# An Investigation on Li-ion Battery Recycling via In-situ Alloying: Influence of Slag Composition on Li and F Evaporation

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# File S1: Supplementary graphs and tables

*Table S1. List of initial sample compositions.*

| StdOrder | RunOrder | MgO (g) | CaO (g) | $SiO_2$ (g) | $Fe_2O_3$ (g) | Co (g) | Ni (g) | Mn (g) | Li (g) | Cu (g) | Al (g) | Fe (g) | C (g) | F (g) | O (g) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **1** | 7 | 0.10 | 1.94 | 1.29 | 8.25 | 1.6 | 0.5 | 0.3 | 0.3 | 0.4 | 0.1 | 0.1 | 3.0 | 0.2 | 1.4 |
| **2** | 8 | 0.05 | 1.06 | 0.71 | 9.00 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.2 | 0.2 | 1.6 |
| **3** | 5 | 0.10 | 2.51 | 0.72 | 8.25 | 1.6 | 0.5 | 0.3 | 0.3 | 0.4 | 0.1 | 0.1 | 3.0 | 0.2 | 1.4 |
| **4** | 14 | 0.05 | 1.37 | 0.39 | 9.00 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.2 | 0.2 | 1.6 |
| **5** | 15 | 0.33 | 1.80 | 1.20 | 8.25 | 1.6 | 0.5 | 0.3 | 0.3 | 0.4 | 0.1 | 0.1 | 3.0 | 0.2 | 1.4 |
| **6** | 3 | 0.18 | 0.98 | 0.65 | 9.00 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.2 | 0.2 | 1.6 |
| **7** | 9 | 0.33 | 2.33 | 0.67 | 8.25 | 1.6 | 0.5 | 0.3 | 0.3 | 0.4 | 0.1 | 0.1 | 3.0 | 0.2 | 1.4 |
| **8** | 11 | 0.18 | 1.27 | 0.36 | 9.00 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.2 | 0.2 | 1.6 |
| **9** | 10 | 0.30 | 3.11 | 1.24 | 7.60 | 1.4 | 0.4 | 0.3 | 0.3 | 0.4 | 0.1 | 0.0 | 2.7 | 0.2 | 1.3 |
| **10** | 12 | 0.10 | 1.05 | 0.42 | 9.12 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.3 | 0.2 | 1.6 |
| **11** | 6 | 0.15 | 0.99 | 1.21 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **12** | 17 | 0.15 | 1.78 | 0.42 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **13** | 2 | 0.01 | 1.67 | 0.67 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **14** | 13 | 0.29 | 1.47 | 0.59 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **15** | 16 | 0.15 | 1.57 | 0.63 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **16** | 4 | 0.15 | 1.57 | 0.63 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |
| **17** | 1 | 0.15 | 1.57 | 0.63 | 8.74 | 1.7 | 0.5 | 0.3 | 0.3 | 0.5 | 0.1 | 0.1 | 3.1 | 0.2 | 1.5 |

*Table S2. Chemical composition of samples dissolved in each dissolution step (wt.%).*

