

Supporting Information (SI)

**Development of a Novel Carbon-Coating Strategy for Producing Core-Shell
Structured Carbon Coated LiFePO₄ for Improved Li-ion Battery Performance**

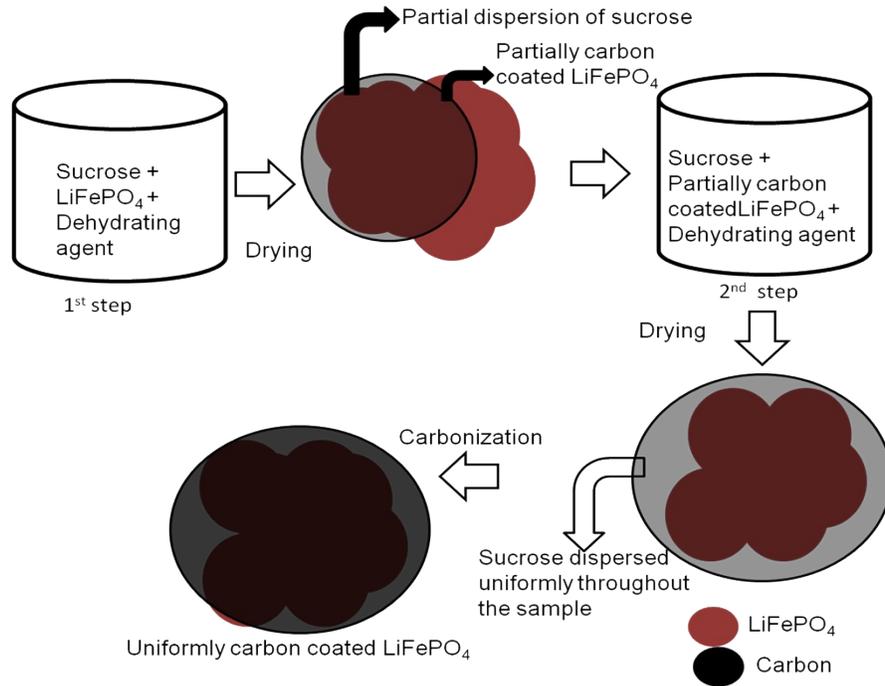
Parakandy Muzhikara Pratheeksha,^{a,d} Erabhoina Hari Mohan,^a Bulusu Venkata Sarada,^b
Mantripragada Ramakrishna,^c Kalyan Hembram,^a Pulakhandam Veera Venkata Srinivas,^a Paul
Joseph Daniel,^d Tata Narasinga Rao,^a Srinivasan Anandan,^{a*}

^a Centre for Nano Materials, ^b Centre for Solar Energy Materials, ^c Centre for Materials
Characterization and Testing, International Advanced Research Centre for Powder Metallurgy
and New Materials, Hyderabad-500 005, India

^d Department of Physics, National Institute of Technology, Warangal-506 004, India

