

Supporting Information for

Improving Reaction Prediction through Chemically Aware Transfer Learning

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Table of Contents

<i>Cope and Claisen Selectivity</i>	S3
<i>Datasets</i>	S3
Cope and Claisen	S3
Ene.....	S4
Nazarov	S4
Artificial Cope and Claisen Rearrangements	S4
<i>Further Prediction Results</i>	S5
Dataset Performances	S5
Chemformer	S7
<i>GitHubs</i>	S8
<i>Artificial Dataset SMILES</i>	S8
<i>Reaxys Dataset IDs</i>	S51
<i>References</i>	S78

Cope and Claisen Selectivity

Datasets

The pericyclic datasets were all extracted from Reaxys¹ and filtered using workflows. The Diels–Alder dataset was created in a previous study and the explanations can be found in Keto et al.² In this study, datasets for the Cope and Claisen, Ene, and Nazarov reactions were created.

Cope and Claisen

From Reaxys, 9,543 unique reactions were extracted when using Cope and Claisen related search terms. Bonding patterns were used to identify reactants. In cases of multiple possible reactants, atom economy was used. Any reactant and product combinations whose weights deviated more than 0.1 AMU were removed. Only 5 reactions had multiple reactants that needed to be identified manually. Products whose double bond count differed from reactants were also removed. After identifying possible reactions, atom mapping was used to verify that the change in bonding occurred on reaction centers. RXNMapper³ was used for this. Finally, the reactions module in RDKit⁴ and a Cope and Claisen reaction template was applied to the reactants to produce a list of possible products. The reaction was kept if the reported product was contained in this list or if a tautomerization of the reported product was in this list. The final dataset contained 3,289 reactions. If the reported product was a tautomer, the non-tautomerized product (from RDKit) was used.

Cope and Claisen rearrangements display diastereoselectivity and site-selectivity. The former is not relevant for the NERF model but can nonetheless form depending on the E/Z arrangement of the double bonds in the reaction centre and the R/S chirality of the SP₃ hybridized carbons. Site-selectivity arises when there are two or more reaction centres. This can be seen when aromatic systems contribute one of the double bonds. The aromatic ring can produce two different outcomes depending on which double bond is used (Fig S1). This can also appear in aliphatic systems.

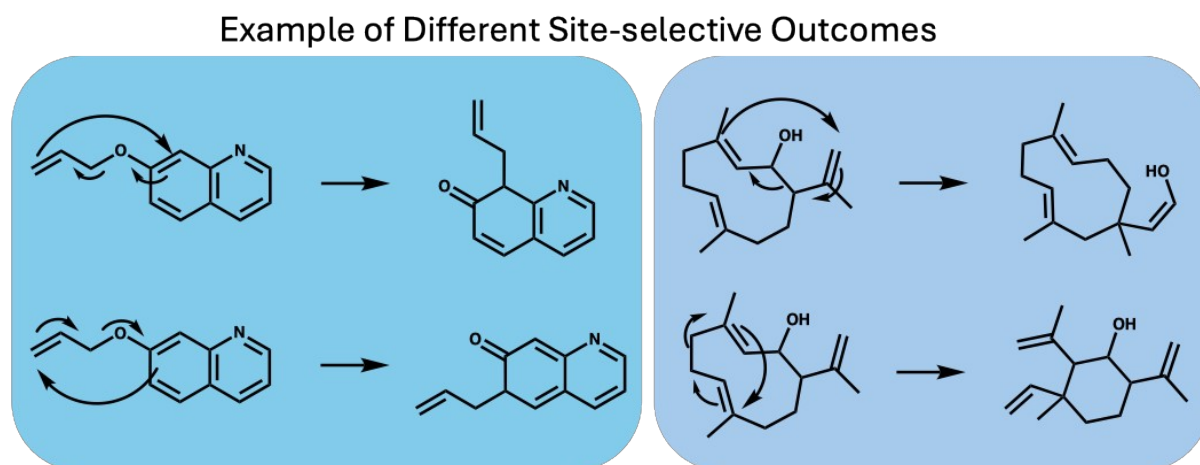


Figure S1. A) The origin of different diastereoselective reaction outcomes. B) Examples of site-selective reactions outcomes present in the Cope and Claisen dataset.