| StdOrder | RunOrder | Dissolution step | Al | Ca | Co | Cu | Fe | Mg | Si | Li | Ni | Mn | F | Total |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **1** | 7 | 1 | 3.9 | 22.0 | 29.1 | 6.9 | 125.7 | 1.1 | 0.0 | 2.4 | 9.0 | 5.5 | 2.3 | 208.1 |
| **1** | 7 | 2 | 1.1 | 5.7 | 4.2 | 1.0 | 17.1 | 0.8 | 4.9 | 0.7 | 1.6 | 1.0 | - | 38.1 |
| **2** | 8 | 1 | 2.2 | 5.6 | 13.5 | 3.0 | 58.7 | 0.6 | 0.0 | 1.2 | 4.4 | 2.6 | 0.6 | 92.5 |
| **2** | 8 | 2 | 1.2 | 0.4 | 0.5 | 0.1 | 2.4 | 0.3 | 0.3 | 0.3 | 0.2 | 0.1 | - | 5.8 |
| **3** | 5 | 1 | 1.7 | 10.0 | 10.3 | 2.4 | 38.6 | 0.4 | 0.0 | 0.5 | 3.2 | 1.9 | 0.5 | 69.5 |
| **3** | 5 | 2 | 0.2 | 1.0 | 1.3 | 0.3 | 5.1 | 0.1 | 0.8 | 0.1 | 0.4 | 0.2 | - | 9.5 |
| **4** | 14 | 1 | 2.9 | 12.0 | 14.7 | 3.1 | 60.8 | 1.0 | 0.1 | 0.7 | 4.5 | 2.1 | 1.2 | 103.1 |
| **4** | 14 | 2 | 1.0 | 4.0 | 2.4 | 0.6 | 10.3 | 0.8 | 1.6 | 0.2 | 0.8 | 0.5 | - | 22.2 |
| **5** | 15 | 1 | 2.2 | 12.7 | 16.5 | 3.1 | 68.7 | 1.3 | 0.0 | 1.3 | 5.2 | 2.4 | 0.5 | 113.9 |
| **5** | 15 | 2 | 0.5 | 2.0 | 3.4 | 0.7 | 13.9 | 0.3 | 0.6 | 0.3 | 1.1 | 0.7 | - | 23.4 |
| **6** | 3 | 1 | 0.1 | 0.6 | 9.9 | 2.3 | 42.6 | 0.0 | 0.2 | 0.3 | 3.9 | 2.1 | 0.1 | 62.1 |
| **6** | 3 | 2 | 0.3 | 0.5 | 1.4 | 0.3 | 9.4 | 0.1 | 1.2 | 0.1 | 0.5 | 0.3 | - | 17.3 |
| **7** | 9 | 1 | 5.2 | 38.4 | 4.0 | 0.9 | 18.8 | 28.5 | 0.5 | 3.5 | 1.3 | 1.4 | 21.3 | 124.0 |
| **7** | 9 | 2 | 6.6 | 41.2 | 0.9 | 0.3 | 5.2 | 7.1 | 0.0 | 4.1 | 0.4 | 0.8 | - | 66.5 |
| **8** | 11 | 1 | 0.2 | 0.8 | 7.8 | 2.2 | 33.9 | 0.1 | 0.0 | 0.3 | 3.1 | 1.7 | 0.1 | 50.1 |
| **8** | 11 | 2 | 0.0 | 0.1 | 31.4 | 6.4 | 105.3 | 0.0 | 2.1 | 0.7 | 9.6 | 6.3 | - | 161.9 |
