Single-Atom Catalysis Data Analysis

Data Description

The single-atom catalysis data is stored in `data/single_atom_catalysis.RData`, and the raw data is available at this Github repo. Here we model the metal/oxide binding energy (the response variable y) using $p = 59$ physical properties of the transition metals and the oxide supports (the primary features X). The response variable y and the primary features X are treated as continuous variables, and we aim to use **iBART** to find an interpretable model with high predictive performance for the metal/oxide binding energy.

A total of 13 transition metals (Cu, Ag, Au, Ni, Pd, Pt, Co, Rh, Ir, Fe, Ru, Mn, V) and 7 oxide supports (CeO_2(111), MgO(100), CeO_2(110), TbO_2(111), ZnO(100), TiO_2(011), α-Al_2O_3(0001)) were studied in the dataset, making a total of $n = 13 \times 7 = 91$ metal/oxide pairs. The primary feature matrix X contains various physical properties of the transition metals and the oxide supports including Pauling Electronegativity (χ_P), $(n-1)^{\text{th}}$ and n^{th} Ionization Energies (IE_{n-1}, IE_n), Electron Affinity (EA), HOMO Energy, LUMO Energy, Heat of Sublimation (ΔH_{sub}), Oxidation Energy of oxide support ($\Delta H_{\text{f,ox,bulk}}$), Oxide Formation Enthalpy ($\Delta H_{\text{f,ox}}$), Zunger Orbital Radius (r), Atomic Number (Z), Meidema Parameters of metal atoms ($\eta^{1/3}, \varphi$), Valance Electron (N_{val}), Oxygen Vacancy Energy of oxide support ($\Delta\text{E}_{\text{vac}}$), Workfunction of oxide support (WF), Surface Energy (γ), Coordination Number (CN), and Bond Valence of surface metal atom (BV). Most of these physical properties are defined for both the transition metals and the oxide supports while a few of them are only defined for either the transition metals or the oxide supports. A detailed description of the 59 primary features X can be find in pages 11–14 of the data supplementary materials published by O'Connor et al.

Package and Data Loading

Before loading the **iBART** package, we must allocate enough memory for Java to avoid out of memory errors.

```r
# Allocate 10GB of memory for Java. Must be called before library(iBART)
options(java.parameters = "-Xmx10g")
library(iBART)
```

Next, we load the real data set and examine what data are needed to run iBART.

```r
load("../data/single_atom_catalysis.RData")
ls()
#> [1] "head" "unit" "X"    "y"
```

The data set consists of 4 objects:

- `y`: a `numeric` vector of metal/oxide binding energy described in Data Description. This is our response variable.
- `X`: a `matrix` of physical properties of the transition metals and the oxide supports described in Data Description. These are our primary features (predictors).
- `head`: a `character` vector storing the column names of `X`.
- `unit`: a (optional) `list` of named numeric vectors. This stores the unit information of the primary features `X`. This can be generated using the helper function `generate_unit(unit, dimension)`. See `?iBART::generate_unit` for more detail.