*Email: anandan@arci.res.in

13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31

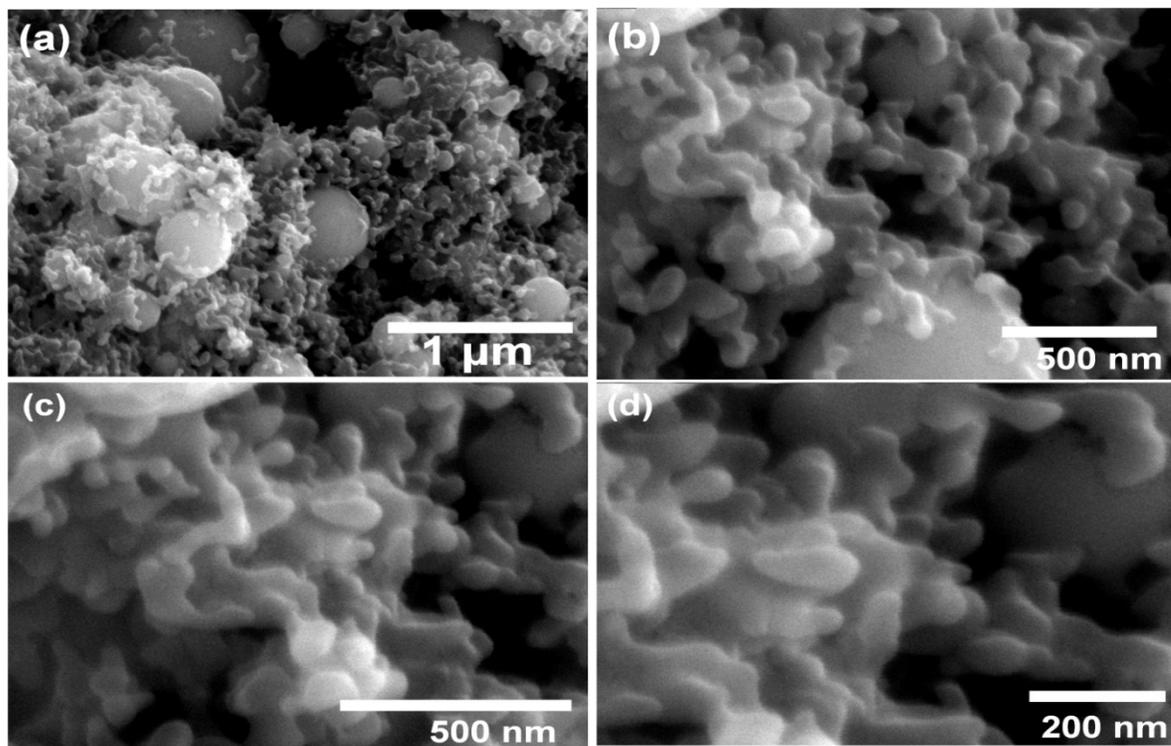


32 **Fig. S1.** Schematic showing the continuous carbon coating by DAP process
33

34 The schematic diagram showing the continuous carbon coating process by DAP process is
35 included in the supporting information as Fig. S1. The dehydrating agent, which when added to
36 the sucrose solution during carbon coating, plays a crucial role in achieving a uniform carbon
37 coverage on LFP. It helps in catenation of carbon atoms through dehydration of water molecule
38 from sucrose molecules, which is followed by thermo-polymerization of dehydrated sucrose
39 molecules and thereby contributing uniform graphitic carbon coating surrounding LFP particles
40 after carbonization.

41 The FE-SEM images of as-synthesized heat-treated and carbon coated LFP have included in **Fig.**
42 **S2, Fig. S3** and **Fig. S4**. It is observed that the morphology of heat-treated LFP and carbon coated
43 LFP (**Fig. S3 and Fig S4**) are similar, implying that carbon coating doesn't change the morphology
44 of LFP particles before and after carbon coating as carbon coating is purely a surface modification.
45 However, the morphology of as-synthesized LFP is different from these LFP particles and the
46 reasons are not clear at this juncture. Similar kind of difference in morphology between as-
47 synthesized and heat-treated particles by flame spray pyrolysis has reported by Wagner research
48 group (c.f. Wagner et al, Transl. Mater. Res, 2016, 3, 025001), who extensively studied the
49 synthesis of various nanoparticles by flame spray pyrolysis unit. They reported that as-synthesized
50 particles showed mixed morphology when obtain from FSP before annealing and later showed
51 uniform morphology after annealing, indicating that some un-reacted molecules during FSP
52 process responsible for mixed morphology. However, later the un-reacted molecules fused
53 together during heat treatment and form uniform morphology. Similarly, in the present study,
54 though as-synthesized LFP shows mixed morphology, the same LFP particles shown uniform
55 morphology after heat treatment, demonstrating that LFP particles produced by FSP upon
56 annealing shows homogeneous morphology.