| **9** | 10 | 1 | 1.3 | 14.2 | 15.8 | 3.3 | 55.9 | 1.4 | 0.0 | 0.9 | 4.9 | 3.1 | 0.8 | 101.8 |
| **9** | 10 | 2 | 0.5 | 2.6 | 1.3 | 0.3 | 5.4 | 0.4 | 13.6 | 0.2 | 0.4 | 0.3 | - | 24.8 |
| **10** | 12 | 1 | 2.5 | 6.0 | 11.9 | 3.2 | 50.5 | 0.6 | 0.0 | 1.0 | 4.3 | 2.6 | 1.0 | 83.6 |
| **10** | 12 | 2 | 0.9 | 0.3 | 0.2 | 0.0 | 1.2 | 0.2 | 9.1 | 0.2 | 0.1 | 0.0 | - | 12.3 |
| **11** | 6 | 1 | 2.5 | 6.7 | 16.8 | 4.0 | 60.7 | 1.3 | 0.0 | 1.5 | 5.5 | 3.3 | 0.4 | 102.7 |
| **11** | 6 | 2 | 1.6 | 1.5 | 1.9 | 0.4 | 8.0 | 0.8 | 3.5 | 0.4 | 0.6 | 0.4 | - | 19.1 |
| **12** | 17 | 1 | 1.8 | 7.9 | 9.7 | 1.9 | 40.9 | 0.6 | 0.1 | 0.4 | 1.7 | 1.5 | 0.0 | 66.4 |
| **12** | 17 | 2 | 0.1 | 0.2 | 0.3 | 0.1 | 1.1 | 0.1 | 0.7 | 0.0 | 0.1 | 0.1 | - | 2.7 |
| **13** | 2 | 1 | 2.8 | 11.1 | 17.7 | 3.9 | 64.9 | 0.5 | 0.0 | 0.8 | 5.8 | 3.4 | 1.2 | 112.2 |
| **13** | 2 | 2 | 0.5 | 0.7 | 1.2 | 0.2 | 5.2 | 0.1 | 2.4 | 0.2 | 0.4 | 0.3 | - | 11.1 |
| **14** | 13 | 1 | 2.4 | 8.4 | 14.5 | 3.2 | 53.1 | 0.7 | 0.0 | 1.0 | 4.5 | 2.7 | 1.0 | 91.6 |
| **14** | 13 | 2 | 0.5 | 1.9 | 1.9 | 0.4 | 7.9 | 0.3 | 4.4 | 0.2 | 0.6 | 0.4 | - | 18.5 |
| **15** | 16 | 1 | 3.9 | 11.1 | 16.1 | 3.2 | 59.4 | 1.2 | 0.0 | 1.0 | 4.7 | 3.1 | 1.3 | 105.0 |
| **15** | 16 | 2 | 0.5 | 1.1 | 1.8 | 0.4 | 7.7 | 0.2 | 0.8 | 0.2 | 0.7 | 0.4 | - | 13.8 |
| **16** | 4 | 1 | 9.1 | 40.5 | 2.2 | 0.5 | 9.0 | 6.3 | 0.9 | 1.7 | 0.5 | 0.5 | 5.9 | 77.1 |
| **16** | 4 | 2 | 1.3 | 5.6 | 2.0 | 0.4 | 8.4 | 1.2 | 1.3 | 0.4 | 0.6 | 0.4 | - | 21.8 |
| **17** | 1 | 1 | 2.5 | 13.5 | 15.2 | 3.1 | 65.0 | 1.6 | 0.0 | 0.9 | 4.7 | 2.9 | 1.2 | 110.7 |
| **17** | 1 | 2 | 0.4 | 1.3 | 2.4 | 0.5 | 9.8 | 0.2 | 1.2 | 0.1 | 0.9 | 0.6 | - | 17.5 |

