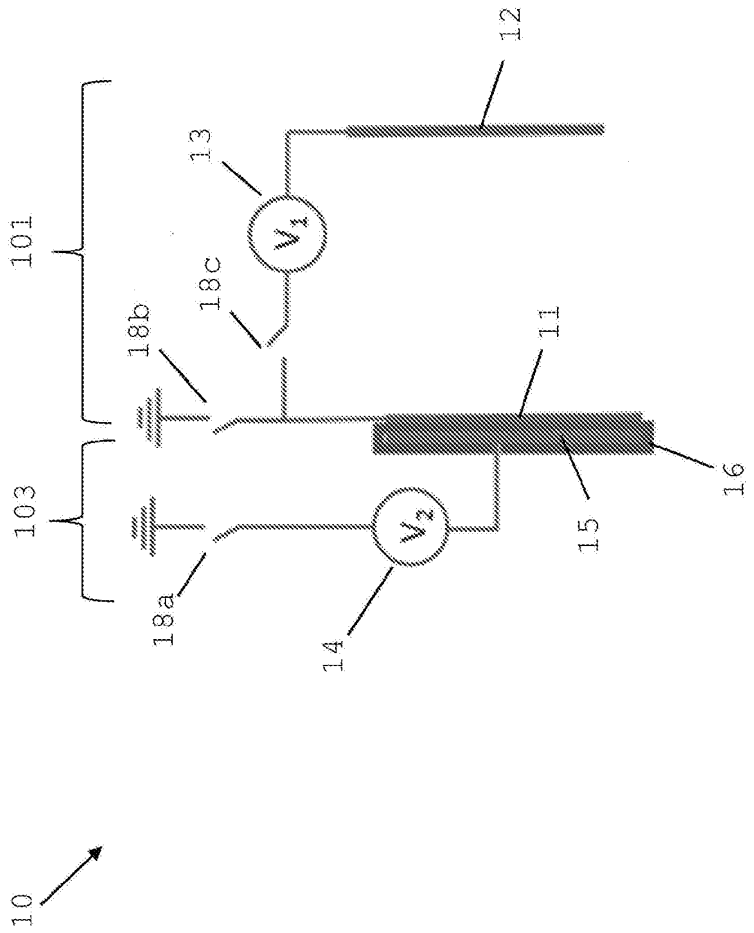

FIGURE 1

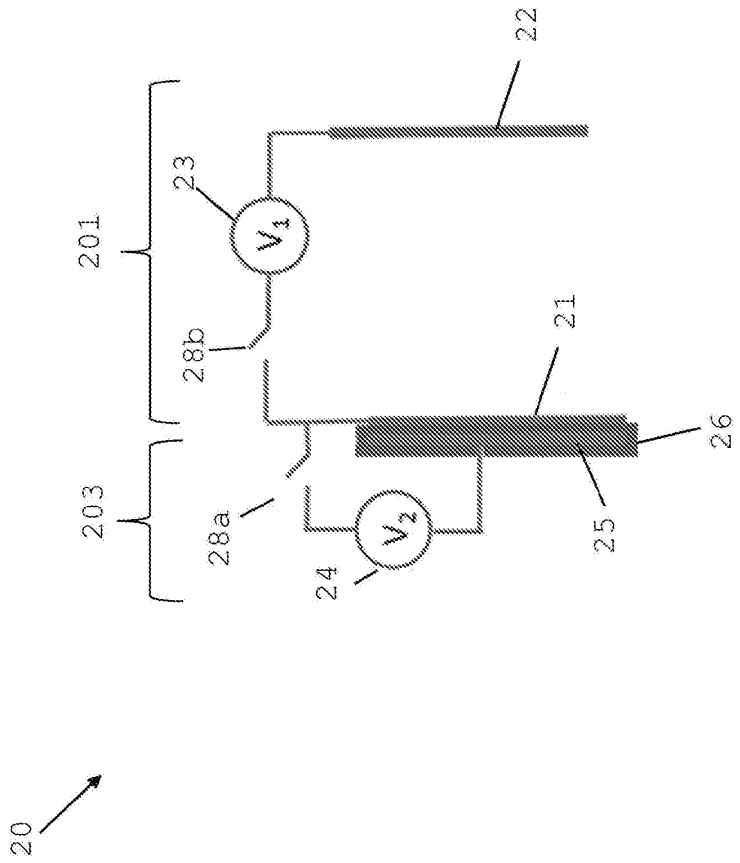

FIGURE 2

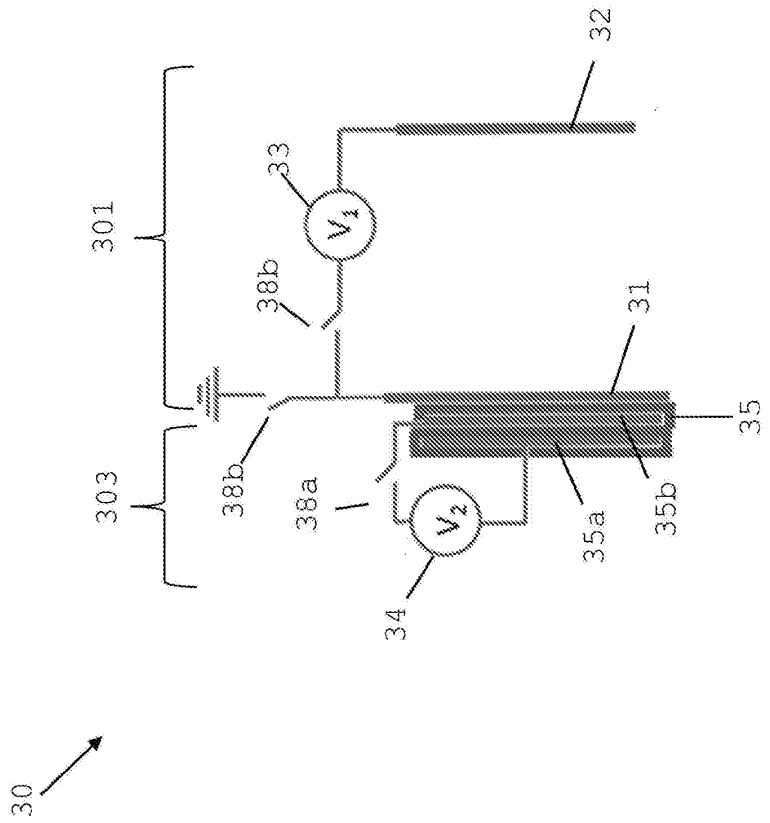

FIGURE 3

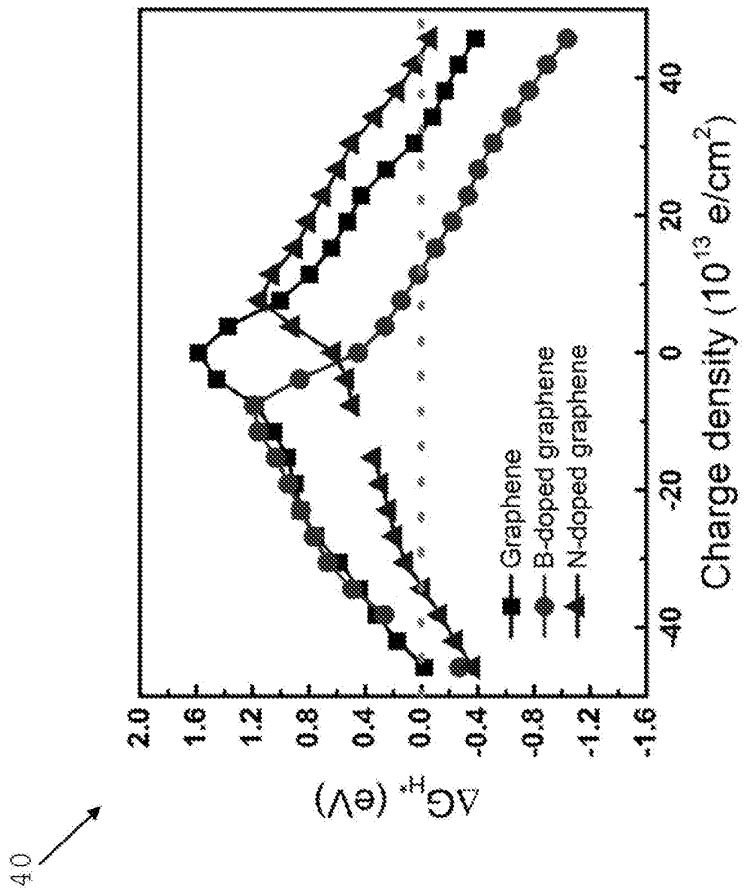
40
ΔG_{H^*} (eV)
2.0
1.6
1.2
0.8
0.4
0.0
-0.4
-0.8
-1.2
-1.6
-40
-20
0
20