57



58

59

60

61

62

63

64

65

66

67

68

69

70

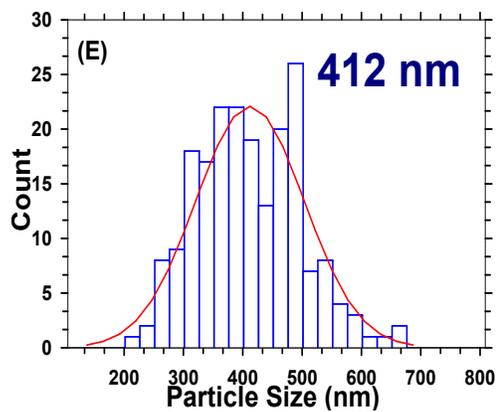
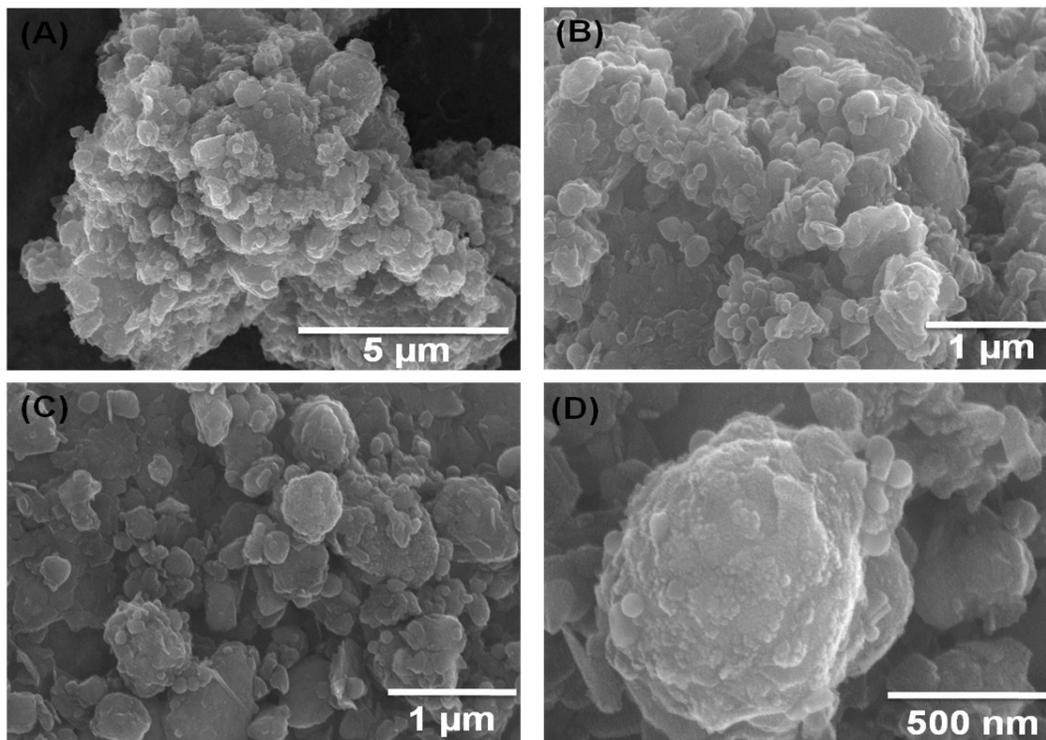
71

72

73 **Fig. S2:** FE-SEM images of as-synthesized LiFePO₄

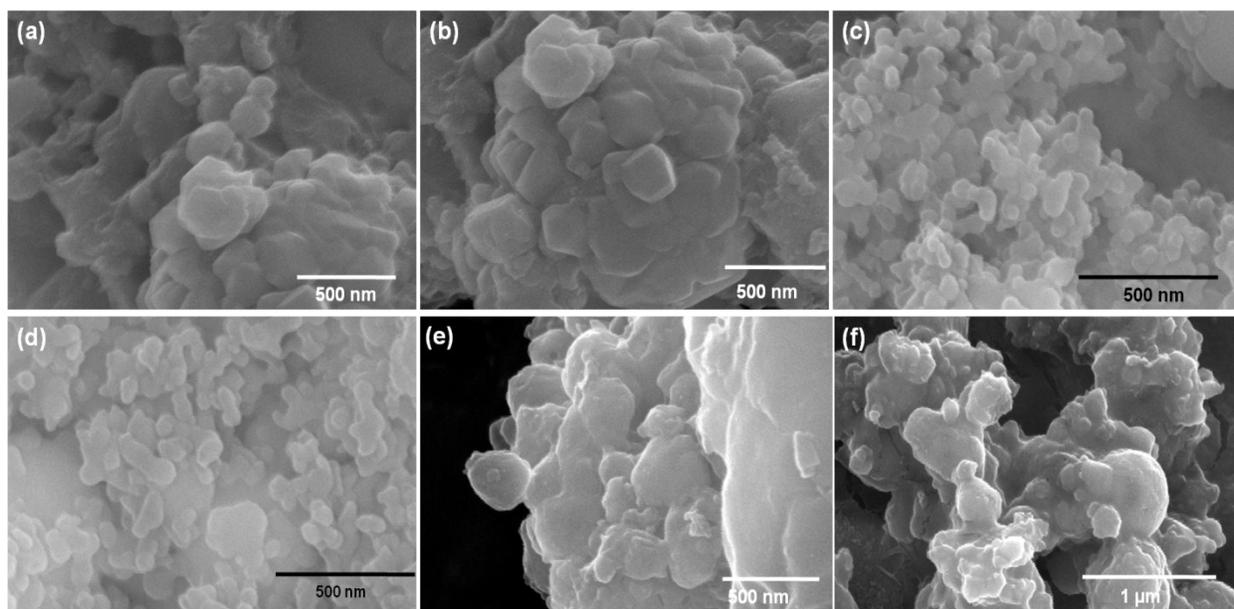
74

75
76
77
78
79
80
81
82
83
84
85
86
87
88
89
90
91
92
93
94
95
96
97
98
99
100
101
102
103
104
105



106
107
108
109
110
111
112
113
114
115
116
117
118
119

Fig. S3: FE-SEM images of heat treated LiFePO_4 (A-D) and Histogram showing particle size Distribution(E)



120 **Fig. S4.** FE-SEM images of LiFePO_4 carbon coated C-LFP-1(a-b), C-LFP-3(c-d) and C-LFP-
121 4 (e-f) using DAP process
122