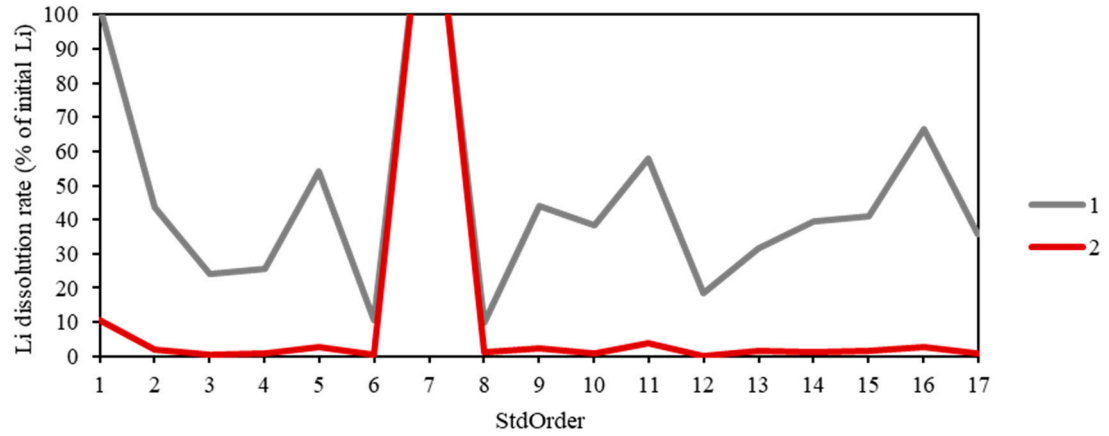



*Figure S1. Li dissolution rate (% of initial Li) in each dissolution step.*

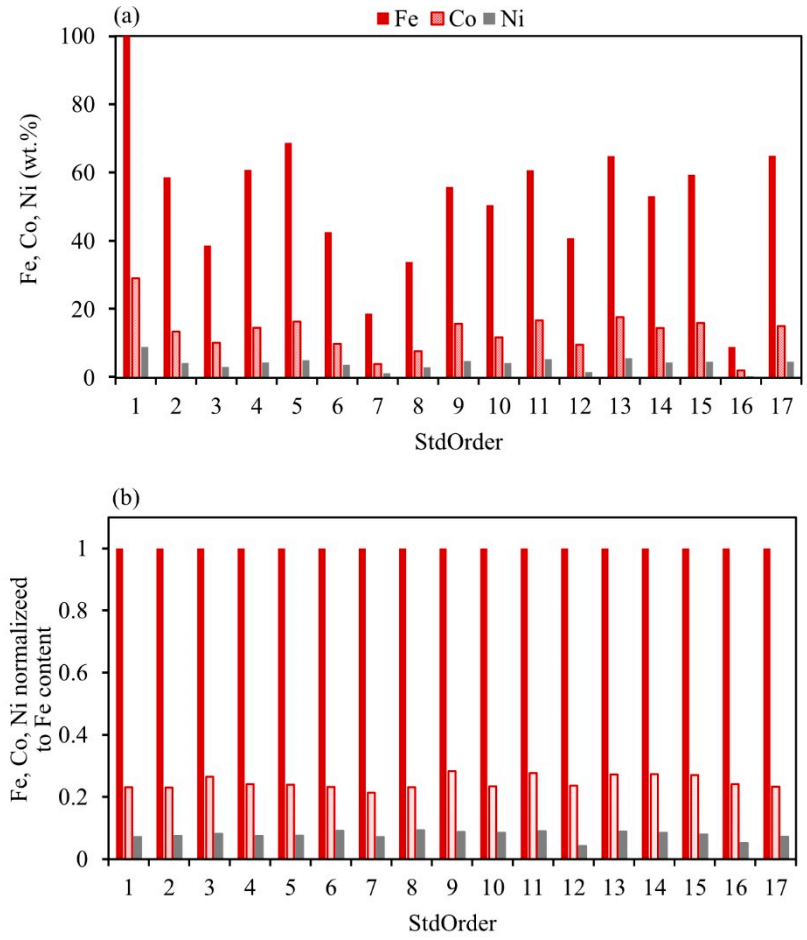



*Figure S2. (a) Fe, Co, and Ni contents in each sample, and (b) Fe, Co, and Ni contents normalized to Fe content in each sample.*

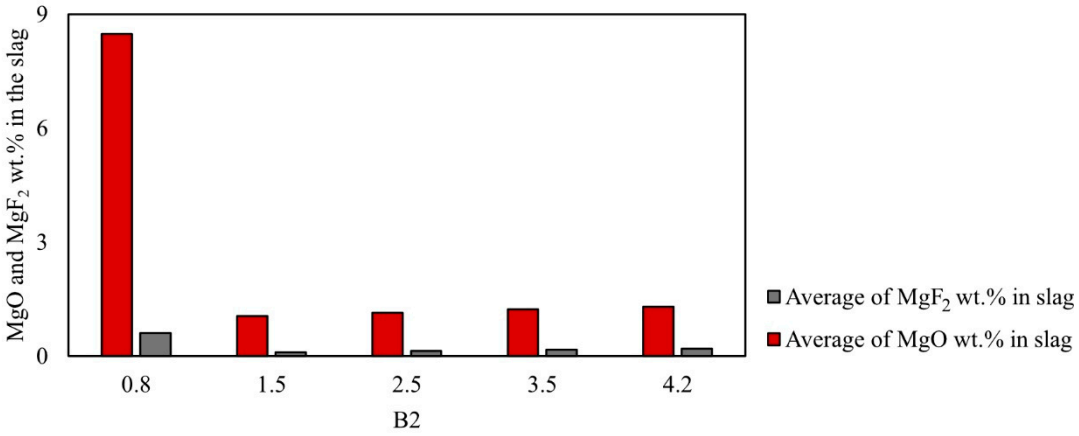



*Figure S3. MgO and $MgF_2$ contents in the slag, modeled by FactSage (Std deviation lower than 0.1).*