40
Charge density (10^{13} e/cm^2)
Graphene
B-doped graphene
N-doped graphene

FIGURE 4

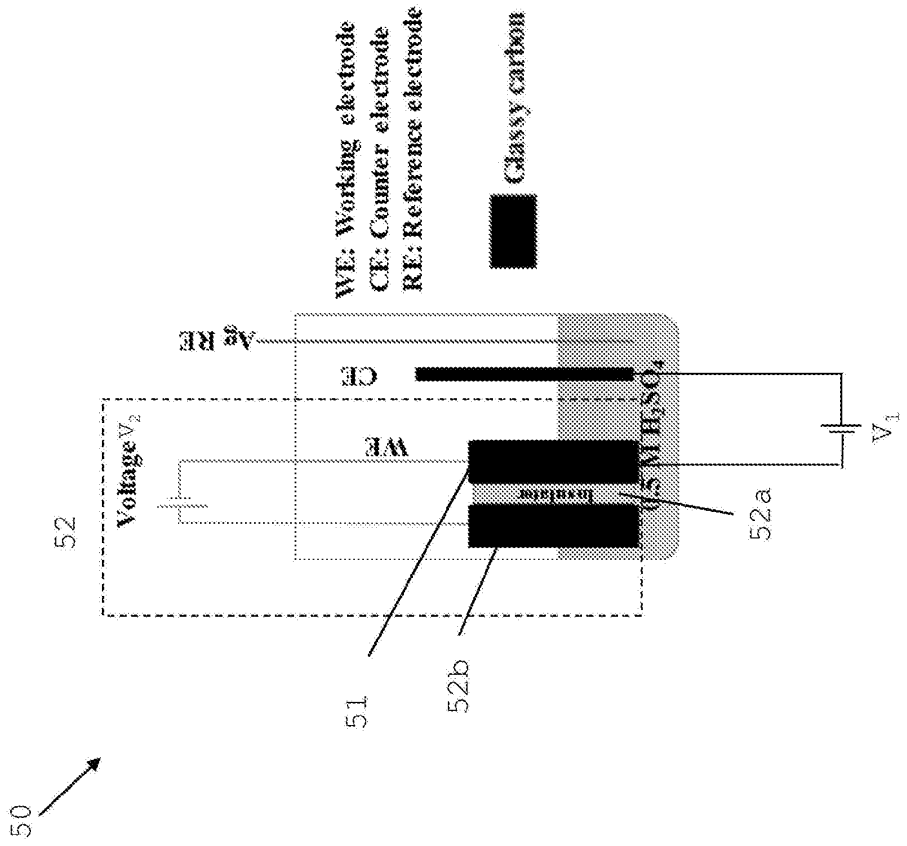
WE: Working electrode
CE: Counter electrode
RE: Reference electrode
Glassy carbon
Ag RE
CE
WE
Voltage V_2
0.5 M H_2SO_4
V_1
52
52a
51
52b
50