123
124
125
126

127

128

129

130

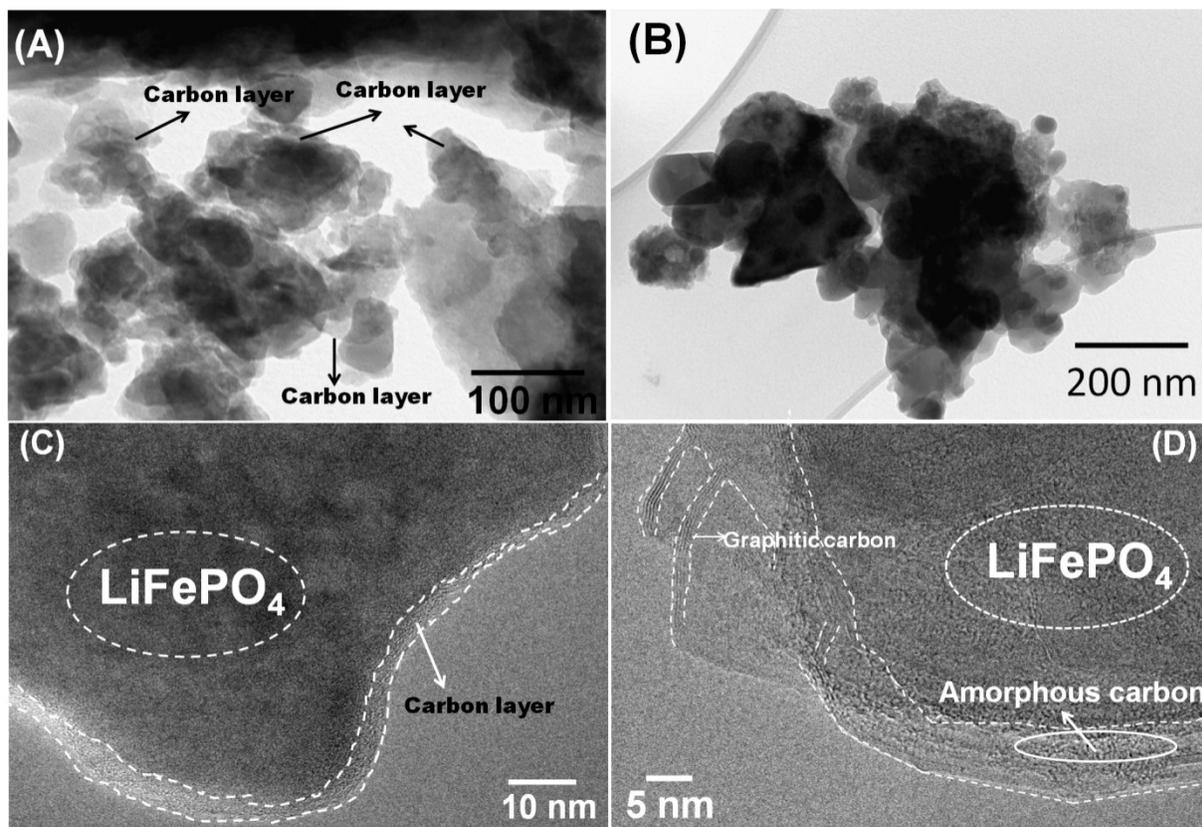
131

132

133

134

135



137

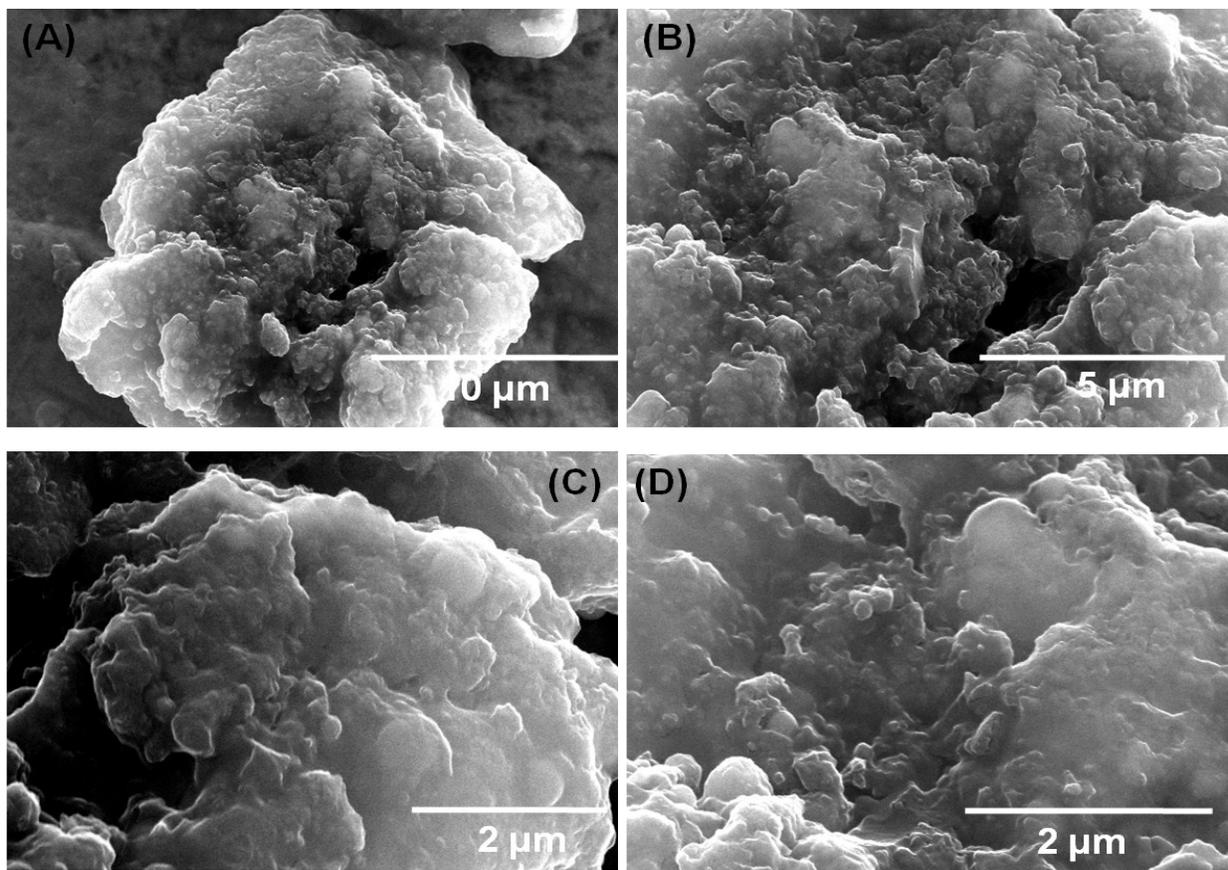
138

139

140 **Fig. S5:** HR-TEM images of C-LFP-2 showing the core shell structure with the presence of
141 graphitic and crystalline carbon on LFP.