Ene

From Reaxys, 3,457 unique reactions were extracted using Ene related search terms. Reactant and product combinations were sorted by the same atom economy filter as with Cope and Claisen reactions, eliminating any reactant and product combinations whose weights differed more than 0.1 AMU. The same product and tautomer enumeration method as for the Cope and Claisen rearrangement was applied here but with an Ene reaction template. RXNMapper was not used to further verify these reactions because the atom mappings were very poor, often mistaking rearrangements for strange substitutions. However, the product enumeration was sufficient for verifying the reaction. The Ene dataset consisted of 2,322 unique reactions.

Nazarov

A dataset of 3,356 unique reactions was extracted from Reaxys using Nazarov related search terms. Atom economy was used once again to identify viable product and reactant combinations. Reactants were then verified by detecting the 1,4-pentadiene bonding pattern. This dataset was originally intended for another study and so only carbon atoms were considered in the 1,4-pentadiene substructure. The reactions were then verified using atom mapping from RXNMapper to check the changing atoms were located on a viable reaction centre. There were 961 reactions with one reaction centre but 86 with two or more possible reaction centres which had to be evaluated manually. The protonated dataset involved finding the divinyl ketone and protonating the ketone using RDKit. The final dataset contained 1,029 Nazarov reactions.

Artificial Cope and Claisen Rearrangements

Artificial data was generated by developing a workflow¹ that randomly created SMILES strings:

1. 10,000 Functional group SMILES were randomly written according to:
 - a. Atoms were selected according to the following weightings:
 - i. C:20, N:2, O:2, S:2, H:5, Cl:1, F:1, Br:1, I:1, P:1
 1. Ex: Carbon appears 10x as often as nitrogen and 20x as often as phosphorus.
 2. Hydrogens can prematurely end a functional group early.
 - b. Only single bonds were allowed.
 - c. Functional groups could not have a main chain longer than 7 atoms.
2. 20 Reaction centres SMILES were randomly written according to:
 - a. Atoms were selected according to the following weightings:
 - i. C:40, N:2, O:2, S:1
3. Functional groups are randomly added to these reaction centres to create 12,000 molecules.
 - a. At each atom of a reaction centre, there was a 25% chance of having a functional group.
4. These molecules undergo a Cope and Claisen reaction using the reactions module in RDKit and RDChiral.⁵
 - a. If multiple unique products were possible, one was randomly selected.

¹ The Python workflow for artificial SMILES generation was written with assistance from ChatGPT.

Further Prediction Results

Dataset Performances

As a preliminary assessment, we trained NERF on the Diels–Alder, Cope and Claisen, Ene, Nazarov (protonated), and Nazarov (unprotonated) reaction datasets without pretraining (Fig S2). NERF can exceed 90% Top-1 accuracy, when using 80% of the dataset as training, for the Diels–Alder and Cope and Claisen rearrangements. The Ene reaction performs similarly well to the Cope and Claisen despite being a smaller dataset (2,322 vs 3,289 reactions). Nazarov cyclizations however prove more difficult, likely because of their small dataset of 1,029 reactions.