FIGURE 5

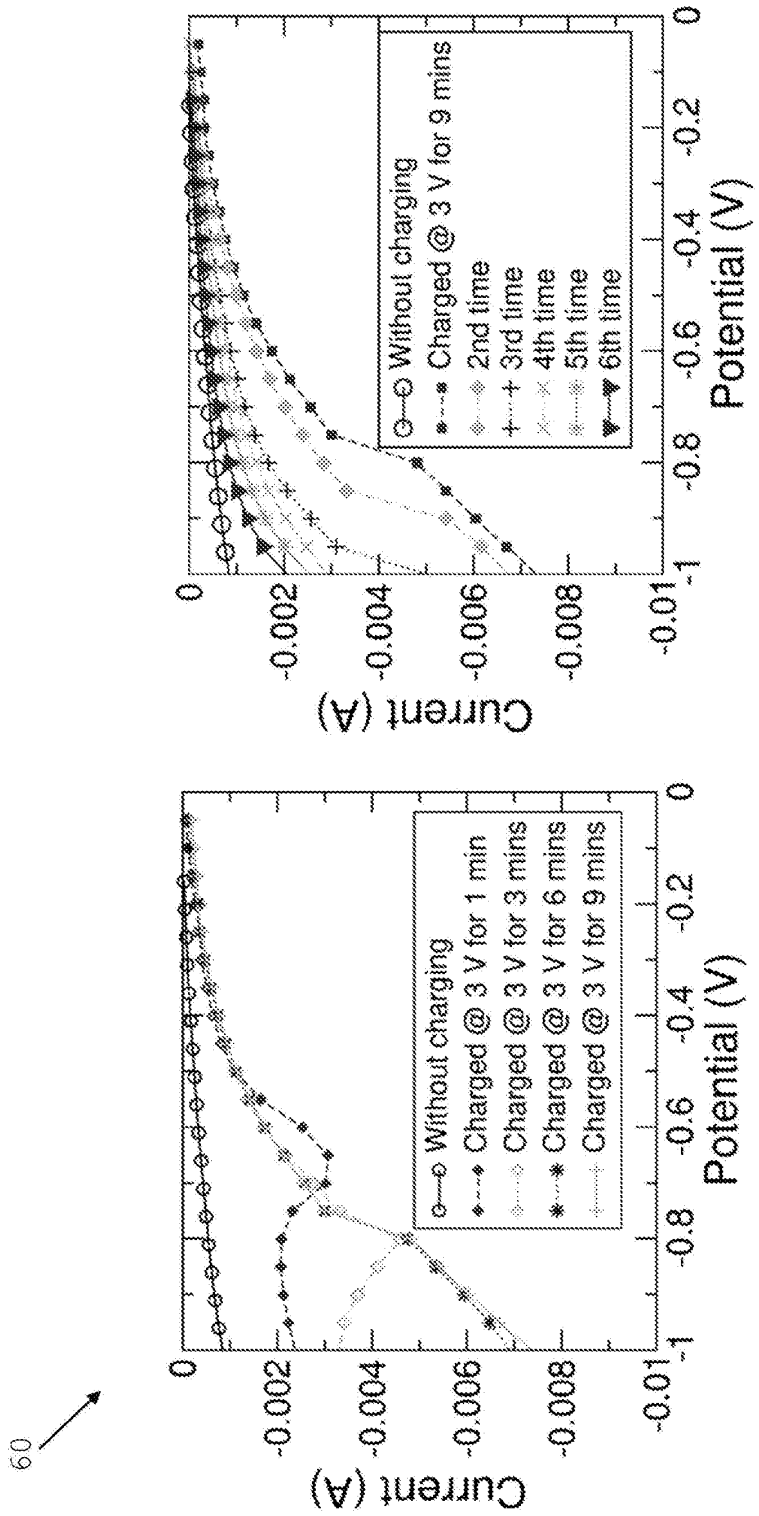

FIGURE 6(a)

FIGURE 6(b)

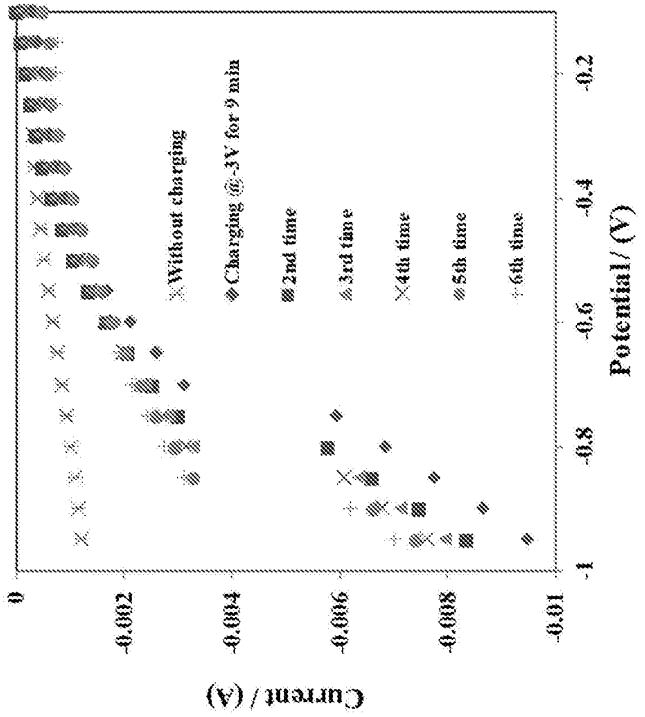

FIGURE 7(b)