iBART

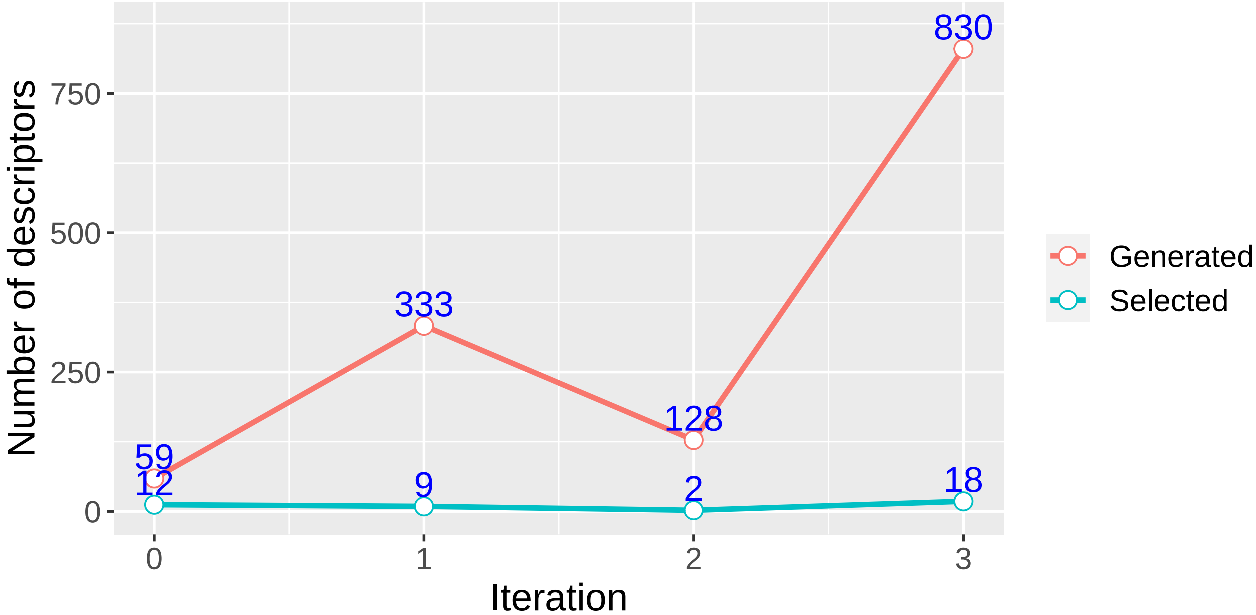
Now let's apply iBART to this data set. Besides the usual regression data (X,y), we need to specify the descriptor generating strategy through `opt`. Here we specify `opt = c("binary", "unary", "binary")`, meaning there will be 3 iterations and we want to alternate between binary and unary operators, starting with binary operators $\mathcal{O}_b$. We can also use all operators O in an iteration. For example, `opt = c("all", "all")` will apply all operators O for 2 iterations.

```
iBART_results <- iBART(X = X, y = y,
                       head = head,  # colnames of X
                       unit = unit,  # units of X
                       opt = c("binary", "unary", "binary"), # binary operator first
                       out_sample = FALSE,
                       Lzero = TRUE,
                       K = 5, # maximum descriptors in l-zero model
                       standardize = FALSE,
                       seed = 888)
#> Start iBART descriptor generation and selection...
#> Iteration 1
#> iBART descriptor selection...
#> avg.........null..................................................
#> Constructing descriptors using binary operators...
#> Iteration 2
#> iBART descriptor selection...
#> avg.........null..................................................
#> Constructing descriptors using unary operators...
#> Iteration 3
#> iBART descriptor selection...
#> avg.........null..................................................
#> Constructing descriptors using binary operators...
#> BART iteration done!
#> LASSO descriptor selection...
#> L-zero regression...
#> Total time: 199.63176202774 secs
```

`iBART()` returns many interesting outputs. For example, `iBART_results$iBART_gen_size` and `iBART_results$iBART_sel_size` store dimension of the newly generated / selected descriptor space for each iteration. Let's plot them and see how **iBART** use nonparametric variable selection for dimension reduction.

```
library(ggplot2)
df_dim <- data.frame(dim = c(iBART_results$iBART_sel_size, iBART_results$iBART_gen_size),
                     iter = rep(0:3, 2),
                     type = rep(c("Selected", "Generated"), each = 4))
ggplot(df_dim, aes(x = iter, y = dim, colour = type, group = type)) +
  theme(text = element_text(size = 15), legend.title = element_blank()) +
  geom_line(size = 1) +
  geom_point(size = 3, shape = 21, fill = "white") +
  geom_text(data = df_dim, aes(label = dim, y = dim + 40, group = type),
            position = position_dodge(0), size = 5, colour = "blue") +
  labs(x = "Iteration", y = "Number of descriptors") +
  scale_x_continuous(breaks = c(0, 1, 2, 3))
#> Warning: Using `size` aesthetic for lines was deprecated in ggplot2 3.4.0.
#> i Please use `linewidth` instead.
#> This warning is displayed once every 8 hours.
```

```
#> Call `lifecycle::last_lifecycle_warnings()` to see where this warning was
#> generated.
```



