Machine Learning Methods for Early Prediction of End-of-Life for Lithium-ion Batteries

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Overview

- Lithium-Ion Batteries Introduction
- The Battery Data Set
 - Severson et al. 2019 (MIT)
 - 2 types of features (1D & 2D)
- Machine Learning Models
 - Support Vector Regression (SVR)
 - Multilayer Perceptron (MLP)
 - Long-Short-Term-Memory RNN (LSTM)
 - Convolutional Neural Network (CNN)
- Results & Conclusion



Lithium-Ion Batteries

Strong future

- Environmental concerns / Renewability
- Electric vehicles
- Consumer electronics
- Grid scale support

• End-of-life

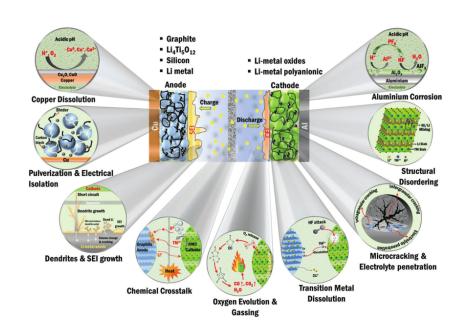
No longer capable of 80% capacity





Battery Degradation

- Complex electrochemical mechanisms
- Highly irregular behavior
- End-of-life
 - Wide range/high variability
 - Hard to predict
 - Severe consequences



Overview of the major degradation mechanisms

Narayan, R., Laberty-Robert, C., Pelta, J., Tarascon, J. M., & Dominko, R. (2021). Self-Healing: An emerging technology for next-generation smart batteries. *Advanced Energy Materials*, *12*(17), 2102652. https://doi.org/10.1002/aenm.202102652



Data-driven modeling

- Mechanism-Agnostic
 - Black box
- Machine learning & artificial intelligence
 - Captures behaviors and patterns
 - Harness information

Data sparsity

- Expensive
- Experiments take months



The Battery Data Set

- Severson et al. 2019 (MIT)
 - Model APR18650M1A
- Largest open-source data set of its kind
- "Cycles"
 - Charged/Discharged until end-of-life
 - Variety of conditions & collect data



Severson, K. A., Attia, P. M., Jin, N., Perkins, N., Jiang, B., Yang, Z., Chen, M. H., Aykol, M., Herring, P. K., Fraggedakis, D., Bazant, M. Z., Harris, S. J., Chueh, W. C., & Braatz, R. D. (2019). Data-driven prediction of battery cycle life before capacity degradation. *Nature Energy*, 4(5), 383–391. doi.org/10.1038/s41560-019-0356-8



Severson et al. 2019

- End-of-life
 - No longer capable of 80% capacity
 - Integer (~300-2500)
 - Number of "cycles"
 - Target of machine learning (y)



GOAL

- Predict end-of-life
- Data from first 100 cycles (or less)



Tabular Data

- Used in original paper
- Calculated summary statistics "1-dimensional"
- "FULL" feature set
 - 10 features
- "DISCHARGE" feature set
 - 6 features
 - Using only discharge capacity-voltage data

TABLE I

SUMMARY FEATURES FROM DISCHARGE VOLTAGE CURVES USING THE 10TH AND 100TH CYCLES (ΔQ100-10(V))

- Minimum (ΔQ₁₀₀₋₁₀(V))
- Variance $(\Delta Q_{100-10}(V))$
- Skewness ($\Delta Q_{100-10}(V)$)
- Kurtosis ($\Delta Q_{100-10}(V)$)
- Discharge capacity of cycle 2
- Max discharge capacity Discharge capacity of cycle 2