# File S2: Statistical Modeling by Minitab on Experimental Results

Note: As three experiments were excluded from statistical calculations, the observation orders (in the "Versus Order" graph) differ slightly from the initial StdOrders in the manuscript. The relationship between the two is detailed in the Table S3:

*Table S3. StdOrder (in manuscript) versus Observation order (in appendix).*

| **StdOrder in manuscript** | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Observation Order in Appendix** | - | 1 | 2 | 3 | 4 | 5 | - | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | - | 14 |

## Regression Analysis: Li evaporation versus Standardized B2

### Regression Equation

Li evaporation = 66.14 + 9.07 Standardized B2

### Coefficients

| Term | Coef | SE Coef | T-Value | P-Value | VIF |
|---|---|---|---|---|---|
| Constant | 66.14 | 3.17 | 20.85 | 0.000 | |
| Standardized B2 | 9.07 | 3.17 | 2.86 | 0.014 | 1.00 |

### Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---|---|---|---|
| 11.8690 | 40.54% | 35.58% | 16.68% |

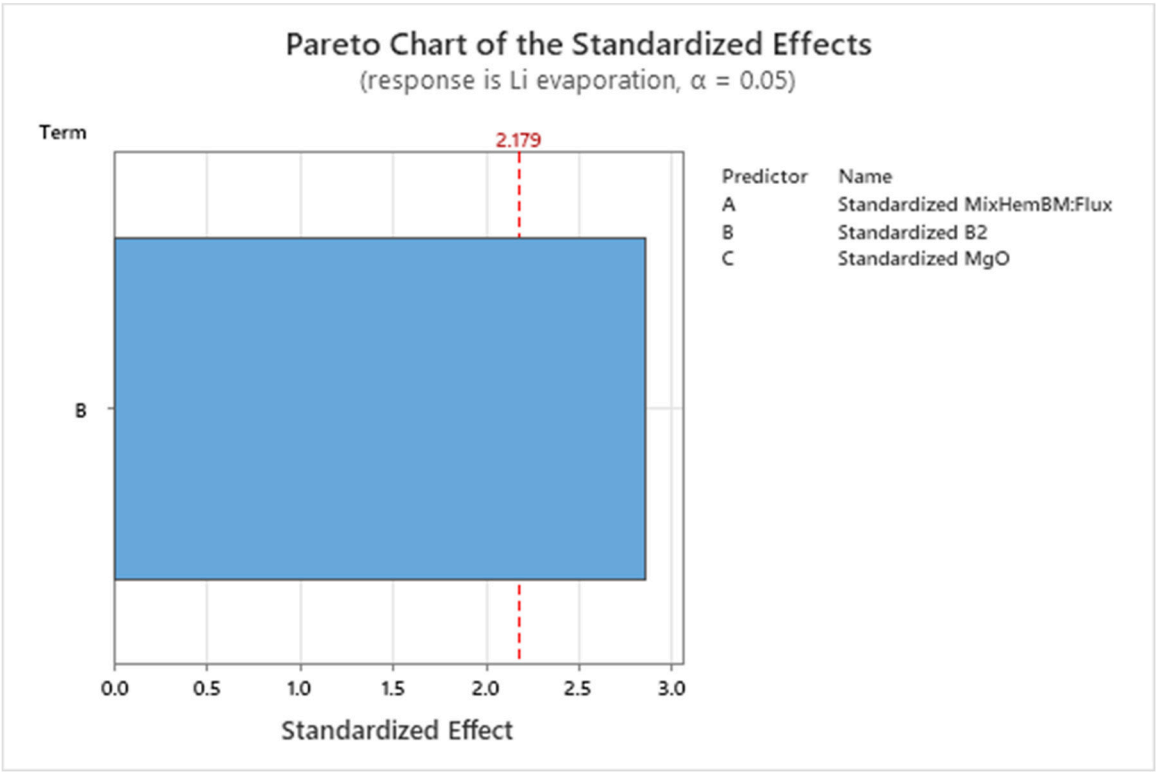
### Analysis of Variance

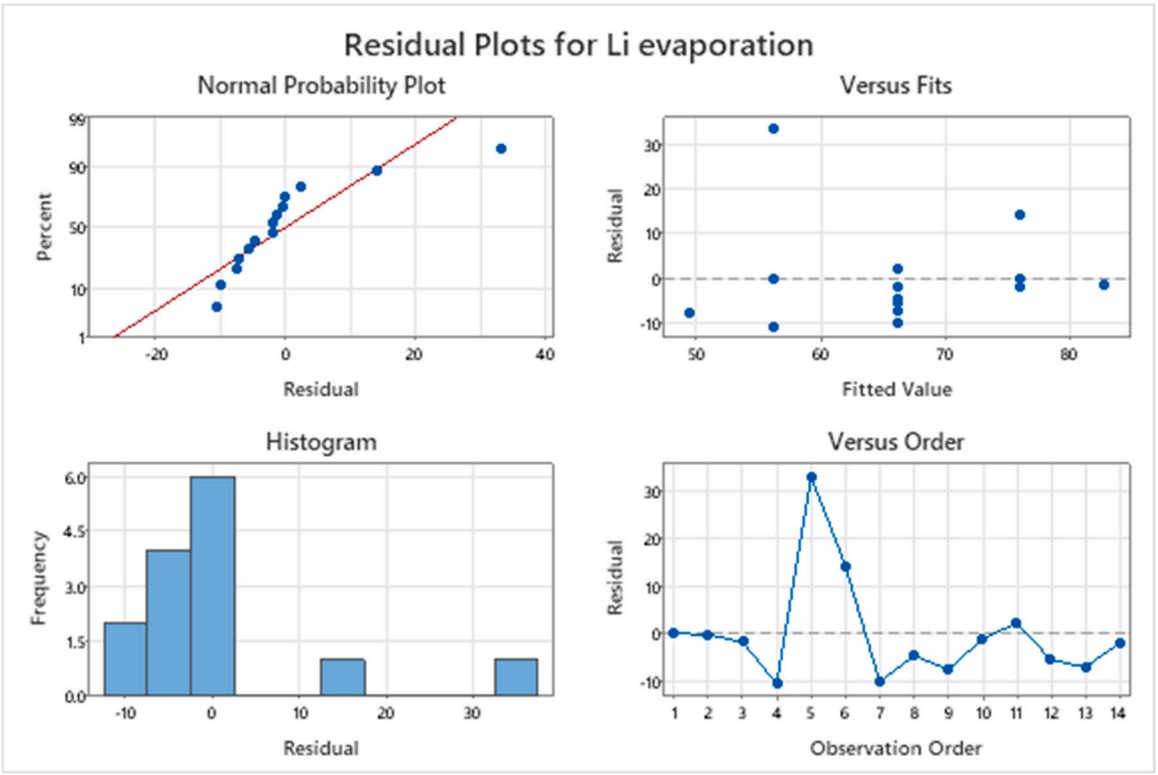
| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|---|---|---|---|---|---|
| Regression | 1 | 1152.46 | 1152.46 | 8.18 | 0.014 |
| Standardized B2 | 1 | 1152.46 | 1152.46 | 8.18 | 0.014 |
| Error | 12 | 1690.49 | 140.87 | | |
| Lack-of-Fit | 11 | 1677.60 | 152.51 | 11.83 | 0.223 |
| Pure Error | 1 | 12.89 | 12.89 | | |
| Total | 13 | 2842.94 | | | |

### Fits and Diagnostics for Unusual Observations

| Obs | Li evaporation | Fit | Resid | Std Resid | |
|---|---|---|---|---|---|
| 5 | 89.49 | 56.20 | 33.29 | 3.06 | R |