We can access the selected k-descriptor via `iBART_results$Lzero_names` and the corresponding regression model in `iBART_results$Lzero_models`. For instance, the selected 3-descriptor model is

```
iBART_results$Lzero_names[[3]]
#> [1] "(s_EA*Hf)"                  "abs((Hfo/Oxv))"
#> [3] "abs(((m_n13/m_N_val)/Oxv))"
summary(iBART_results$Lzero_models[[3]])
#>
#> Call:
#> lm(formula = y_train ~ ., data = dat_train)
#>
#> Residuals:
#>      Min       1Q   Median       3Q      Max
#> -1.70871 -0.42326  0.05825  0.44715  1.97315
#>
#> Coefficients:
#>                                 Estimate Std. Error t value Pr(>|t|)
#> (Intercept)                     -0.01707    0.12675  -0.135    0.893
#> `(s_EA*Hf)`                      0.40427    0.04441   9.104 2.75e-14 ***
#> `abs((Hfo/Oxv))`                -0.58838    0.09857  -5.969 5.05e-08 ***
#> `abs(((m_n13/m_N_val)/Oxv))` -19.62963    4.25098  -4.618 1.33e-05 ***
#> ---
#> Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
#>
#> Residual standard error: 0.6378 on 87 degrees of freedom
#> Multiple R-squared:  0.9534, Adjusted R-squared:  0.9518
#> F-statistic: 593.9 on 3 and 87 DF,  p-value: < 2.2e-16
```

OIS vs non-OIS

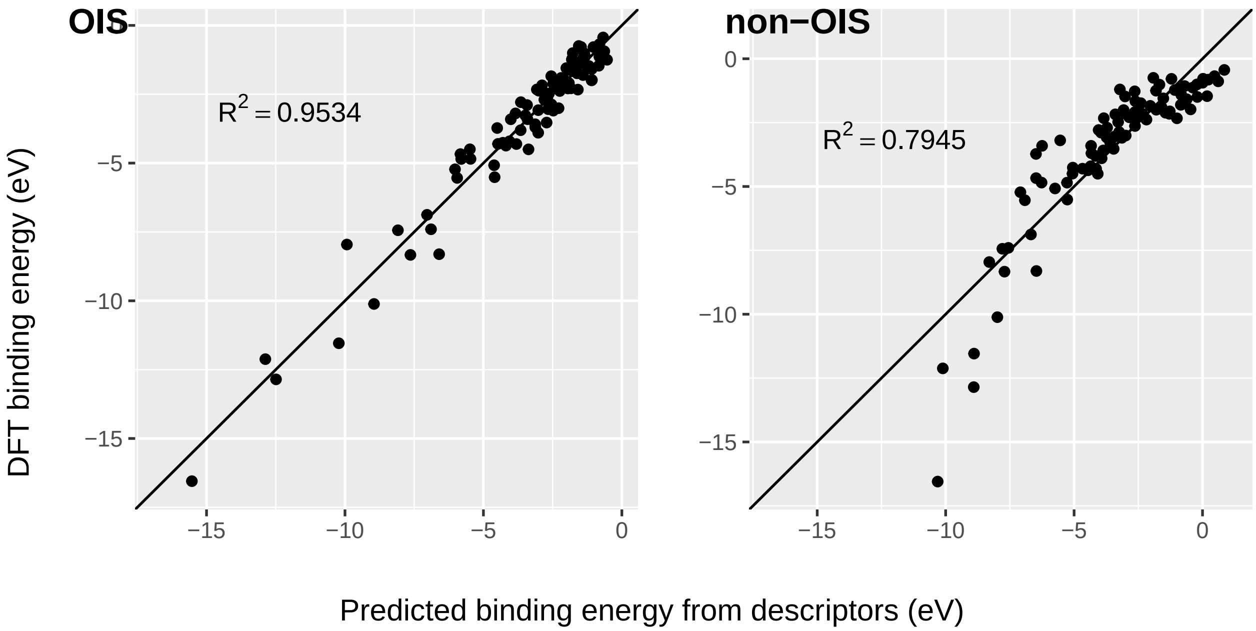
The OIS model differs from the non-OIS model in that the former builds on nonlinear descriptors (composition of $\mathcal{O}$ on X) while the latter builds on the primary features X. The OIS model has many advantages. In particular, it reveals interpretable nonlinear relationship between y and X, and improves prediction accuracy over a simple linear regression model (or non-OIS model). We showcase the improved accuracy over non-OIS model using Figure 7 in the paper.

```
# Train a non-OIS model with 3 predictors
set.seed(123)
model_no_OIS <- k_var_model(X_train = X, y_train = y, k = 3, parallel = FALSE)

#### Figure 7 ####
library(ggpubr)
model_OIS <- iBART_results$Lzero_model[[3]]

# Prepare data for plotting
data_OIS <- data.frame(y = y, y_hat = model_OIS$fitted.values)
data_no_OIS <- data.frame(y = y, y_hat = model_no_OIS$models$fitted.values)

p1 <- ggplot(data_OIS, aes(x = y_hat, y = y)) +
  geom_point() +
  geom_abline() +
  xlim(c(min(data_OIS$y_hat, data_OIS$y) - 0.2, max(data_OIS$y_hat, data_OIS$y) + 0.2)) +
  ylim(c(min(data_OIS$y_hat, data_OIS$y) - 0.2, max(data_OIS$y_hat, data_OIS$y) + 0.2)) +
  xlab("") +
  ylab("") +
  annotate("text", x = -12, y = -3, parse = TRUE,
           label = paste("R^{2} ==", round(summary(model_OIS)$r.squared, 4)))
p2 <- ggplot(data_no_OIS, aes(x = y_hat, y = y)) +
  geom_point() +
  geom_abline() +
  xlim(c(min(data_no_OIS$y_hat, data_no_OIS$y) - 0.2, max(data_no_OIS$y_hat, data_no_OIS$y) + 0.2)) +
  ylim(c(min(data_no_OIS$y_hat, data_no_OIS$y) - 0.2, max(data_no_OIS$y_hat, data_no_OIS$y) + 0.2)) +
  xlab("") +
  ylab("") +
  annotate("text", x = -12, y = -3, parse = TRUE,
           label = paste("R^{2} ==", round(summary(model_no_OIS$models)$r.squared, 4)))
fig <- ggarrange(p1, p2,
                 labels = c("OIS", "non-OIS"),
                 ncol = 2, nrow = 1)
annotate_figure(fig,
                bottom = text_grob("Predicted binding energy from descriptors (eV)"),
                left = text_grob("DFT binding energy (eV)", rot = 90))
```



