Open Challenges in Developing Generalizable Large Scale Machine Learning Models for Catalyst Discovery

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Abstract

The development of machine learned potentials for catalyst discovery has predominantly been focused on very specific chemistries and material compositions. While effective in interpolating between available materials, these approaches struggle to generalize across chemical space. The recent curation of large-scale catalyst datasets has offered the opportunity to build a universal machine learning potential, spanning chemical and composition space. If accomplished, said potential could accelerate the catalyst discovery process across a variety of applications (CO\textsubscript{2} reduction, NH\textsubscript{3} production, etc.) without additional specialized training efforts that are currently required. The release of the Open Catalyst 2020 Dataset (OC20)\textsuperscript{1} has begun just that, pushing the heterogeneous catalysis and machine learning communities towards building more accurate and robust models. In this perspective, we discuss some of the challenges and findings of recent developments on OC20. We examine the performance of current models across different materials and adsorbates to identify notably underperforming subsets. We then discuss some of the modeling efforts surrounding energy-conservation, approaches to finding and evaluating the local minima, and augmentation of off-equilibrium data. To complement the community’s ongoing developments, we end with an outlook to some of the important challenges that have yet to be thoroughly explored for large-scale catalyst discovery.

Introduction

Catalysts have played a key role in the synthesis of everyday chemicals and fuels necessary for a 21st century society. As renewable energy prices continue to decrease, traditional chemical synthesis processes are being revisited for more sustainable alternatives. At the center of this, catalyst discovery plays a key role in the advancement of renewable energy processes and sustainable chemical production, i.e. ammonia for fertilizer and hydrogen production. Unfortunately, the search space for catalyst materials is enormous for even high-throughput experiments.\textsuperscript{2} This presents a need for computational tools to simulate systems through quantum mechanical (QM) models like Density Functional Theory (DFT). QM approaches have made notable advancements in bridging computational results to experimental findings.\textsuperscript{3,4} While effective, QM tools scale very poorly, $O(N^3)$ or
Figure 1: Summary of challenges associated with training on large dataset with large ML potentials discussed in the paper. *Top left* Trade offs in direct and gradient GNN force predictions. *Top right* An example system for a case where the distance metrics are relatively good for the direct approach but the force metrics are worse. *Bottom left* Demonstration of inconsistent error across a metallic surface and a non-metal through an example. *Bottom right* Augmenting existing relaxation datasets with off-equilibrium data can aid in relaxation performance.

worse in the number of electrons. The computational cost associated with QM tools render them infeasible to the scale of the systems and search space desired for catalyst discovery. As a result, the catalysis community has moved towards a more data driven approach. With the QM data available, researchers are often interested in building machine learning surrogates for a particular chemical property. Such efforts, however, were limited to the finite data available, often for a very specific chemistry or system, limiting the generalizability ability of such models. Fortunately, as the community continues to curate larger, and more diverse datasets, machine learning models will continue to improve as they move towards larger, and more sophisticated architectures.

In the field of small molecules, a vast collection of datasets have been developed for varying use cases, including molecular dynamics simulations (MD17, ANI-1, COL1) and quantum mechanical properties (QM9, Alchemy). These datasets are often limited to a few (5-10) unique elements, on average 10-20 atoms per system, and training set sizes in the range of 10k-1M samples. In the field of heterogeneous catalysis, datasets are often much more limited with training set sizes between 100 - 50k. These datasets were often created for very specific applications involving a handful of small adsorbates (i.e. hydrogen containing adsorbates on transition metal surfaces, CO₂ re-
duction catalysts, etc.). The release of OC20 marks a push towards a large, sparse collection of the material space. OC20 spans 55 unique elements, 82 adsorbates and includes a collection of unary, binary and ternary materials. A total of 1.28 million DFT relaxations were performed, comprising \( \sim 260M \) single point evaluations of system energy and per-atom forces.