Tabular Data

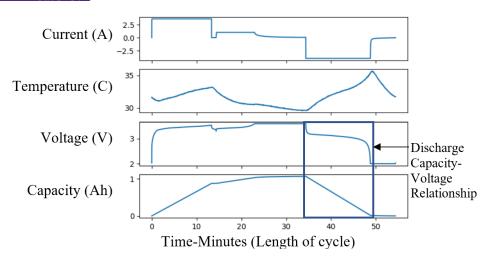
	4									•			
In [135	data	dataset											
Out[135		cell	cell_life	minimum_dQ_100_10	variance_dQ_100_10	skewness_dQ_100_10	kurtosis_dQ_100_10	mean_dQ_100_10	Slope_Cap_Fade_2_100	Intercept_Cap_Fade			
	0	b1c0	1852.0	-2.072648	-5.014861	-0.274041	0.129790	-2.541602	1.692000e-05	1.			
	1	b1c1	2160.0	-1.958457	-5.013960	-0.367163	0.012464	-2.387257	5.535293e-06	1.			
	2	b1c2	2237.0	-1.764058	-4.737000	0.033502	-0.457627	-2.348070	1.029563e-05	1.			
	3	b1c3	1434.0	-1.722149	-4.442613	-0.357486	0.039579	-2.127507	1.713635e-05	1.			
	4	b1c4	1709.0	-1.855177	-4.647744	-0.440634	0.125101	-2.240332	1.899432e-05	1.			
	119	b3c39	1156.0	-1.758008	-4.454635	-0.533817	0.001307	-2.191895	-6.583238e-06	1.			
	120	b3c40	796.0	-1.656517	-4.295108	-0.501096	0.056338	-2.041969	-7.692147e-06	1.			
	121	b3c41	786.0	-1.616310	-4.219509	-0.447169	0.026586	-2.011082	9.732245e-06	1.			
	122	b3c44	940.0	-1.585275	-4.131496	-0.611514	0.092860	-1.949579	4.689737e-06	1.			
	123	b3c45	1801.0	-1.783097	-4.520851	-0.483937	0.070824	-2.146804	-1.258454e-07	1.			

124 rows × 18 columns





- Capacity values (Ah) during "discharge"
- Interpolated at 100 evenly spaced voltages (3.5V-2V)



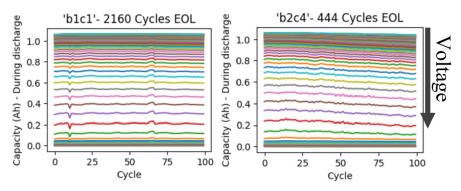
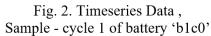


Fig. 3. Discharge Capacity-Voltage Curves Comparison of batteries with high and low end-of-life





Discharge Capacity-Voltage Data

- Two-dimensional
 - 99 cycles X 100 voltages
 - (skip cycle 1)
 - 9900 capacities (Ah) per battery
- Used in follow-up paper
 - Attia et al.
 - (and other papers)

	Cycle 2	Cycle 3		Cycle 100
3.5 V	Ah	Ah	Ah	Ah
485 V	Ah	Ah	Ah	Ah
•••	Ah	Ah	Ah	Ah
2 V	Ah	Ah	Ah	Ah

100 evenly spaced voltages

3.485



Discharge Capacity-Voltage Data

- Two-dimensional
 - 99 cycles X 100 voltages
 - (skip cycle 1)

```
print(train_Qdlin.std(),train_Qdlin.mean())
print(test_Qdlin.std(),test_Qdlin.mean())
print(secondary_test_Qdlin.std(),secondary_test_Qdlin.mean())

tensor(1.0000, dtype=torch.float64) tensor(-0.6982, dtype=torch.float64)
tensor(0.8786, dtype=torch.float64) tensor(-0.6188, dtype=torch.float64)
tensor(0.4862, dtype=torch.float64) tensor(-0.3183, dtype=torch.float64)

In [160... # torch.save(train_Qdlin, r'data/DischargeCapacity-InterpolatedData_Train.pt')
# torch.save(test_Qdlin, r'data/DischargeCapacity-InterpolatedData_Test.pt')
# torch.save(secondary_test_Qdlin, r'data/DischargeCapacity-InterpolatedData_Secondary_Test.pt')
```



Machine Learning Models

- Tabular data (1D)
 - Support Vector Regression (SVR)
 - Multilayer Perceptron (MLP)
- Discharge capacity-voltage data (2D)
 - Long-Short-Term-Memory RNN (LSTM)
 - Convolutional Neural Network (CNN)



Support Vector Regression (SVR)

- SVM- Projection to higher dimensional feature space
 - Kernel (Sigmoid, RBF, etc.)
 - Hyperplane ► real value (Regression)
- Hyperparameters (cross-validation)
 - Epsilon (margin)
 - C (L2 regularization)
 - Gamma