R Session Info

```
sessionInfo()
#> R version 4.0.5 (2021-03-31)
#> Platform: x86_64-apple-darwin17.0 (64-bit)
#> Running under: macOS Big Sur 10.16
#>
#> Matrix products: default
#> BLAS:   /Library/Frameworks/R.framework/Versions/4.0/Resources/lib/libRblas.dylib
#> LAPACK: /Library/Frameworks/R.framework/Versions/4.0/Resources/lib/libRlapack.dylib
#>
#> locale:
#> [1] en_US.UTF-8/en_US.UTF-8/en_US.UTF-8/C/en_US.UTF-8/en_US.UTF-8
#>
#> attached base packages:
#> [1] stats     graphics  grDevices utils     datasets  methods   base
#>
#> other attached packages:
#> [1] ggpubr_0.6.0  ggplot2_3.4.4 iBART_0.0.3.3
#>
#> loaded via a namespace (and not attached):
#>  [1] shape_1.4.6        tidyselect_1.2.0   xfun_0.40
#>  [4] purrr_1.0.2        splines_4.0.5      rJava_1.0-4
#>  [7] lattice_0.20-44    carData_3.0-5      colorspace_2.1-0
#> [10] vctrs_0.6.4        generics_0.1.3     htmltools_0.5.6.1
#> [13] yaml_2.3.7         utf8_1.2.4         survival_3.2-11
#> [16] rlang_1.1.1        pillar_1.9.0       glue_1.6.2
#> [19] withr_2.5.1        foreach_1.5.1      lifecycle_1.0.3
#> [22] munsell_0.5.0      ggsignif_0.6.4     gtable_0.3.4
#> [25] codetools_0.2-18   evaluate_0.22      labeling_0.4.3
#> [28] knitr_1.44         fastmap_1.1.1      parallel_4.0.5
#> [31] fansi_1.0.5        itertools_0.1-3    broom_1.0.5
#> [34] bartMachine_1.2.6  scales_1.2.1       backports_1.4.1
```

```
#> [37] abind_1.4-5          farver_2.1.1         gridExtra_2.3
#> [40] digest_0.6.33        rstatix_0.7.2        dplyr_1.1.3
#> [43] cowplot_1.1.1        grid_4.0.5           cli_3.6.1
#> [46] tools_4.0.5          magrittr_2.0.3       missForest_1.4
#> [49] glmnet_4.1-1         tibble_3.2.1         randomForest_4.6-14
#> [52] crayon_1.5.2         tidyr_1.3.0          car_3.1-2
#> [55] pkgconfig_2.0.3      Matrix_1.6-1.1       bartMachineJARs_1.1
#> [58] rmarkdown_2.25       rstudioapi_0.15.0    iterators_1.0.13
#> [61] R6_2.5.1             compiler_4.0.5
```