*R Large residual*


Pareto Chart of the Standardized Effects
(response is Li evaporation, α = 0.05)
Term
2.179
B
0.0
0.5
1.0
1.5
2.0
2.5
3.0
Standardized Effect
Predictor Name
A Standardized MixHemBM:Flux
B Standardized B2
C Standardized MgO


Residual Plots for Li evaporation
Normal Probability Plot
Percent
Residual
Versus Fits
Residual
Fitted Value
Histogram
Frequency
Residual
Versus Order
Residual
Observation Order

# Regression Analysis: F evaporation versus Standardized B2

## Regression Equation

F evaporation = 46.09 + 0.51 Standardized B2 + 15.11 Standardized B2*Standardized B2

## Coefficients

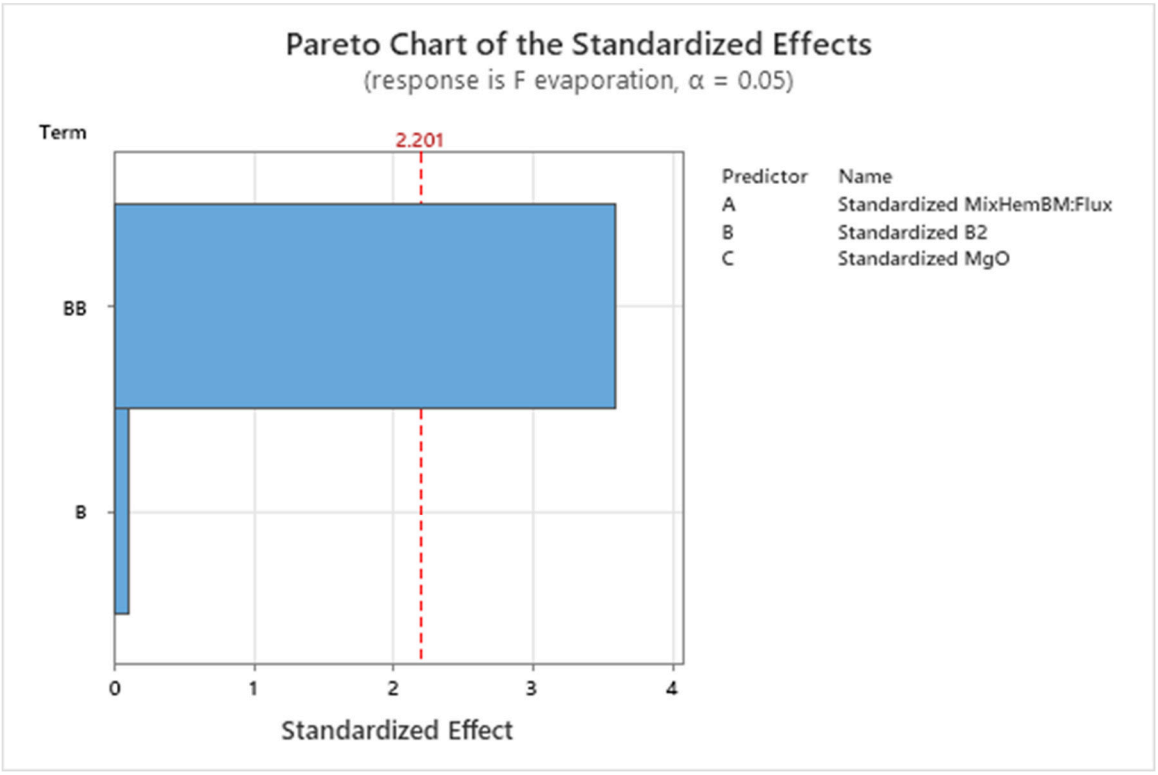
| Term | Coef | SE Coef | T-Value | P-Value | VIF |
|---|---|---|---|---|---|
| Constant | 46.09 | 6.33 | 7.28 | 0.000 | |
| Standardized B2 | 0.51 | 4.73 | 0.11 | 0.917 | 1.00 |
| Standardized B2*Standardized B2 | 15.11 | 4.20 | 3.60 | 0.004 | 1.00 |