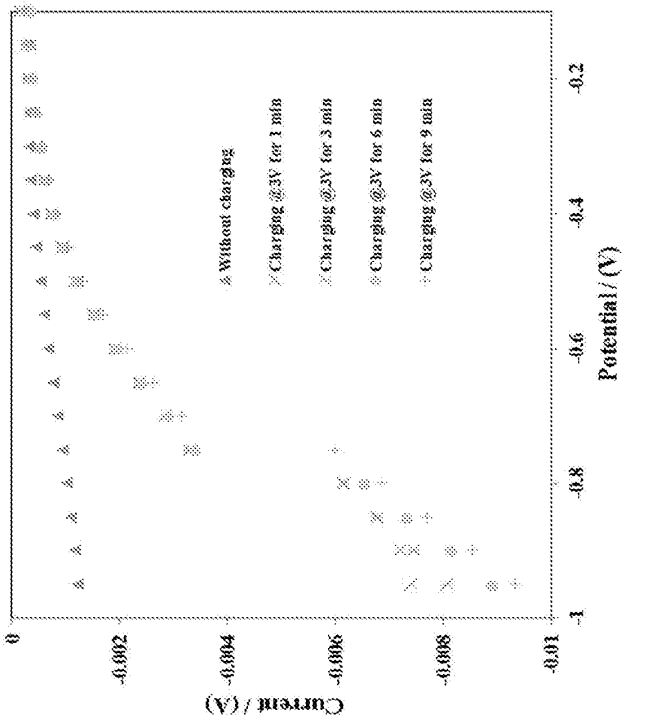

FIGURE 7(a)

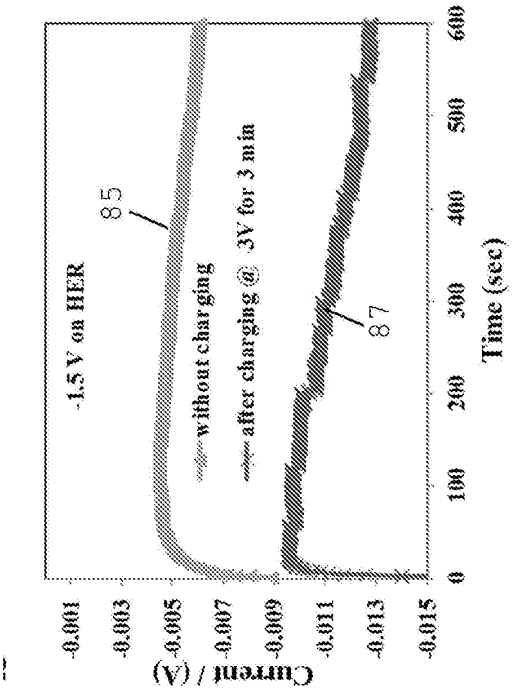

FIGURE 8(b)

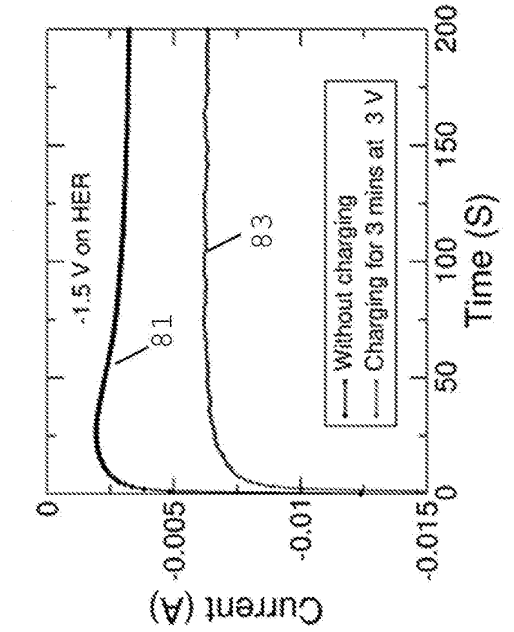

FIGURE 8(a)

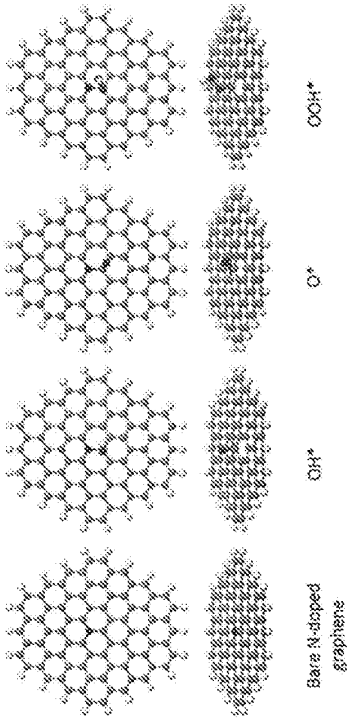

FIGURE 9 (c)

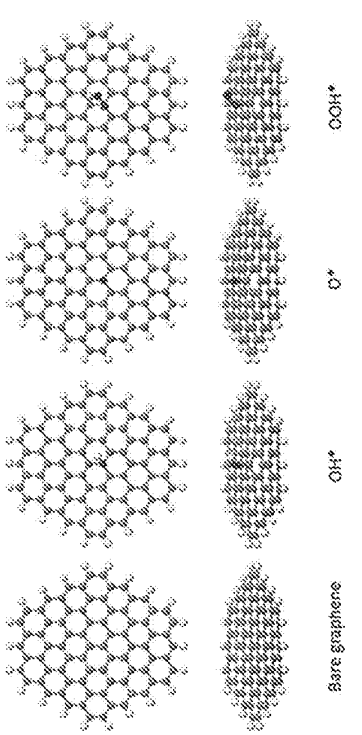

FIGURE 9(a)