OC20 presented several practical tasks for the community to work towards. The most general of the tasks, Structure to Energy and Forces (S2EF) evaluates a model’s ability to serve as a surrogate to DFT - predicting a configuration’s energy and per-atom forces. Initial Structure to Relaxed Energy (IS2RE) asks to predict the relaxed state energy, given only the initial structure. Initial Structure to Relaxed Structure (IS2RS) explores how well the relaxed structure can be predicted given only the initial configuration. In the scope of OC20, all energies were referenced to represent adsorption energy. For more details, we refer readers to the original manuscript.

In this perspective we shed light on the challenges of training Graph Neural Networks (GNNs) on large-scale datasets spanning material and composition space, illustrated in Figure 1. We begin with a quick overview on the current state of the community’s progress and share some takeaways from what we have observed. We then discuss some telling trends on the performance of models across different adsorbates and material types. We discuss how different approaches and modeling decisions impact the prediction tasks and highlight the challenges associated with each. Further, we explain what the accuracies in various proposed metrics mean and some of the challenges in analyzing them. Finally, we share our outlook on the direction the community is headed and what still remains to achieve a large scale, generalizable potential for catalyst discovery.

**Community progress in developing ML models for catalysis**

Molecular modeling has progressed at an incredible rate over the past few decades. Simple linear models, neural networks, and kernel methods were originally developed relying on hand-crafted atomic representations, or descriptors as inputs to the models. Descriptors capture invariant geometric information in the form of bonds and angles of the local environment of an atom. While effective, the parameterization of such descriptors has been a challenging and non-trivial task. The past few years has seen a shift towards deep learning approaches. Rather than relying on hand crafted representations, models are being developed to learn similar or more expressive representations, specifically by exploiting the graphical nature of molecules using Graph Neural Networks (GNNs). Such models only take in 3D atomic coordinates and atomic numbers. A graph is then generated, where atoms are treated as nodes, and the distance between them as edges. Once a graph has been constructed, GNNs will undergo several rounds of message passing in which node representations are updated based off messages sent between neighboring nodes. While models may differ in their exact architecture, the update and message functions often include a series of multi-layer perceptrons and nonlinearities. Unlike traditional descriptor based models, GNNs end up learning node representations as part of the training process. Learned representations proceed through a final output block where a final prediction is made. In recent years, GNNs have come to surpass traditional descriptor based models. While typically data hungry, recent models like NequIP are demonstrating great performance with as little as 100 samples. GNNs continue to gain traction as models continue to demonstrate state of the art performance on molecular datasets.

Since the release of OC20, the community has been rapidly developing new approaches to improve existing baselines. Models being de-
Figure 2: Community progress on the OC20 dataset since release. Left: IS2RE performance for both direct and relaxation based approaches. The current error target of 0.10 eV would make these models more practically useful for researchers’ applications. Right: S2EF performance as evaluated by mean absolute error of the forces. IS2RE and S2EF MAEs for their median baselines are 1.756 eV and 0.084 eV/Å, respectively.

Where are molecular GNNs still erroneous?

Most of the independent work done in developing ML potentials has been confined to datasets built for certain applications. For example, ML potentials for the applications of CO2RR are usually just trained with CO and H adsorbates.46 While this approach might interpolate well across materials, extrapolation to different adsorbates or more complicated materials will likely suffer in performance. A universal ML potential, if possible, would first require a large, diverse dataset that spans material and chemical space. OC20 dataset was created to build ML potentials that cover a large and diverse space of heterogeneous catalysts.

Errors across material types: With over 300k unique surfaces, OC20 spans a vast range of material compositions. When training large GNNs on the entire OC20 dataset, we observe that the accuracies are not uniform across element and adsorbate types. To analyze this, we divide the validation set into four different material types: intermetallics, metalloids, nonmetals and halides, Figure 3(a). The distribution of data across these classes of materials is not the same, we have significantly more intermetallics and relatively fewer halides. We observe that the performance on non-metals is significantly worse, although both nonmetals and...
metaloids contribute to similar percentage of training data (Figure 3(b)). On the other hand, models tend to do much better across the board for intermetallics. Inaccuracies coming from non-metals disproportionately contribute to the overall errors, leading to worse performance for both force and energy predictions.