RBF Kernel:
$$K(x,y) = e^{-}(\gamma ||x-y||^2)$$

Sigmoid Kernel:
$$K(x,y) = tanh(\gamma . x^T y + r)$$





SVR

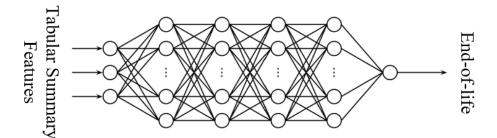
Code sklearn.svm.SVR

SVR Hyperparameters

```
def SVRmodel(features, target, params):
    X train=Training Data[features].to numpy()
   y_train=Training_Data[target].to_numpy()
   X_test=Testing_Data[features].to_numpy()
   y_test=Testing_Data[target].to_numpy()
   X sec=Secondary Data[features].to numpy()
   y_sec=Secondary_Data[target].to_numpy()
    scaler = preprocessing.StandardScaler().fit(X train)
   X_train = scaler.transform(X_train)
   X_test = scaler.transform(X_test)
   X sec = scaler.transform(X sec)
###################
    model = SVR()
    grid=GridSearchCV(estimator=model,param_grid=params,cv=40)
grid.fit(X train, y train)
    print(grid.best params )
   y_pred_train=10**(grid.predict(X_train))
   y pred test=10**(grid.predict(X test))
   y pred sec=10**(grid.predict(X sec))
    y_train=10**(y_train)
   y_test=10**(y_test)
   y_sec=10**(y_sec)
    RMSE=list()
   MPE=list()
    RMSE.append(np.sqrt(mean_squared_error(y_train, y_pred_train)))
   MPE.append(float(np.mean(np.abs((y_train - y_pred_train.reshape(-1,1))) / y_train * 100)))
    RMSE.append(np.sqrt(mean squared error(y test, y pred test)))
   MPE.append(float(np.mean(np.abs((y_test - y_pred_test.reshape(-1,1))) / y_test * 100)))
    RMSE.append(np.sqrt(mean_squared_error(y_sec, y_pred_sec)))
   MPE.append(float(np.mean(np.abs((y_sec - y_pred_sec.reshape(-1,1))) / y_sec * 100)))
```

Multilayer Perceptron (MLP)

- Neural network for tabular data
- Nonlinear activation
 - Relu
- Optimization
 - Adaptive momentum
- Regularization
 - L2 (alpha hyperparameter)







MLP

Code sklearn.neural_network MLPRegressor

```
def MLPmodel(features, target, params):
   X train=Training Data[features].to numpy()
   y train=Training Data[target].to numpy()
   X_test=Testing_Data[features].to_numpy()
   y test=Testing Data[target].to numpy()
   X sec=Secondary Data[features].to numpy()
   v sec=Secondary Data[target].to numpy()
   scaler = preprocessing.StandardScaler().fit(X train)
   X train = scaler.transform(X train)
   X test = scaler.transform(X test)
   X sec = scaler.transform(X sec)
model = MLPRegressor(random state=0, max iter=10000)
   grid = GridSearchCV(estimator=model, param grid=params)
#################
   grid.fit(X_train, y_train)
   print(grid.best params )
   y pred train=10**(grid.predict(X train))
   y pred test=10**(grid.predict(X test))
   v pred sec=10**(grid.predict(X sec))
```

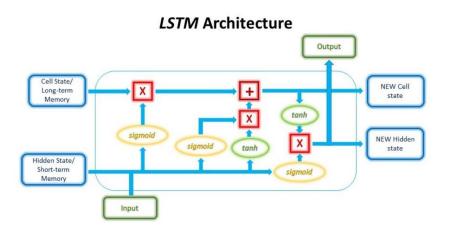
MLP Hyperparameters

The hidden layer Sizes used for cross validation were chosen to match the Murphy's PML1 page 429, Neural Networks For Tabular data, Which references the following

website by Brendan Hasz. As well as the shape of the MLP used for MNIST(Page 427). The size to was reduced match this data's Small feature set.