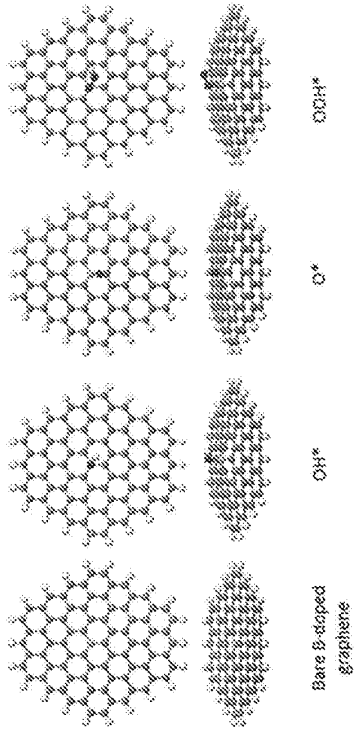

FIGURE 9(b)

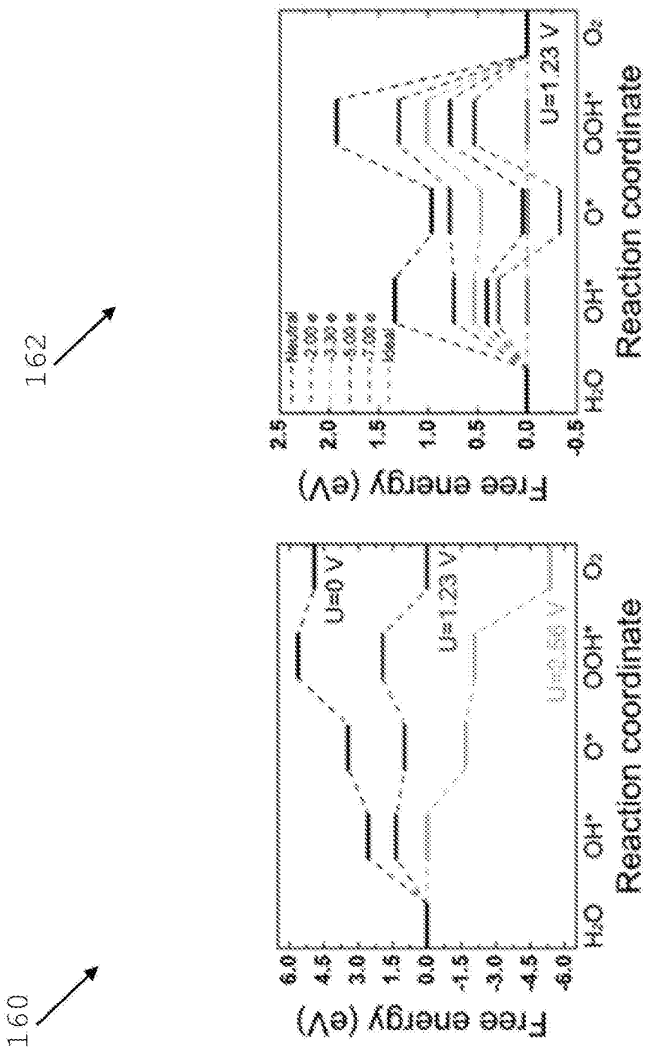

FIGURE 10(b)

FIGURE 10(a)

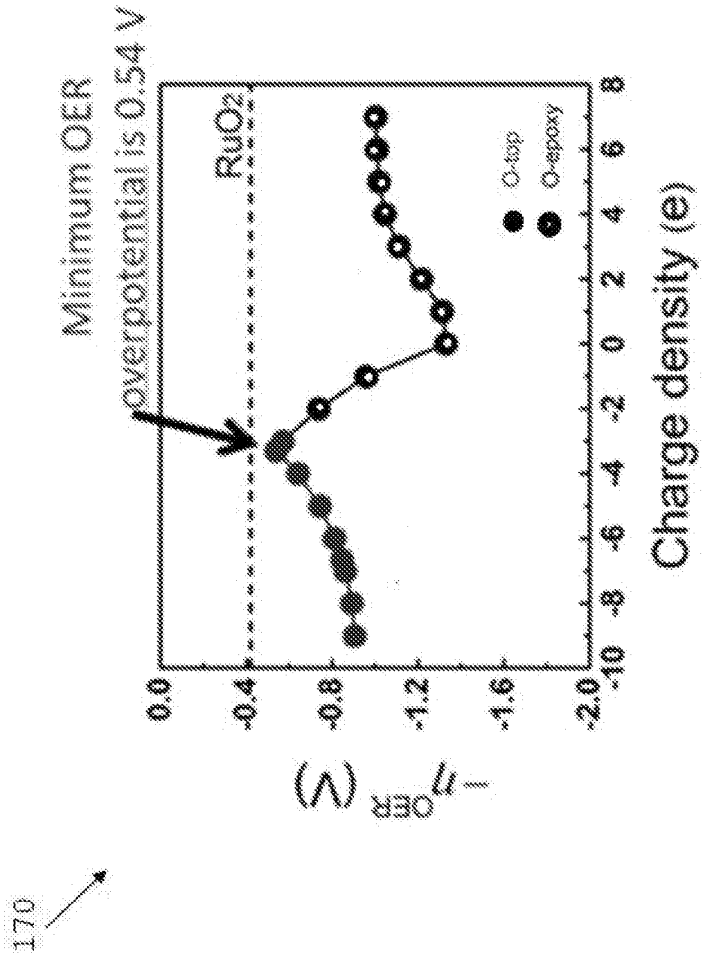
Minimum OER
overpotential is 0.54 V
RuO2
O-top
O-epoxy
Charge density (e)
$-\eta^{OER}$ (V)
0.0
-0.4
-0.8
-1.2
-1.6
-2.0
-10
-8
-6
-4
-2
0
2
4
6
8
170