142

143



144

145

146 **Fig. S6.** FE-SEM images of C-LFP-2 taken at low magnification (A-D)

147

148

149

150

151

152

153

154

155

156

157

158

159
160
161
162
163
164
165
166
167
168
169
170
171
172
173
174

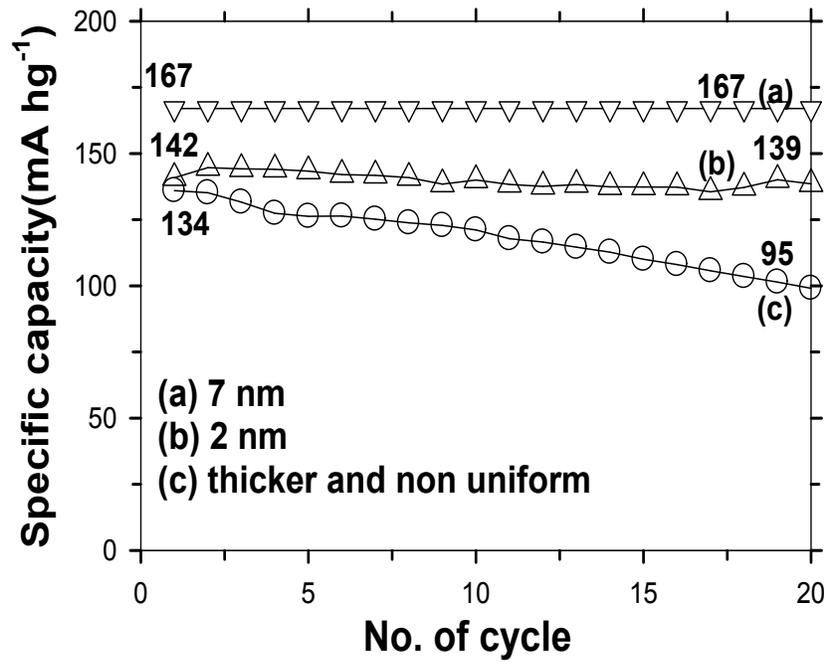
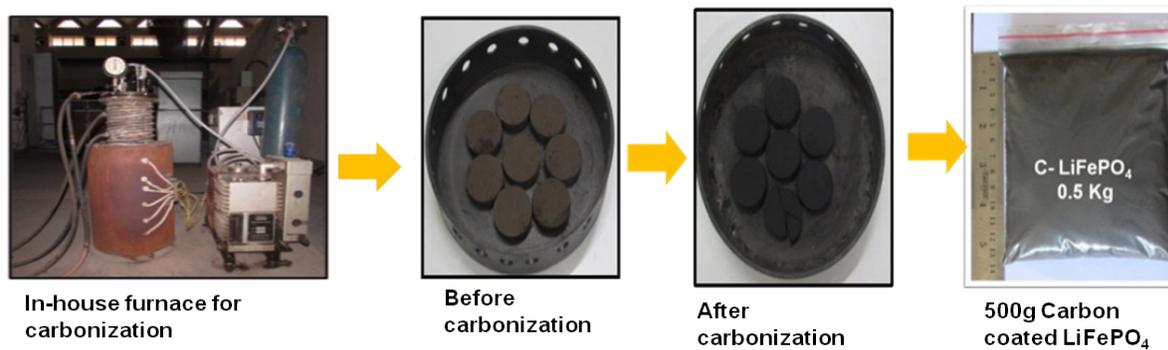