Errors across adsorbates: Large adsorbates are inherently more complicated as the degrees of freedom increases with the number of atoms. However, we observe no correlation with our model’s performance and the size of the adsorbate. Model accuracies are poor for bidentate adsorbates like *CH*COH, *N*NO, *CH2*O, shown in Figure 3(c). Figure 3(d) also shows that adsorbates with N and O are generally more erroneous.

### Modeling trade-offs

#### Energy-conserving forces

Force predictions play an important role in the applications of ML models for catalyst discovery. While some tasks may only be interested in property predictions like adsorption or formation energy, \[ \text{48,52,53} \] forces are necessary to study dynamics such as structural relaxations, molecular dynamics, and transition state calculations. \[ \text{113,36-54} \]

Physically, energy-conserving forces are derived as the gradient of energy with respect to atomic positions:

\[
F_i = -\frac{dE}{dx_i}
\]  

(1)

Energy-conservation is critical in studying molecular dynamics accurately. ML models estimating energy-conserving forces must ensure the architecture is continuous and differentiable, often satisfied by appropriate non-linear
Table 1: Results on the OC20 S2EF task via gradient-derived or direct force predictions. All models were trained on the OC20 S2EF All dataset. Results reported for the validation set. Energy metrics are unavailable for the gradient based SpinConv model due to being optimized only on forces.

| Model    | Energy MAE (eV) ↓ | Force MAE (eV/Å) ↓ |
|----------|-------------------|---------------------|
|          | ID OOD Ads OOD Cat OOD Both | ID OOD Ads OOD Cat OOD Both |
| Median   | 2.04 2.42 1.99 | 0.081 0.080 0.079 |

|          | Gradient forces | Direct forces |
|----------|-----------------|---------------|
| SpinConv | - - - 0.31 | 0.26 - 0.35 | 0.47 0.027 0.030 0.029 0.37 |
| GemNet-dT | 0.36 0.39 0.48 | 0.58 0.030 0.034 0.033 0.42 |
|           | 0.35 0.38 0.34 | 16.06% 12.62% 13.17% 11.06% |

Table 2: Results on the OC20 IS2RE task using one of two approaches. Direct: Directly predicting the relaxed state energy and Relaxation: Training a model for energy and force predictions, followed by an iterative ML-based geometry optimization to arrive at a relaxed structure and energy. Relaxation results on the 2M subset suggest that competitive results are still possible with a limited compute budget. Results reported for the test set.

| Model    | Approach | Dataset Size | Energy MAE [eV] ↓ | Energy within Threshold (EwT) ↑ |
|----------|----------|--------------|-------------------|-------------------------------|
|          |          |              | ID OOD Ads OOD Cat OOD Both | ID OOD Ads OOD Cat OOD Both |
| Median baseline | - - | 1.75 1.88 1.71 | 1.66 | 0.71% 0.72% 0.89% 0.74% |
| DimeNet++ | Direct | 460,328 | 0.56 0.57 0.58 | 0.66 | 4.25% 2.07% 4.10% 2.41% |
| SpinConv | Direct | 460,328 | 0.56 0.72 0.57 | 0.67 | 4.80% 2.26% 3.82% 2.33% |
| NoisyNodes | Direct | 460,328 | 0.42 0.57 0.44 | 0.47 | 9.12% 3.49% 8.01% 4.64% |
| Graphormer | Direct | 460,328 | 0.40 0.57 0.42 | 0.50 | 8.97% 3.45% 8.18% 3.79% |
| DimeNet++ + LF + LE | Relaxation | 2,000,000 | 0.53 0.57 0.56 | 0.52 | 6.79% 4.71% 6.49% 4.54% |
| SpinConv | Relaxation | 2,000,000 | 0.46 0.51 0.44 | 0.44 | 7.38% 4.82% 7.05% 5.31% |
| GemNet-dT | Relaxation | 2,000,000 | 0.44 0.44 0.45 | 0.42 | 9.37% 6.59% 8.42% 6.40% |
| GemNet-OC | Relaxation | 2,000,000 | 0.41 0.42 0.42 | 0.39 | 11.02% 8.68% 10.10% 7.82% |
| DimeNet++ - LF + LE | Relaxation | 133,934,018 | 0.50 0.54 0.58 | 0.61 | 6.57% 4.34% 5.09% 3.93% |
| SpinConv | Relaxation | 133,934,018 | 0.42 0.44 0.46 | 0.42 | 9.37% 7.47% 8.16% 6.50% |
| GemNet-dT | Relaxation | 133,934,018 | 0.39 0.39 0.43 | 0.38 | 12.37% 9.11% 10.09% 7.87% |
| GemNet-OC | Relaxation | 133,934,018 | 0.35 0.35 0.38 | 0.34 | 16.06% 12.62% 13.17% 11.06% |