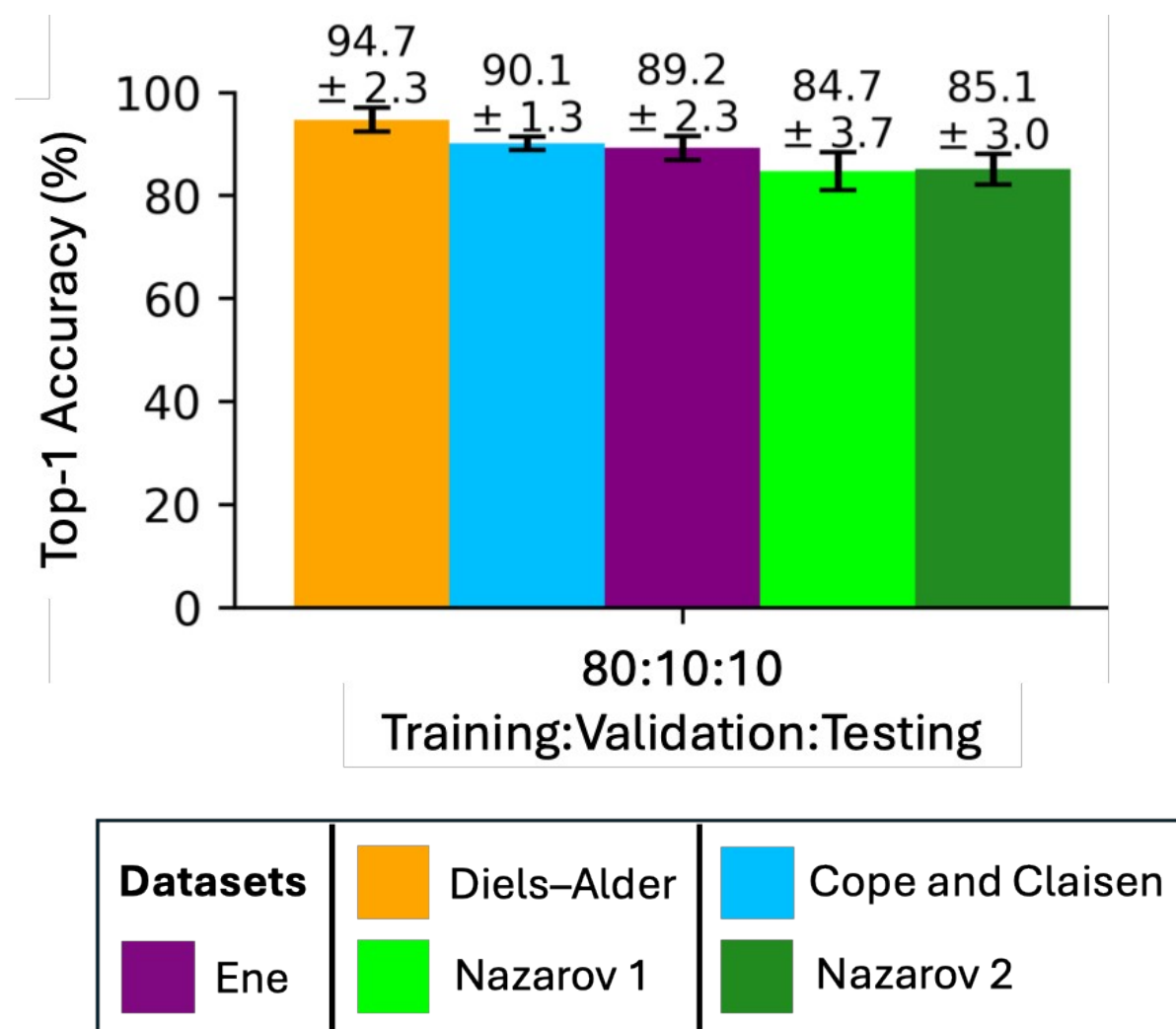


Figure S2. Top-1 product prediction performance for experimental datasets of Diels–Alder, Cope and Claisen, Ene, Nazarov (Nazarov 1), and protonated Nazarov (Nazarov 2) dataset. The NERF model is used and a 80:10:10 training:validation:testing data split is used without any pretraining.

The performance increases from pretraining (as seen in Figure 2 in the manuscript) are shown in tables S1 and S2 with standard deviations. In all cases, the standard deviations were low, indicating the models were robust.

Table S1. Comparison of pretraining approaches for predicting Cope and Claisen rearrangements with NERF model. Results are shown for increasing amounts of Cope and Claisen training data. Top-1 Accuracies, the average from 10 runs, are reported alongside the standard deviation.