Fig. S7: Cyclic stability of DAP carbon coated LFP studied with varying thickness at 1C rate

175



176

177 **Fig. S8:** Carbonization strategy for bulk carbon coating.

178

179

180

181

182

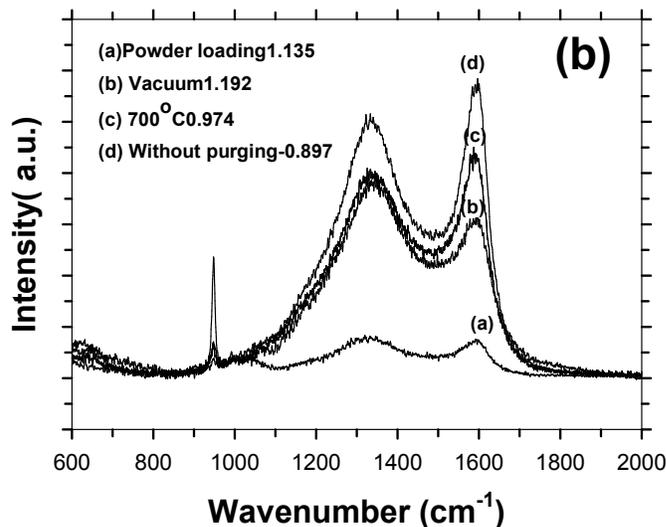
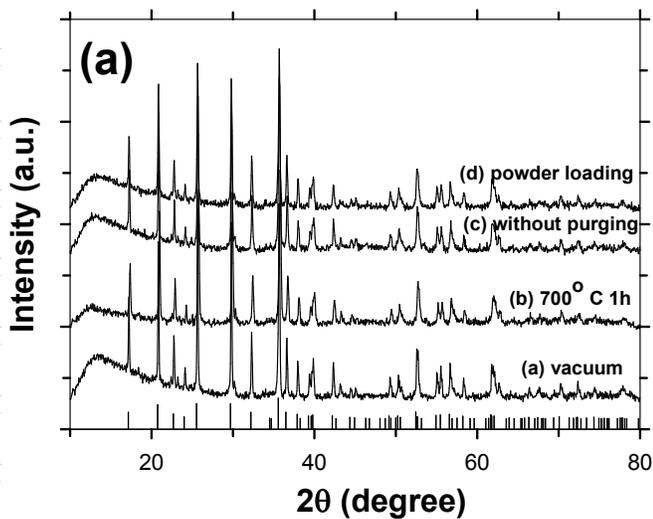
183