Geometrically, forces derived in an energy-conserving manner ensures forces are rotationally equivariant, a necessary physical relation of molecular systems. Unfortunately, a gradient calculation increases model overhead in both memory usage and computational time by a factor of 2-4. For datasets like MD17, calculating forces as a gradient is known to help in model accuracies as that is an important physical prior to the model. Models trained on MD17 are often used to run molecular dynamics, further necessitating the need for energy-conservation. However, for the OC20 dataset, particularly in the task of geometric optimization, we observe that the gradient approach for calculating forces to perform worse than direct prediction of forces for GemNet-dT and Spinconv. DimeNet++ and ForceNet were built for gradient and direct approach respectively. The gradient approach could also make the training unstable in certain cases, which has been observed for ForceNet and GemNet-OC. Table 1 compares performance on the S2EF task for two recent top performing models, GemNet-dT and SpinConv. Not only are the force accuracies worse for the gradient approach, but the corresponding relaxed structure and relaxed energy
metrics calculated via optimization are also significantly worse.\textsuperscript{[20]}

While energy-conservation plays a critical role in many molecular applications, we observe that direct force computations bring efficiency and performance advantages.\textsuperscript{[40,56]}\textsuperscript{[10,56]} Models trained for direct force predictions are limited to applications where strict enforcement of energy-conservation can reasonably be ignored, i.e. OC20's structural relaxations. Here, atomic positions are updated solely from force estimates.\textsuperscript{[4,10,56]} If necessary, DFT, or a subsequent ML model, can then be used to make reliable energy predictions on the ML optimized structure. Similarly, transition states or saddle points can be derived in a similar manner with direct-force models. We want to emphasize that although unorthodox, direct-force models still prove to be useful in certain catalyst applications, i.e. OC20-like tasks.

**Prediction of relaxed energy and structure**

Adsorption energy is one of many properties that helps inform catalyst performance.\textsuperscript{[58]} Computationally, this is computed via a series of QM structural relaxations. The relaxed energy is then referenced to represent the adsorption energy, see Chanussot et al.\textsuperscript{[1]} García-Muelas et al.\textsuperscript{[59]} for more details. From a data-driven approach, we can predict the relaxed energy or the relaxed structure of an atomic system usually via two methods. First, we can build a surrogate to DFT, approximating system energy and per-atom structures, and running ML optimizations to find the minimum energy, a common approach within the field. Alternatively, given a large enough dataset of relaxed structures and energies, we can try to predict these properties directly using a ML model instead of optimizing via an iterative loop. The advantage of the direct method over the relaxation approach is that it requires only a single call to the ML model, whereas the relaxation approach could require on average 200-300 calls for a single relaxation. Direct approaches are particularly advantageous when we talk about the computational cost of approaching large scale inference on the order of hundreds of millions to billions of systems.