Long-Short-Term-Memory RNN (LSTM)

- Timeseries data
- Cell state
- Hidden state



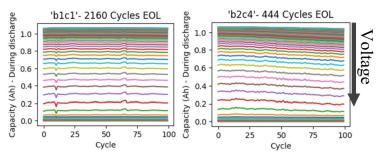


Fig. 3. Discharge Capacity-Voltage Curves Comparison of batteries with high and low end-of-life

$$\begin{split} i_t &= \sigma(W_{ii}x_t + b_{ii} + W_{hi}h_{t-1} + b_{hi}) \\ f_t &= \sigma(W_{if}x_t + b_{if} + W_{hf}h_{t-1} + b_{hf}) \\ g_t &= \tanh(W_{ig}x_t + b_{ig} + W_{hg}h_{t-1} + b_{hg}) \\ o_t &= \sigma(W_{io}x_t + b_{io} + W_{ho}h_{t-1} + b_{ho}) \\ c_t &= f_t \odot c_{t-1} + i_t \odot g_t \\ h_t &= o_t \odot \tanh(c_t) \end{split}$$

Fig. 6. The sequence of calculations at each cell in LSTM [10] (input gate, forget gate, cell gait, & output gate)



Long-Short-Term-Memory RNN (LSTM)

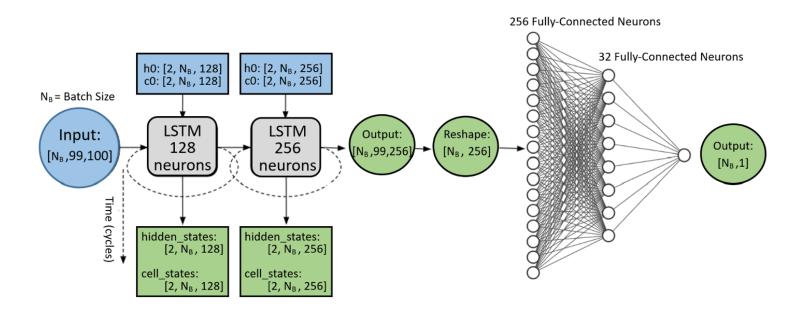


Fig. 5. Architecture of Long-Short-Term-Memory
Recurrent Neural network



LSTM

- Pytorch

```
from torch.utils.data import Dataset
class CustomDataset(Dataset):
    def __init__(self, features, targets):
        self.features = features
        self.targets = targets

def __getitem__(self, idx):
        return self.features[idx], self.targets[idx]

def __len__(self):
    return len(self.features)
```

```
train_Dataset = CustomDataset(train_Qdlin,y_train)
test_Dataset = CustomDataset(test_Qdlin,y_test)
secondary_test_Dataset = CustomDataset(secondary_test_Qdlin,y_sec)

train_Qdlin_loader = DataLoader(train_Dataset, batch_size=41, shuffle=True)
test_Qdlin_loader = DataLoader(test_Dataset, batch_size=42, shuffle=True)
secondary_test_Qdlin_loader = DataLoader(secondary_test_Dataset, batch_size=40, shuffle=True)
```