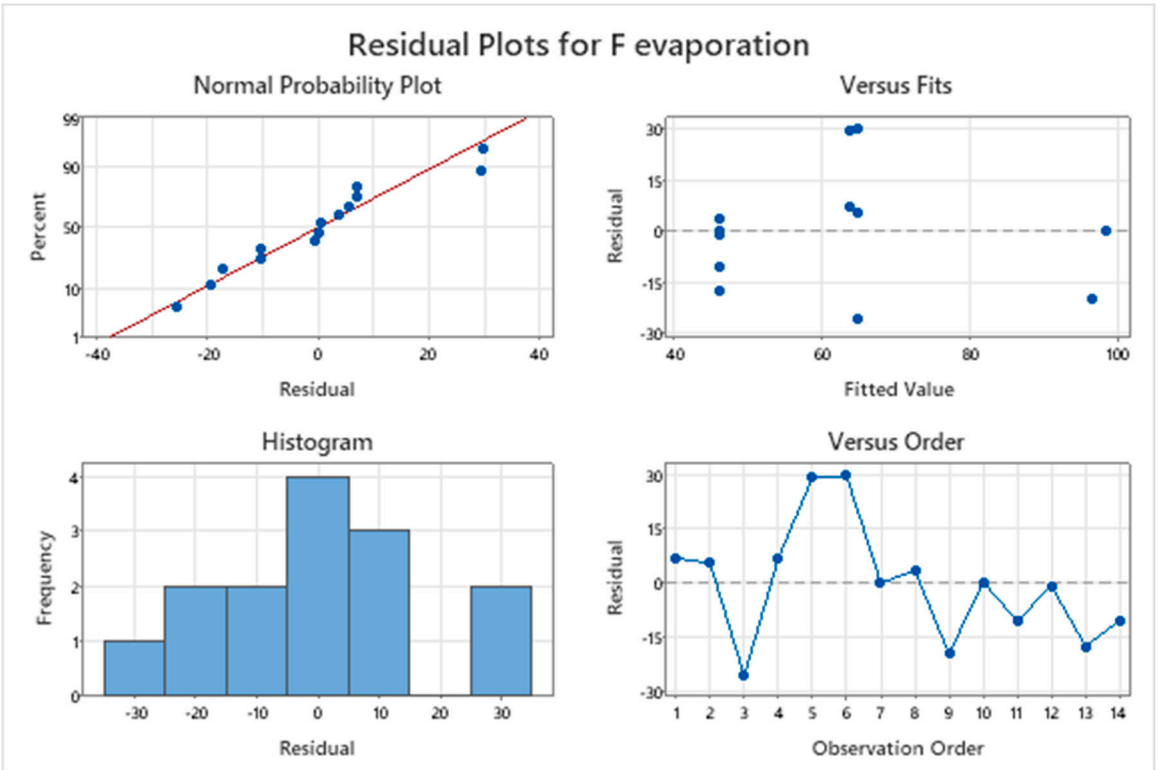
## Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---|---|---|---|
| 17.7035 | 54.04% | 45.69% | 4.39% |

## Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|---|---|---|---|---|---|
| Regression | 2 | 4054.45 | 2027.23 | 6.47 | 0.014 |
| Standardized B2 | 1 | 3.59 | 3.59 | 0.01 | 0.917 |
| Standardized B2*Standardized B2 | 1 | 4050.86 | 4050.86 | 12.92 | 0.004 |
| Error | 11 | 3447.57 | 313.42 | | |
| Lack-of-Fit | 10 | 3423.01 | 342.30 | 13.94 | 0.206 |
| Pure Error | 1 | 24.56 | 24.56 | | |
| Total | 13 | 7502.02 | | | |


Pareto Chart of the Standardized Effects
(response is F evaporation, α = 0.05)
Term
2.201
BB
B
0
1
2
3
4
Standardized Effect
Predictor Name
A Standardized MixHemBM:Flux
B Standardized B2
C Standardized MgO


Residual Plots for F evaporation
Normal Probability Plot
Percent
Residual
Versus Fits
Residual
Fitted Value
Histogram
Frequency
Residual
Versus Order
Residual
Observation Order