persson, K. A. & Ong, S. P. Surface energies of elemental crystals. *Scientific Data* **3**, 160080 (2016).
21. Liao, Y.-L., Wood, B., Das, A. & Smidt, T. *EquiformerV2: Improved Equivariant Transformer for Scaling to Higher-Degree Representations* 2024.
22. Batatia, I. *et al.* A foundation model for atomistic materials chemistry. *The Journal of Chemical Physics* **163**, 184110 (2025).
23. Chanussot, L., Das, A., Goyal, S., Lavril, T., Shuaibi, M., Riviere, M., Tran, K., Heras-Domingo, J., Ho, C., Hu, W., Palizhati, A., Sriram, A., Wood, B., Yoon, J., Parikh, D., Zitnick, C. L. & Ulissi, Z. Open Catalyst 2020 (OC20) Dataset and Community Challenges. *ACS Catalysis* **11**, 6059–6072 (2021).
24. Neumann, M., Gin, J., Rhodes, B., Bennett, S., Li, Z., Choubisa, H., Hussey, A. & Godwin, J. Orb: A Fast, Scalable Neural Network Potential. *arXiv* (2024).
25. Mazitov, A., Bigi, F., Kellner, M., Pegolo, P., Tisi, D., Fraux, G., Pozdnyakov, S., Loche, P. & Ceriotti, M. PET-MAD as a lightweight universal interatomic potential for advanced materials modeling. *Nature Communications* **16**, 10653 (2025).
26. Park, Y., Kim, J., Hwang, S. & Han, S. Scalable Parallel Algorithm for Graph Neural Network Interatomic Potentials in Molecular Dynamics Simulations. *J. Chem. Theory Comput.* **20**, 4857–4868 (2024).
27. Burte, A. S., Nair, A., Grabow, L. C., Dauenhauer, P. J., Scott, S. L. & Abdelrahman, O. A. CatTestHub: A benchmarking database of experimental heterogeneous catalysis for evaluating advanced materials. *Journal of Catalysis* **442**, 115902 (2025).