LSTM

– Pytorch torch.nn.LSTM

```
def train(model,train_loader,criterion,optimizer,epochs,device):
    for epoch in range(epochs):
       model.train()
        for inputs, targets in train_loader:
            optimizer.zero_grad()
            inputs = inputs.to(device)
            targets = targets.to(device)
            outputs = model(inputs)
            outputs.to(device)
            loss = criterion(outputs, targets)
            loss.backward()
            optimizer.step()
        model.eval()
        if(epoch%100 == 0):
            print('--',epoch,'-epoch--RMSE-- ',
                  np.sqrt(mean_squared_error(10**(targets.cpu().detach().numpy()+y_train_mean.numpy()),
                                          10**(outputs.cpu().detach().numpv()+v train mean.numpv()))))
    return model
```

```
class LSTMRNN(nn.Module):
    def init (self, input size, hidden size, hidden size2, num layers,
                 dropout, num_dense_neurons):
       super(LSTMRNN, self).__init__()
       self.input_size = input_size
       self.hidden size = hidden size
       self.hidden size2 = hidden size2
       self.num layers = num layers
       self.dropout = dropout
       self.num dense neurons = num dense neurons
       self.firstEpochFlag = True
       self.dropout layer = nn.Dropout(self.dropout)
       self.lstm = nn.LSTM(input_size = self.input_size,
                           hidden size = self.hidden size,
                            num layers = self.num layers,
                             dropout = self.dropout,
                           batch first = True)
       self.lstm2 = nn.LSTM(input size = self.hidden size.
                           hidden size = self.hidden size2,
                            num layers = self.num layers,
                             dropout = self.dropout,
                           batch first = True)
       self.linear laver = nn.Sequential(
            #nn.Dropout(self.dropout)
           nn.Linear(self.hidden size2, self.num dense neurons),
           nn.ReLU().
           nn.Linear(num_dense_neurons,1),
    def forward(self, x):
       if(self.firstEpochFlag):print(x.shape)
       h0 = Variable(torch.zeros(self.num_layers, x.shape[0], self.hidden_size,device=x.device))
       c0 = Variable(torch.zeros(self.num layers, x.shape[0], self.hidden size,device=x.device))
       lstm output, (h1, c1) = self.lstm(x, (h0, c0))
       if(self.firstEpochFlag):print(lstm_output.shape)
       h00 = Variable(torch.zeros(self.num layers, x.shape[0], self.hidden size2,device=x.device))
       c00 = Variable(torch.zeros(self.num_layers, x.shape[0], self.hidden_size2,device=x.device))
       lstm output2,(h2, c2) = self.lstm2(lstm output,(h00, c00))
       if(self.firstEpochFlag):print(lstm output2.shape)
       flat = lstm output2[:, -1, :]#last projected timestep (cycle)
       if(self.firstEpochFlag):print(flat.shape)
       output = self.linear layer(flat)
       if(self.firstEpochFlag):
            (output.shape)
            self.firstEpochFlag=False
       return output
```

LSTM

– Pytorch torch.nn.LSTM

Train & Test

```
criterion=nn.MSELoss()
  model=LSTMRNN(input size, hidden size, hidden size2, num layers,dropout, num dense neurons)
  optimizer=optim.Adam(model.parameters(),weight decay=weight decay, lr=lr)
  model = model.to(device)
  model = train(model,train_Qdlin_loader,criterion,optimizer,epochs,device)
torch.Size([41, 99, 100])
torch.Size([41, 99, 128])
torch.Size([41, 99, 256])
torch.Size([41, 256])
-- 0 -epoch--RMSE-- 325.713504214364
-- 100 -epoch--RMSE-- 191.60093282624348
-- 200 -epoch--RMSE-- 118.58940215229036
-- 300 -epoch--RMSE-- 84.2295624764314
-- 400 -epoch--RMSE-- 65.92709414591444
-- 500 -epoch--RMSE-- 52.18300708080519
-- 600 -epoch--RMSE-- 46.43477470967451
-- 700 -epoch--RMSE-- 43.97749221237031
-- 800 -epoch--RMSE-- 42.366674639828105
-- 900 -epoch--RMSE-- 38.88688856498636
-- 1000 -epoch--RMSE-- 37.52256793585603
-- 1100 -epoch--RMSE-- 36.52446165852783
-- 1200 -epoch--RMSE-- 35.78776480837691
-- 1300 -epoch--RMSE-- 35.32158666670042
-- 1400 -epoch--RMSE-- 34.85211871891183
  testALL(model)
35.13341232095763 --- RMSE--- Train
1.8556565652851051 ---MPE---Train
101.92579699870431 --- RMSE--- Test
11.284582893046336 --- MPE--- Test
193.827633377642 --- RMSE--- Test Secondary
14.209379170879178 ---MPE---Test Secondary
```

```
class LSTMRNN(nn.Module):
   def init (self, input size, hidden size, hidden size2, num layers,
                 dropout, num_dense_neurons):
       super(LSTMRNN, self).__init__()
       self.input_size = input_size
       self.hidden size = hidden size
       self.hidden size2 = hidden size2
       self.num layers = num layers
       self.dropout = dropout
       self.num dense neurons = num dense neurons
       self.firstEpochFlag = True
       self.dropout layer = nn.Dropout(self.dropout)
       self.lstm = nn.LSTM(input_size = self.input_size,
                           hidden size = self.hidden size,
                            num layers = self.num layers,
                             dropout = self.dropout,
                           batch first = True)
       self.lstm2 = nn.LSTM(input size = self.hidden size.
                           hidden size = self.hidden size2,
                            num layers = self.num layers,
                             dropout = self.dropout,
                           batch first = True)
       self.linear laver = nn.Sequential(
            #nn.Dropout(self.dropout)
           nn.Linear(self.hidden size2, self.num dense neurons),
           nn.ReLU(),
           nn.Linear(num dense neurons,1),
    def forward(self, x):
       if(self.firstEpochFlag):print(x.shape)
       h0 = Variable(torch.zeros(self.num_layers, x.shape[0], self.hidden_size.device=x.device))
       c0 = Variable(torch.zeros(self.num layers, x.shape[0], self.hidden size,device=x.device))
       lstm output,(h1, c1) = self.lstm(x,(h0, c0))
       if(self.firstEpochFlag):print(lstm_output.shape)
       h00 = Variable(torch.zeros(self.num layers, x.shape[0], self.hidden size2,device=x.device))
       c00 = Variable(torch.zeros(self.num_layers, x.shape[0], self.hidden_size2,device=x.device))
       lstm output2,(h2, c2) = self.lstm2(lstm output,(h00, c00))
       if(self.firstEpochFlag):print(lstm output2.shape)
       flat = lstm output2[:, -1, :]#last projected timestep (cycle)
       if(self.firstEpochFlag):print(flat.shape)
       output = self.linear layer(flat)
       if(self.firstEpochFlag):
            (output.shape)
            self.firstEpochFlag=False
       return output
```