Entry	Pretraining Datasets	Cope and Claisen Training:Validation:Testing Splits				
		10:45:45	40:30:30	60:20:20	80:10:10	85:05:10
Pretraining						
1	Diels–Alder 1	76 ± 1.4	87.2 ± 0.6	89.6 ± 1.6	91.9 ± 2.2	92.3 ± 1.6
2	Diels–Alder 2	73.2 ± 1.3	86.1 ± 1.1	89.1 ± 1.9	92 ± 1.1	91.5 ± 1.4
3	Ene	66.7 ± 1.5	83.5 ± 1.5	86.8 ± 2.3	90 ± 1.1	90.3 ± 2
4	Nazarov 1	65 ± 1.4	83.2 ± 1.3	86.4 ± 1.8	90.2 ± 1.4	89.8 ± 1.2
5	Nazarov 2	64.1 ± 1.4	82.6 ± 1.6	86 ± 2.3	89.5 ± 1.6	89.9 ± 1.8
6	USPTO-MIT	68.9 ± 1.4	85.5 ± 1	88.7 ± 1.7	91 ± 1.5	91.5 ± 1.4
7	No Pretraining	62.7 ± 1.8	82.4 ± 1.1	86.6 ± 2.1	90.1 ± 1.3	90.7 ± 0.9

Table S2. Comparison of pretraining approaches for predicting Diels–Alder reactions with NERF model. Results are shown for increasing amounts of Diels–Alder training data. Top-1 Accuracies, the average from 10 runs, are reported alongside the standard deviation.

Entry	Pretraining Datasets	Diels–Alder Training:Validation:Testing Splits				
		10:45:45	40:30:30	60:20:20	80:10:10	85:05:10
Pretraining						
1	Cope and Claisen	78.5 ± 1	91.8 ± 0.8	94.1 ± 0.7	95.5 ± 0.8	95.8 ± 0.6
2	Ene	73.6 ± 0.8	91.3 ± 0.7	93.8 ± 0.8	95.6 ± 0.8	95.5 ± 1.3
3	Nazarov 1	72.4 ± 1.9	91.8 ± 0.7	94.1 ± 1	95.5 ± 0.8	95.9 ± 0.6
4	Nazarov 2	69.6 ± 1.3	91.9 ± 0.6	94.1 ± 0.7	95.2 ± 1.8	96.2 ± 0.7
5	USPTO-MIT	82.9 ± 0.8	94.3 ± 0.5	95.6 ± 0.5	96.5 ± 0.7	96.8 ± 0.8
6	No Pretraining	69.3 ± 2	91.3 ± 1.8	94 ± 1	94.7 ± 2.3	96.1 ± 0.6

When performing the equal size dataset comparison, there was variation in both the pretraining model data split as well as training model data split. As such, the results here are shown per training split. For each training split, the accuracy and standard deviation arise from the 10 pretrained models (and subsequently these become 10 transfer learned models). This is shown below when predicting Cope and Claisen rearrangements (Table S3) and Diels–Alder reactions (Table S4).

Table S3. Comparison of pretraining approaches for predicting Cope and Claisen rearrangements with NERF model when using equal sized pretraining datasets of 1,000 reactions. For each training seed and pretraining combination, 10 pretraining data splits were used to create 10 pretrained models. These models were subsequently trained on the specific training data split (seed) shown. Top-1 Accuracies, the average from these 10 runs, are reported alongside the standard deviation.

Entry	Pretraining Datasets	Cope and Claisen 10:45:45 Splits				
		Seed 1	Seed 2	Seed 3	Seed 4	Seed 5
1	No Pretraining	65.4	63.9	62.7	64.5	60.7
2	Diels–Alder	67.6 ± 1.6	67.0 ± 1.4	66.5 ± 1.0	66.0 ± 2.2	67.2 ± 1.2
3	Ene	64.7 ± 1.2	65.1 ± 1.8	64.8 ± 1.2	64.5 ± 1.8	64.1 ± 1.6
4	Nazarov 1	64.4 ± 1.5	63.5 ± 1.7	62.6 ± 1.6	62.7 ± 1.1	64.4 ± 1.5
5	Nazarov 2	64.9 ± 1.4	64.5 ± 1.3	63.6 ± 1.8	63.0 ± 1.8	63.6 ± 2.1
6	USPTO-MIT	58.6 ± 2.1	61.0 ± 1.3	61.6 ± 1.6	60.2 ± 2.0	61.4 ± 1.6
7	Artificial Cope and Claisen	63.4 ± 1.3	65.0 ± 1.4	63.8 ± 1.7	63.6 ± 1.8	64.1 ± 1.4
More Seeds						
		Seed 6	Seed 7	Seed 8	Seed 9	Seed 10
8	No Pretraining	59.4	61.7	62.1	61.9	64.7