The community has made tremendous progress in predicting adsorption energy as evaluated by the OC20 IS2RE task (Figure [2]). Direct approaches, despite using 300x less data, are approaching the competitive relaxation based approaches of GemNet-XL and GemNet-OC. Inference time aside, models trained on the full 133M dataset for the relaxation based approaches are typically compute intensive, using between 128-512 GPUs.\textsuperscript{[1,35,40,56]} While this is certainly a small price to pay if the models developed accelerate the discovery process, it does make it difficult for the community to engage in and aid in development. This has been particularly observed in the NeurIPS 2021 Open Catalyst Challenge,\textsuperscript{[44]} where of the 30 submissions, 0 were made via the relaxation approach. Here, we show that models trained on a 2M subset of the full dataset are still able to provide competitive results and even, averaged across all splits, out perform direct approaches. Given the trends in the 2M dataset correlate well with the full 133M dataset,\textsuperscript{[45]} this should help incentivize the community to explore other approaches even with resource limitations. Although the relaxation approach is computationally expensive for both training and inference, we have observed that the models trained through this approach tend to generalize better on out-of-distribution (OOD) data, Table [2].

Direct relaxed energy predictions are an easier ML problem than direct structure predictions. For a system of size N, energy predictions require a single scalar output, while structure predictions require 3N components. We find that for relaxed energy prediction tasks, metrics are closer for direct and relaxation approaches whereas for structure prediction task the metrics are worse. The OC20 paper provides a baseline for relaxed structure prediction only via the relaxation approach.\textsuperscript{[1]} In Table [3] we provide baselines for direct relaxed structure prediction. A considerable gap exists between the direct and relaxation based approaches (especially in the DFT based metrics).
Metrics for finding local minima

Relaxed structure prediction is less straightforward than some of the other common energy and force prediction tasks. Given a dataset like OC20 where relaxed structures are not necessarily global minima, a model trained on such a dataset could either (1) predict and arrive at the same local minima, (2) arrive at a different, but still suitable minima, or (3) fail to arrive at any sort of minima.

To account for this, two main metrics have been presented in the OC20 paper. Average Distance within Threshold (ADwT) is a distance based metric and measures how close the predicted structure compares to the actual structure. This is similar to the Global Distance Test (GDT) metric in the protein folding task. ADwT takes an average across different thresholds varying from 0.1 to 0.5 Å to ensure a signal is captured. For the OC20 dataset, we evaluate this metric for the input initial structures for an accuracy of 21.18% on the in-domain validation set.

Models, at the bare minimum, should perform better than this baseline. To ensure invariance to arbitrary coordinate reference frames, we predict the difference between initial and final positions instead of the final position Cartesian coordinates. Predicting the delta difference helps simplify this task and results in improved ADwT accuracies.

Table 3: Baseline metrics for IS2RS direct task in comparison with the relaxation approach. Metrics are reported on a 2k subset of the validation set, across all splits. DwT is evaluated at a threshold of 0.04 Å. For compute reasons, DFT-based metrics were evaluated on a 200 system subset of the 2k, 50 systems from each split.

| Model           | DwT (at 0.04 Å) | ADwT | FbT | AFbT |
|-----------------|-----------------|------|-----|------|
| ForceNet        | 0.70            | 45.69% | 0.00% | 0.00% |
| Direct          | 1.05            | 47.76% | 0.00% | 0.00% |
| GemNet-dT       | 1.73            | 45.87% | 0.00% | 0.08% |
| ForceNet        | 1.45            | 46.51% | 0.00% | 7.64% |
| Relaxation      | 8.20            | 55.81% | 0.00% | 12.55% |
| GemNet-dT       | 13.95           | 60.88% | 0.00% | 0.35% |