Convolutional Neural Network (CNN)

Convolution layers

Max Pool layers

Fully connected layers

- Discharge capacity-voltage data
 - Two-dimensional (99X100)
- Tabular summary data (discharge features)
 - 6 features





Convolutional Neural Network (CNN)

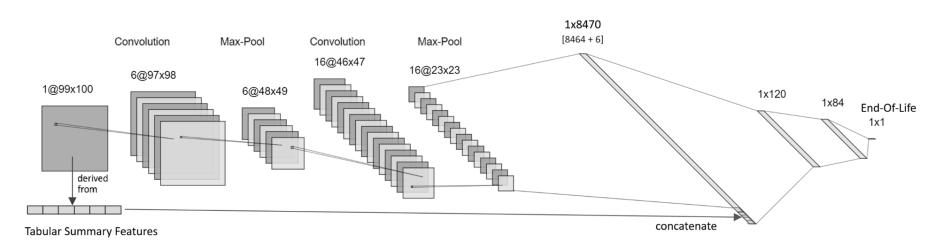


Fig. 7. Architecture of Convolutional Neural network



CNN

– Pytorch torch.nn.Conv2d

```
from torch.utils.data import Dataset
class CustomDataset(Dataset):
    def __init__(self, features, targets, tabular):
        self.features = features
        self.targets = targets
        self.tabular = tabular #Merge tabular data set

def __getitem__(self, idx):
        return self.features[idx], self.targets[idx], self.tabular[idx]

def __len__(self):
    return len(self.features)
```

```
class CNN(nn.Module):
    def __init__(self,inputSize):
        super(CNN, self). init ()
       self.firstEpochFlag=True
       # Calculates size of first linear layer
        # This allows the Module to be used with different number of cycles
        self.linearSize=((((inputSize-2)//2)-2)//2)*23*16+6
        # Convolutional architecture from Attia et al.
        self.convolutional laver = nn.Sequential(
           nn.Conv2d(in_channels=1, out_channels=6, kernel_size=(3,3)),
            nn.ReLU(),
            nn.MaxPool2d(kernel_size=2, stride=2),
            #nn.BatchNorm2d(3).
            nn.Conv2d(in_channels=6, out_channels=16, kernel_size=(3,3)),
            nn.ReLU(),
            nn.MaxPool2d(kernel size=2, stride=2, padding=0),
        self.linear_layer = nn.Sequential(
            nn.Linear(self.linearSize,120),
            #nn.Dropout(.1)
            nn.ReLU(),
            nn.Linear(120,84),
            nn.ReLU(),
            nn.Linear(84,1),
    def forward(self, x,tabular):
        #Print the shapes of the data on the first Epoch
       if(self.firstEpochFlag):print(x.shape)
       x = x.view(-1, 1.x.shape[1].99)
       if(self.firstEpochFlag):print(x.shape)
        clout = self.convolutional layer(x)
       if(self.firstEpochFlag):print(clout.shape)
       flat = torch.flatten(clout, 1)
       if(self.firstEpochFlag):print(flat.shape)
        #Concatenate tabular features!
       flat=torch.cat((flat, tabular), -1)
       if(self.firstEpochFlag):
            print(tabular.shape)
            print(flat.shape)
            self.firstEpochFlag=False
       output = self.linear_layer(flat)
       return output
```