9	Diels–Alder	65.3 ± 1.4	67.2 ± 1.4	66.7 ± 1.9	65.7 ± 1.6	64.9 ± 1.6
10	Ene	63.8 ± 1.6	65.4 ± 1.7	64.5 ± 1.5	64.2 ± 1.9	64.7 ± 2.1
11	Nazarov 1	63.1 ± 2.0	62.9 ± 1.9	63.4 ± 1.9	63.8 ± 1.6	63.3 ± 1.6
12	Nazarov 2	62.9 ± 1.5	63.2 ± 1.4	64.9 ± 1.3	63.8 ± 1.4	63.8 ± 1.8
13	USPTO-MIT	60.2 ± 1.4	59.9 ± 2.1	60.3 ± 1.0	59.5 ± 1.8	60.2 ± 2.0
14	Artificial Cope and Claisen	63.9 ± 2.1	63.9 ± 1.5	63.7 ± 1.6	64.3 ± 1.5	63.3 ± 1.7

Table S4. Comparison of pretraining approaches for predicting Diels–Alder reactions with NERF model when using equal sized pretraining datasets of 1,000 reactions. For each training seed and pretraining combination, 10 pretraining data splits were used to create 10 pretrained models. These models were subsequently trained on the specific training data split (seed) shown. Top-1 Accuracies, the average from these 10 runs, are reported alongside the standard deviation.

Entry	Pretraining Datasets	Diels–Alder 3:48:48 Splits				
		Seed 1	Seed 2	Seed 3	Seed 4	Seed 5
1	No Pretraining	40.3	40.9	43.6	40.8	39.7
2	Cope and Claisen	45.9 ± 2.0	52.4 ± 1.8	53.0 ± 1.4	55.1 ± 2.4	50.7 ± 3.2
3	Ene	45.3 ± 1.3	43.2 ± 0.7	44.8 ± 1.6	45.1 ± 0.8	44.3 ± 1.4
4	Nazarov 1	36.9 ± 2.2	39.8 ± 1.4	38.6 ± 1.7	40.2 ± 3.0	41.6 ± 2.0
5	Nazarov 2	38.5 ± 1.8	39.5 ± 1.6	39.3 ± 1.8	39.0 ± 2.7	39.0 ± 1.5
6	USPTO-MIT	24.3 ± 1.8	28.2 ± 1.6	32.9 ± 1.5	29.1 ± 1.3	29.0 ± 1.7
		More Seeds				
		Seed 6	Seed 7	Seed 8	Seed 9	Seed 10
7	No Pretraining	38.2	36.4	36.8	41.0	36.9
8	Cope and Claisen	48.4 ± 2.5	54.1 ± 1.4	55.5 ± 1.6	56.0 ± 1.6	54.1 ± 5.0
9	Ene	43.0 ± 2.0	46.0 ± 1.2	46.7 ± 1.2	47.0 ± 1.5	42.7 ± 1.3
10	Nazarov 1	39.6 ± 1.7	37.3 ± 2.6	42.7 ± 1.7	40.3 ± 2.0	39.5 ± 2.9
11	Nazarov 2	40.0 ± 1.7	38.2 ± 2.4	42.9 ± 1.2	38.8 ± 1.7	41.2 ± 0.9
12	USPTO-MIT	29.4 ± 1.4	29.5 ± 1.4	29.7 ± 1.9	27.7 ± 1.5	29.5 ± 1.5