FIGURE 11

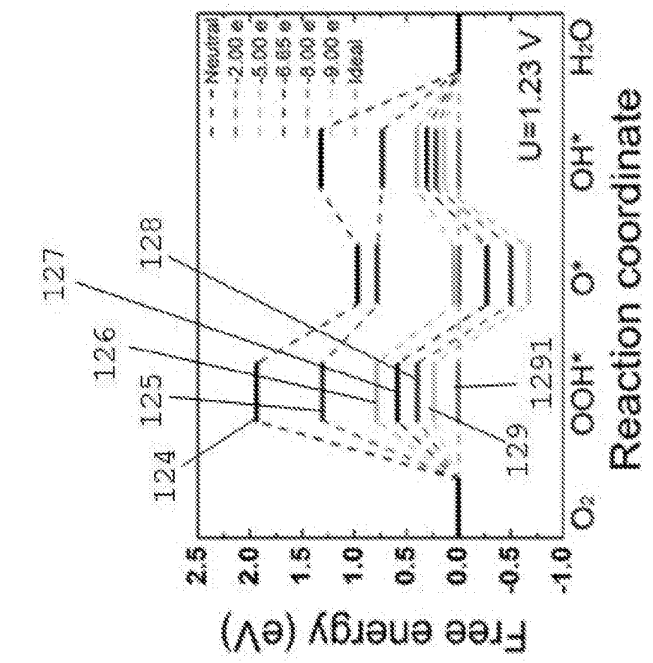

FIGURE 12(b)

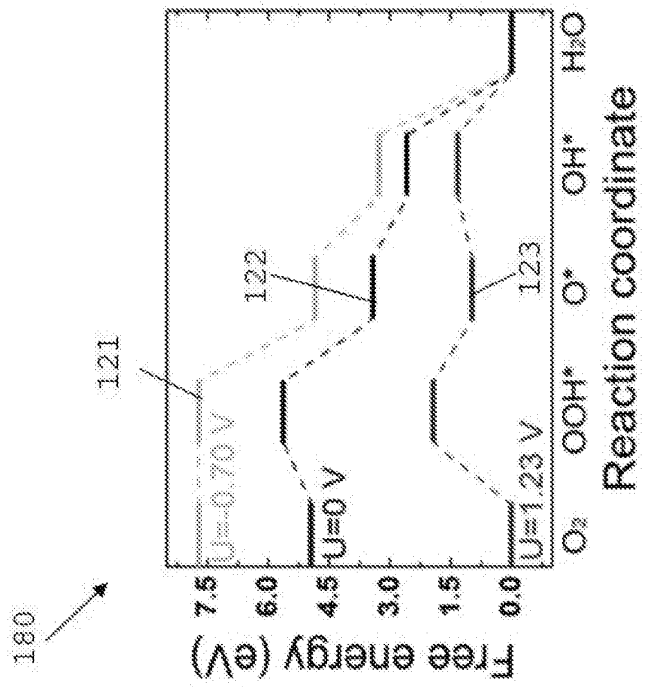

FIGURE 12(a)

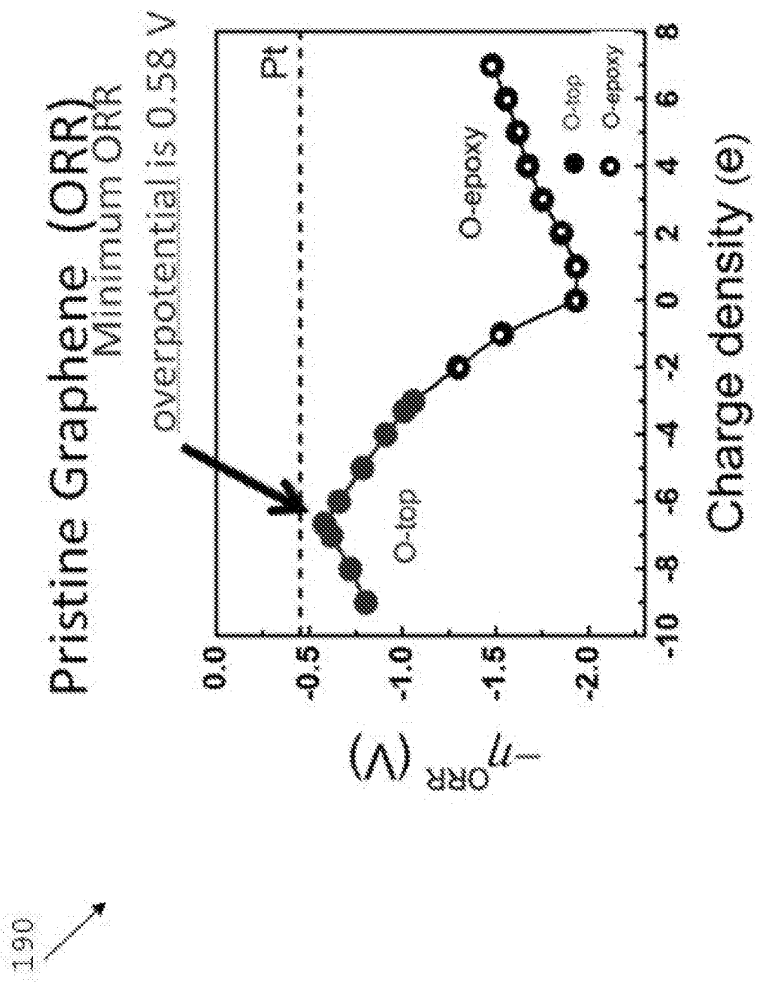
Pristine Graphene (ORR)
Minimum ORR overpotential is 0.58 V
Pt
O-top
O-epoxy
O-top
O-epoxy
$-\eta^{ORR}$ (V)
0.0
-0.5
-1.0
-1.5
-2.0
-10
-8
-6
-4
-2
0
2
4
6
8
Charge density (e)
100