A model that predicts a relaxed structure that is not identical to its DFT reference may still be considered successful for two reasons. (1) the model could have predicted a symmetrically identical site on the surface and (2) the model predicted a different, but still suitable local minima. The former is more a concern surrounding the distance-based metric, as ADwT, although accounts for periodic-boundary conditions, does not consider symmetrically identical sites. While it is rather unlikely an adsorbate initialized over a particular site will hop several sites over to a symmetrically identical site, it is worth raising awareness to the possibility. On the other hand, a model that arrives at a different relaxed structure entirely will fail according to ADwT. However, to verify whether the model has predicted a different suitable minima, we can evaluate the DFT forces corresponding to the ML predicted structures. This metric is called Average Force below Threshold (AFbT) and it measures the percent of structures having their forces close to zero. Since models are expected to predict relaxed structures, DFT forces should be close to zero. This is a stricter metric as compared to ADwT. However, this is far more expensive due to the additional DFT calculations. A more practically useful metric would be number of DFT calculations required to find the relaxed structure starting from the ML relaxed structure. This would give us an idea of the percent of DFT calculations that the current ML models can reduce. Although useful, this is a significantly more expensive metric than AFbT calculations. While it is not something Open Catalyst Project’s (OCP) tracks on their public leaderboard, we bring awareness to it as there could be instances where models do poorly on ADwT and AFbT but resulting structures are only a few DFT steps away from the relaxed structure.

In Table 3 we compare relaxed structure prediction via a direct and relaxation approach. We observe that direct methods, although having competitive ADwT metrics, have AFbT metrics that are significantly worse. This suggests that direct models do a reasonable job at getting close to the relaxed structure but are in high-force configurations, failing to capture repulsive physical interactions. We speculate models struggle with this since small perturbations distances can have large consequences on
forces, e.g. moving two atoms at an equilibrium bond length fractions of an angstrom towards each other. Relaxed structure prediction via the relaxation approach avoids this issue by using ML forces to drive a geometric optimizer.

We observe that distance metrics at tighter thresholds correlate better with force based metrics, however, going below 0.04 Å does not give sufficient signal and the accuracies for most systems fall to zero. Moreover, the Distance within Threshold (DwT) at 0.04 Å isn’t a good enough signal that can replace AFbT. For example, DwT (at 0.04Å ) for ForceNet relaxation approach and GemNet-dT direct approach are similar, however, the AFbT metrics still differ by 7.56% (as shown in Table 3). We believe that finding non DFT-based metrics that correlate well with DFT-based metrics is still an open and important question in the community which would make model evaluation computationally less expensive.

### Additional data

The OC20 paper released two additional data subsets generated with ab-initio molecular dynamics (‘MD’) and structural perturbations (‘Rattled’). These provide 38M and 17M additional S2EF training data points respectively.

Table 4 presents results for GemNet-OC models trained on S2EF, Rattled, and MD data compared against similar analysis from the OC20 paper for DimeNet++. First, on the force MAE metric, addition of MD data hurts DimeNet++ while it improves GemNet-OC. We speculate this to be another artifact of modeling forces as negative gradients of energy (as in DimeNet++) vs. direct prediction (as in GemNet-OC). Second, consistent with the OC20 paper, adding MD data to the training set provides a useful signal for IS2RS structure relaxations as per the AFbT metric. Finally, adding Rattled data helps with IS2RS metrics, but did not help or marginally hurt the S2EF force MAE. This could be due to a variety of reasons – random perturbations being too large / small to be useful, intermediate structures along a trajectory being less useful compared to closer to the local minimum (as in MD initial structures), etc. A promising direction here could be active learning approaches to optimally query additional training data points.

### Summary and Outlook

The development of generalizable or universal ML models has only recently been seriously considered with the emergence of large-scale datasets like OC20. Since its release, the catalysis and ML communities have both made tremendous progressive in developing models for catalyst applications. As the community continues to grow and as more datasets emerge that span material and composition space, the prospect of large-scale generalizable models is within reason. Progress thus far has demonstrated several challenges in accomplishing this feat: classes of materials and adsorbates with inconsistent errors, energy-conserving forces, relaxed vs direct approaches, DFT metrics, and data augmentation strategies. In this perspective, we discussed these challenges in detail and provided some insights as to how and why they are important. Although these challenges were discussed in the context of OC20, we anticipate similar challenges to future datasets of its kind.