184

185

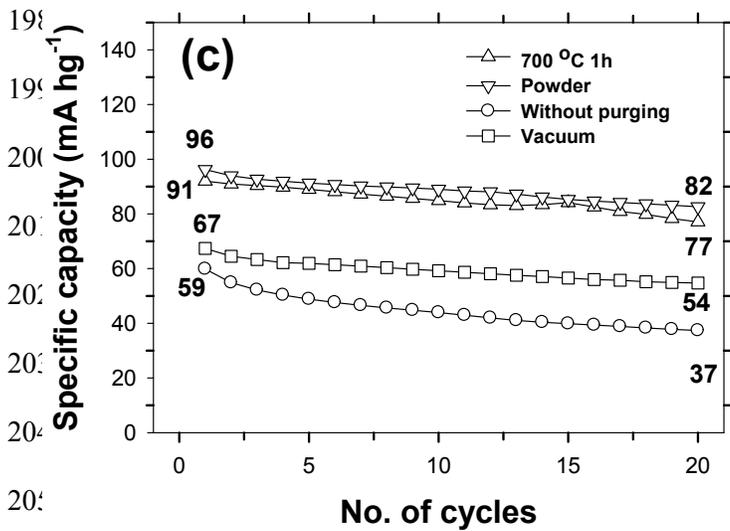
186

187



196

197



200

206

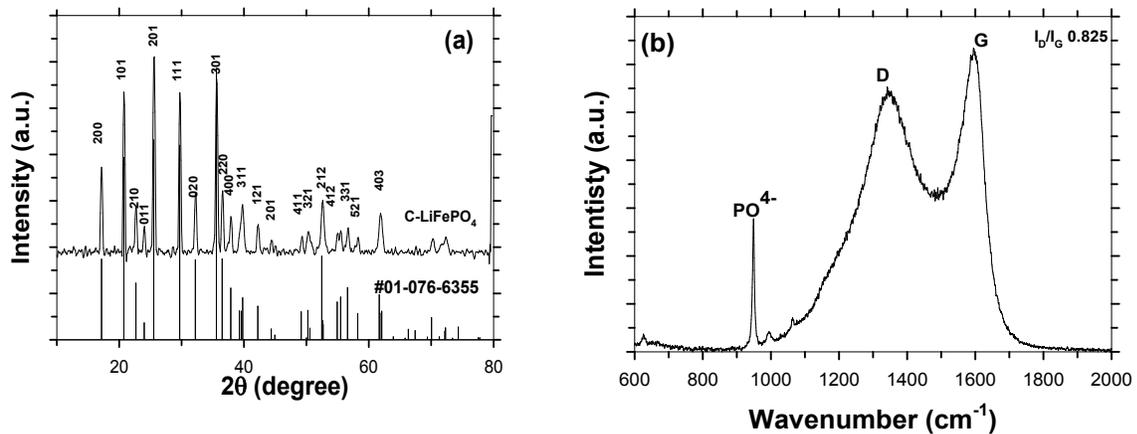
207 **Fig. S9:** Comparison of (a) X-ray diffraction analysis, (b) Raman analysis and (c) cyclic stability
 208 for optimization conditions of bulk carbon coated LiFePO₄

209

210

211

212



213

214 **Fig. S10:** (a) X-ray diffraction pattern and (b) Raman spectra of C-LFP-2B