FIGURE 13

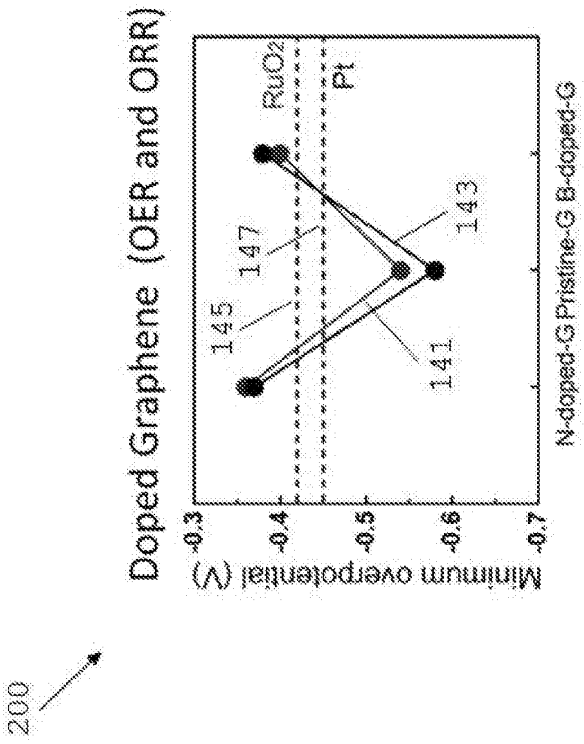
200
Doped Graphene (OER and ORR)
Minimum overpotential (V)
-0.3
-0.4
-0.5
-0.6
-0.7
RuO2
Pt
145
147
143
141
N-doped-G Pristine-G B-doped-G

FIGURE 14

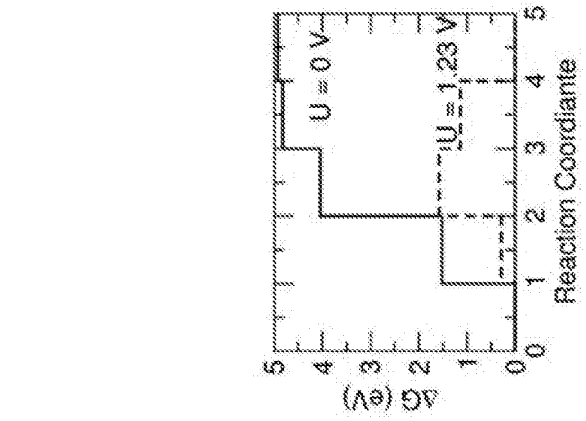

FIGURE 15

210

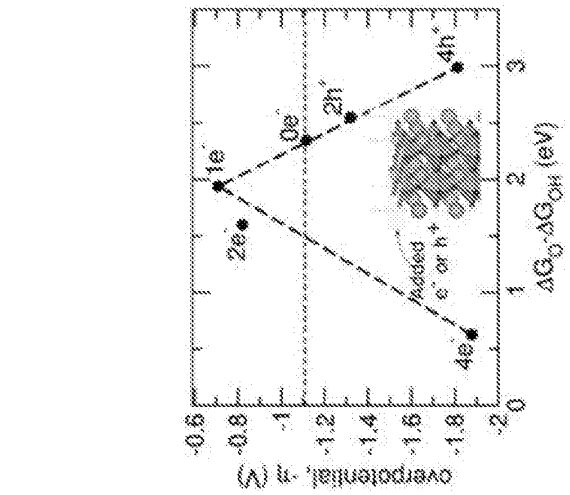
overpotential, -η (V)
-0.6
-0.8
-1
-1.2
-1.4
-1.6
-1.8
-2.0
0
1
2
3
ΔG_O-ΔG_{OH} (eV)
1e⁻
2e⁻
0e⁻
2h⁺
4h⁺
4e⁻
Added e⁻ or h⁺
220