Chemformer

Chemformer⁶ was pretrained on two different splits of the Diels–Alder dataset and the USPTO-MIT dataset before being retrained for Cope and Claisen rearrangements. The two Diels–Alder splits were the same as before, where the training:validation:testing set ratios were 80:10:10 (Diels–Alder 1) and 40:30:30 (Diels–Alder 2). The larger USPTO-MIT dataset proved a considerably better source of pretraining at all amounts of Cope and Claisen training data (Table S5). The USPTO-MIT pretrained model was then further pretrained on several pericyclic datasets and fine-tuned for Cope and Claisen rearrangements. As before, a non-protonated Nazarov (Nazarov 1) and protonated Nazarov (Nazarov 2) was used. At low training splits, this additional pretraining data was useful over the non-pretrained baseline but this became less significant with increasing amounts of training data. The smaller the amount of additional pretraining data, the less significant the impact.

Table S5. Comparison of pretraining approaches for predicting Cope and Claisen rearrangements with Chemformer model. Results are shown for increasing amounts of Cope and Claisen training data. Initial models were pretrained on two Diels–Alder datasets and the USPTO-MIT dataset. Further models took the USPTO-MIT pretrained model, retrained on another pericyclic dataset, and were fine-tuned and tested on Cope and Claisen data. Top-1 Accuracies are reported.

Entry	Pretraining Datasets	Cope and Claisen				
		Training	Validation	Testing	Splits	
		10:45:45	40:30:30	60:20:20	80:10:10	85:05:10
Pretraining						
1	Diels–Alder 1	24.1 ± 1.5	54.4 ± 1.3	64.7 ± 0.8	71.5 ± 1.9	71.7 ± 2.2

2	Diels–Alder 2	5.1 ± 0.4	20.3 ± 1.2	30.6 ± 1.7	41.9 ± 2.7	41.8 ± 3.3
3	USPTO-MIT Pretraining	76.7 ± 1.3	91.7 ± 0.8	93.8 ± 0.8	94.6 ± 1.1	94.2 ± 1.3
USPTO-MIT Pretraining + Additional Pretraining						
4	USPTO-MIT + Diels–Alder 1	81.5 ± 1	92.4 ± 0.7	94.2 ± 0.9	94.8 ± 0.8	94.6 ± 0.9
5	USPTO-MIT + Diels–Alder 2	81.7 ± 1.1	92.2 ± 0.6	94.1 ± 0.5	94.7 ± 1.4	95 ± 0.7
6	USPTO-MIT +Ene	78.4 ± 1.4	91.9 ± 0.9	94 ± 0.9	94.4 ± 1.2	94.4 ± 1.3
7	USPTO-MIT + Nazarov 1	77.1 ± 1.3	91.3 ± 0.7	93.5 ± 0.9	94.3 ± 0.9	94.5 ± 1.2
8	USPTO-MIT + Nazarov 2	76.7 ± 1.7	91.6 ± 0.9	93.8 ± 0.9	94.3 ± 1.4	94.4 ± 1.1

GitHubs

Jupyter notebooks to reproduce the pericyclic datasets, assuming one has a valid Reaxys license, can be found on the following GitHubs:

Cope and Claisen rearrangements, Ene reactions, and Nazarov cycloadditions:

<https://github.com/angusketo/PericyclicTL/>

Diels–Alder reactions (previous work²): https://github.com/angusketo/DA_DataExtraction

Details about the Conda environment and Python package versions used to run NERF can be found on the PericyclicTL GitHub. Additionally, a Jupyter notebook is included to more easily extract the results from the pickle and output files that NERF produces:

<https://github.com/angusketo/PericyclicTL/>

The NERF model itself can be found as a supplementary .zip file in the associated conference proceedings: <https://proceedings.mlr.press/v139/bi21a.html>

These Jupyter notebooks, Conda environment information, and datasets (artificial data and Reaxys IDs), can also be found on Zenodo: [10.5281/zenodo.15056997](https://zenodo.org/record/15056997)

Artificial Dataset SMILES

The artificial Cope and Claisen rearrangements are displayed with their atom-mapping included.

```
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Cope and Claisen:

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