Results & Conclusion

RMSE & MAE

- Repeat training/testing
 - 60 cycles
 - 40 cycles

$$MAE = \frac{1}{n} \sum_{i=1}^{n} \left| \widehat{y}_i - y_i \right|$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (\widehat{y}_i - y_i)^2}$$



Summary data results

TABLE II
RESULTS FROM TABULAR SUMMARY DATA

MODEL	RMSE TRAIN	RMSE TEST	RMSE TEST #2	MPE TRAIN	MPE TEST	MPE TEST #2
Severson et al. Elastic Net "Variance" model	(103)	(138)	(196)	(14)	(13)	(11)
"Discharge" model	(76)	(86)	(173)	(10)	(10)	(9)
"Full" model	(51)	(100)	(214)	(6)	(8)	(11)
Support Vector Regression - "Discharge" features	79	79	172	10	10	9
Support Vector Regression - "Full" features	54	115	200	7	9	11
Multilayer Perceptron - "Discharge" features	67	105	159	8	8	8
Multilayer Perceptron - "Full" features	39	138	176	4	8	10



Discharge capacity-voltage data results

TABLE III
RESULTS FROM DISCHARGE CAPACITY-VOLTAGE DATA

MODEL	RMSE TRAIN	RMSE TEST	RMSE TEST #2	MPE TRAIN	MPE TEST	MPE TEST #2
Xu et al. [9] LSTM 100 cycles	(42)	(91)	(215)			
60 cycles	(48)	(88)	(212)			
40 cycles	(51)	(127)	(224)			
LSTM 100 cycles	35	101	193	1	11	14
60 cycles	38	316	220	2	18	17
40 cycles	49	127	218	3	12	17
Attia et al. [8] Convolutional Neural Network	(17)	(72)	(204)			
Convolutional neural network 100 cycles	16	83	141	1	6	8
60 cycles	14	184	203	1	8	12
40 cycles	11	80	206	1	6	11



Discharge capacity-voltage data results

- Convolutional neural network (100 cycles)
 - Best model

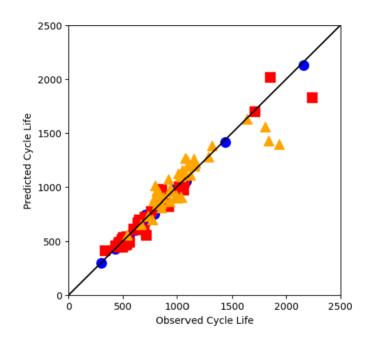


Fig. 8. Predicted vs Observed End-Of-Life CNN - first 100 cycles





Discussion

Overfitting



Future work

- Use more data
 - Voltage
 - Current
 - Temperature
 - Internal resistance
- Trade-off
 - Capacity data is "free"

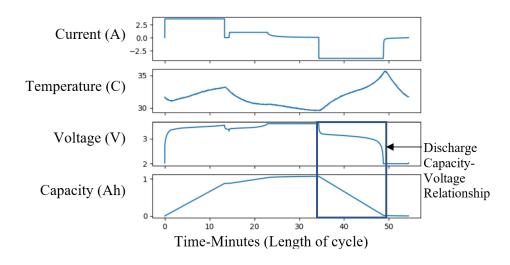


Fig. 2. Timeseries Data, Sample - cycle 1 of battery 'b1c0'



Machine Learning
Methods for Early
Prediction of End-of-Life
for Lithium-ion Batteries

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