Datasets like OC20 has offered new ways to how we think about building large, generalizable, and reliable models. While model performance has been the focal point of community progress thus far, we provide an outlook of other important challenges that we hope the community to engage in.
Training strategies. OC20 was released with predefined training, validation, and test sets. Its splits were curated in a manner to tackle the problem of building a single generalizable model for catalysis. However, it could be the case that multiple models for different subsets of the data, e.g. adsorbates, compositions, materials, do better. In the case of nonmetals, for instance, we have shown that this actually hurts performance - a possible consequence of the reduced dataset size.

Uncertainty and active learning. While model performance is a necessary step for the discovery process, it is not always sufficient. A practical ML-aided catalyst discovery pipeline will ultimately turn to experiments to validate whether the ML predicted “great” catalyst is at all effective. Having confidence in these predictions is particularly important to avoid wasted expensive experiments. Uncertainty quantification has been a particularly popular topic within the catalysis community, often focused on the small data regime and active learning. The effectiveness of traditional uncertainty estimation techniques on large datasets like OC20 is a necessary and important step for the future of this work. Similarly, how to best leverage active learning for either dataset generation and/or augmentation or online active learning at the scale of OC20 will be an exciting future direction.

Model efficiency. In addition to model performance and reliability, model efficiency will continue to be critical for all applications. For training, faster, more data efficient models can help attract the community to tackle some of the bigger challenges like a surrogate to DFT, i.e. OC20’s S2EF task. Progress so far has shown that the best models are also the largest models. From an inference perspective, this poses obvious challenges of slower speeds and ultimately reduced screening throughput. While models still remain orders of magnitude faster than DFT, when considering the possibility of screening billions of systems, computational costs add up. Recent models encoding equivariant representations have shown incredible scaling and efficiency gains that could be promising to explore. Moving forward, efficient architectures and model distillation will be an important contribution to reduce the computational cost of large-scale inference, even if it means sacrificing some accuracy.

Data augmentation. The scale of OC20 makes data augmentation a non-trivial challenge. With 130M+ training data points, randomly adding 10-100k data points will likely have negligible impact on the models. We observed that models using the additional MD data are able to perform the best, while the rattled data has little impact. Identifying strategies to combine and train large molecular and material datasets like ANL and OQMD with OC20 could help improve models even further. The biggest challenge surrounding this comes from combining datasets of varying levels of DFT theory.

Energy-conserving forces. In the context of OC20, we have observed that the best performing models make a direct force-prediction. While this may be suitable for some applications, the more physically motivated gradient approach to force prediction is desired for other applications like MD. The same direct models applied to MD17 observe the opposite effect, better performance via the gradient method. It remains an open question why this is the case, and we encourage others to investigate this observation.

Physics-based modeling. The majority of models submitted to OC20 have followed a purely data-driven approach, only taking in atomic numbers and positions as inputs. Exploring ways to leverage OC20 charge density or Bader charge data could prove useful, particularly in the low data regime. Additionally, models like UNiTE or OrbNet that leverage tight binding DFT for featurization could be interesting to explore for catalyst applications.

To be made publically available at https://github.com/Open-Catalyst-Project/ocp/blob/main/DATASET.md
Glossary

ADwT  Average Distance within Threshold. 8

AFbT  Average Force below Threshold. 8, 9

DFT   Density Functional Theory. 1, 8

DwT   Distance within Threshold. 9

EwT   Energy within Threshold. 6

GNNs  Graph Neural Networks. 3, 4

IS2RE Initial Structure to Relaxed Energy. 3, 4, 6

IS2RS Initial Structure to Relaxed Structure. 3, 9

OC20  Open Catalyst 2020 Dataset. 1, 3–10

OCP   Open Catalyst Project’s. 8

S2EF  Structure to Energy and Forces. 3, 4, 9
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