215

216

217

218

219

220

221

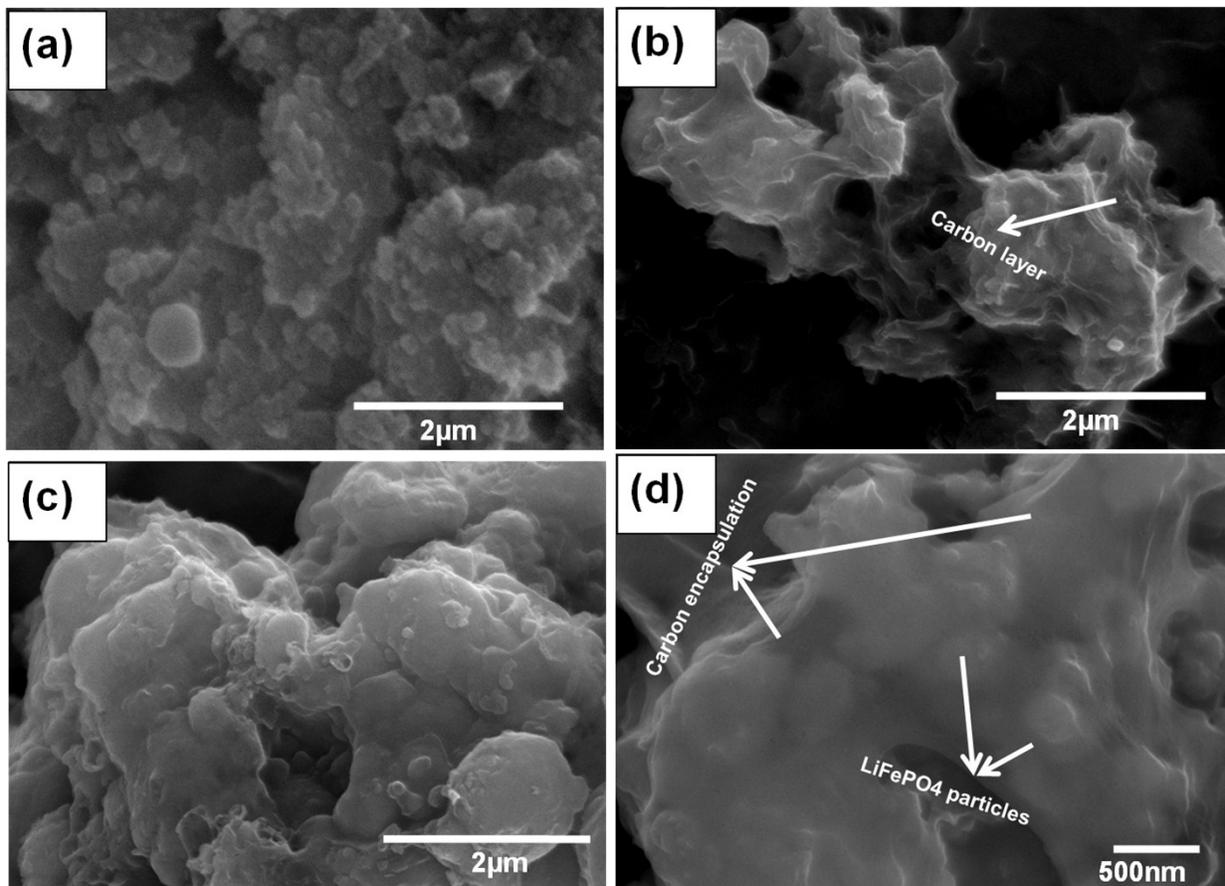
222

223

224

225

226



227

228 **Fig. S11:** FE-SEM images of C-LFP-2B

229

230

231

232

233

234

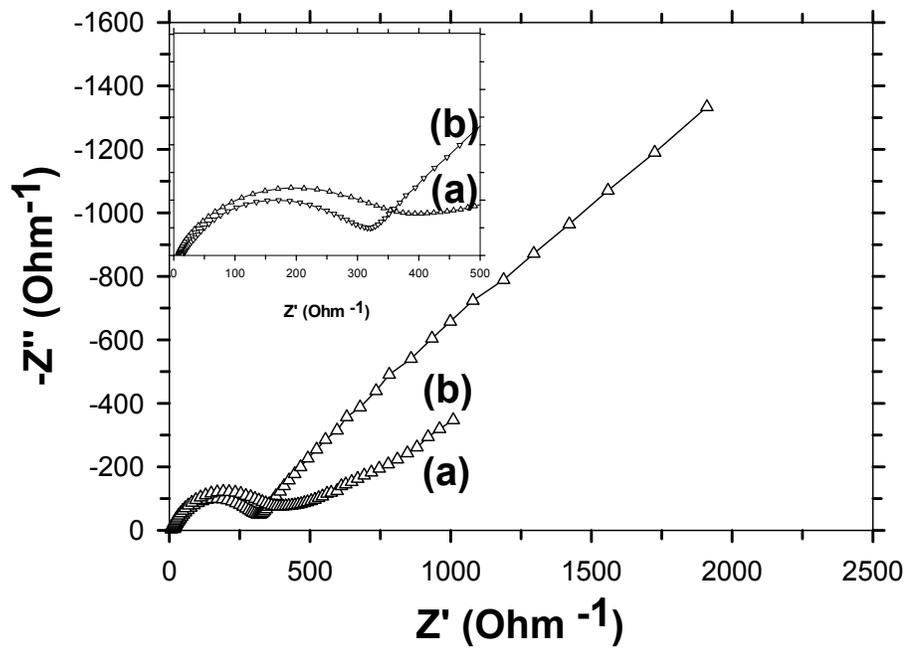
235

236

237

238

239



249 **Fig. S12:** EIS spectra of pristine LFP (a) and C-LFP-2B (b) carried out in a
 250 frequency of 0.01mHz to 100 kHz

251

252

253

254

255

256

257

258

259 Calculation of specific energy:

260 The specific energy of the full cell was calculated by the following equation as reported
261 previously.^{1,2}

$$262 E = (V * m * C) / W$$

263 Where

264 V = Nominal cell voltage (V)

265 m = Active material weight (g)

266 C = Cathode limited specific capacity (mA h g⁻¹)

267 W = Weight of the cell components (Active material weight of cathode + active material weight
268 of anode + weight of separator + weight of electrolyte)

269 The specific energy of cathode (C-LFP) developed in the present study in full cell is calculated as
270 follows:

$$271 E = (1.87 \text{ V} * 0.0067 \text{ g} * 115 \text{ mA h g}^{-1}) / (0.0787 \text{ g})$$

$$272 E = 18 \text{ W h kg}^{-1}$$

273 For full cell using commercial electrodes specific energy is calculated as

$$274 E = (1.87 \text{ V} * 0.0062 \text{ g} * 127 \text{ mA h g}^{-1}) / (0.07797 \text{ g})$$

$$275 E = 18.8 \text{ W h kg}^{-1}$$

276 References

277. H. G. Jung, M. W. Jang, J. Hassoun, Y. K. Sun and B. Scrosati, *Nat. Commun.*, 2011, **2**, 1-5.

278. B. D. McCloskey, *J. Phys. Chem. Lett.*, 2015, **6**, 4581-4588.