FIGURE 16

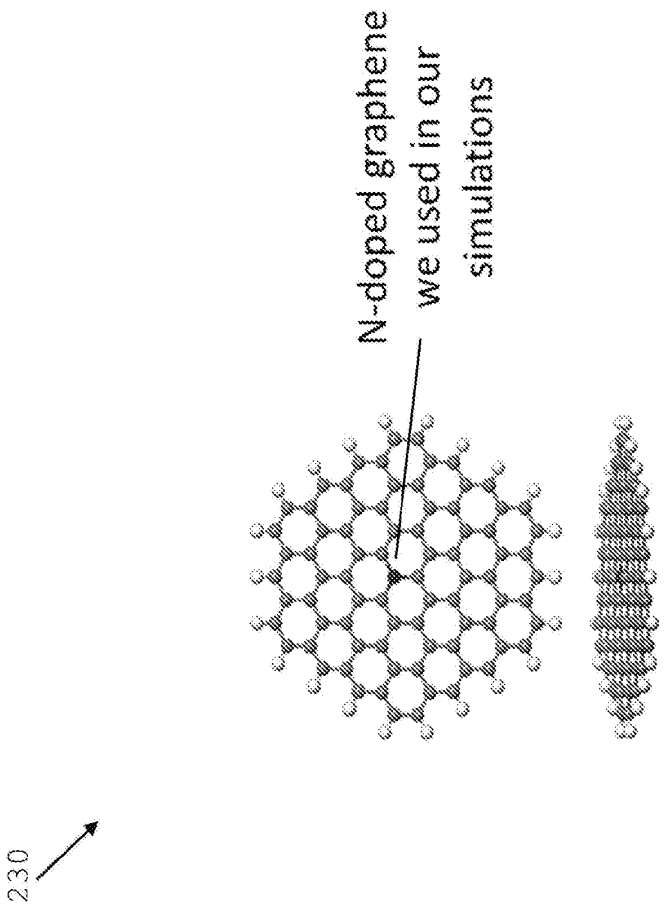

FIGURE 17

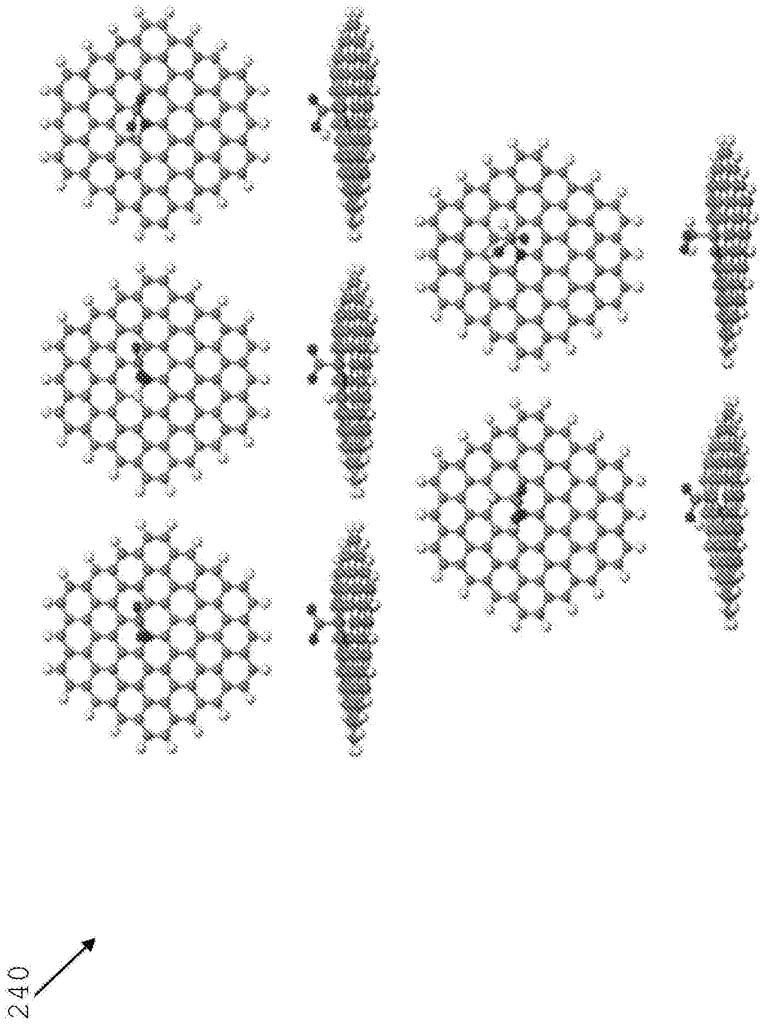

FIGURE 18

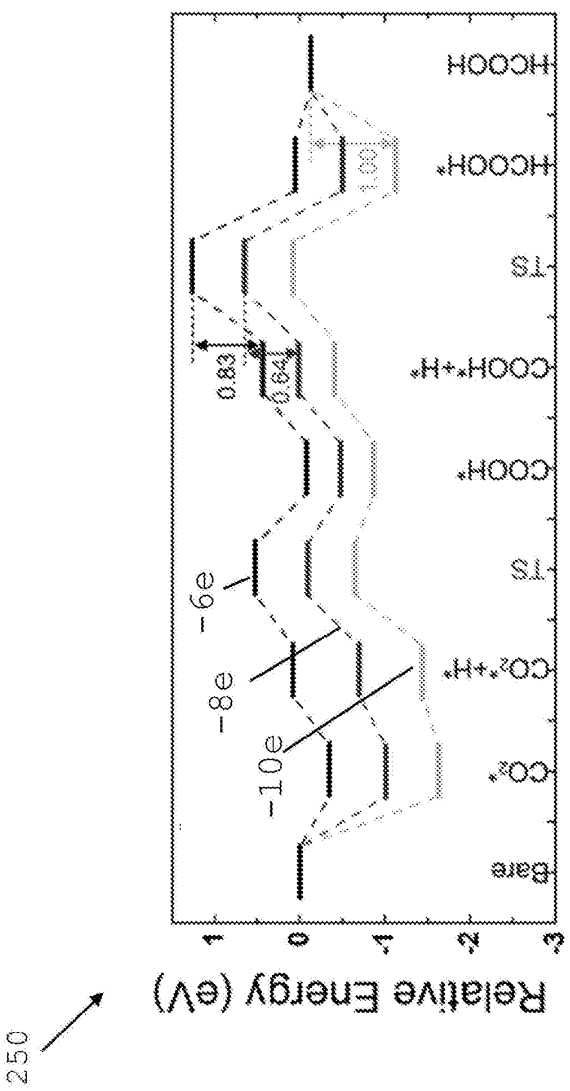
250
Relative Energy (eV)
1
0
-1
-2
-3
Bare
CO2*
CO2*+H*
TS
COOH*
COOH*+H*
TS
HCOOH*
HCOOH
-6e
-8e
-10e
0.83
0.64
1.00
Reaction Pathway
Largest barriers of CO2 reduction at different charge densities are shown
Small charge density (N=-6 e): weak adsorption of intermediates and large reaction barrier (0.83 eV)
Mediate charge density (N=-8 e): CO2 reduction to formate acid with small barrier (0.64 eV)
Large charge density (N=-10 e): strong adsorption of intermediates and large desorption barrier (1.00 eV)

FIGURE 19