

# Galvanic effects induced by siderite and cementite surface layers on carbon steel in aqueous CO<sub>2</sub> environments

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## SUPPLEMENTARY MATERIAL

A light microscope image of the etched X65 carbon steel surface is shown in **Figure S1**.

Nyquist plots used to determine solution resistance for all conditions are shown in **Figure S2**.

OCP measurements during Fe<sub>3</sub>C and FeCO<sub>3</sub> layer formation experiments (Method I and Method II), measured in between LPR measurements shown in Figure 3 in the main manuscript, are shown in **Figure S3**.

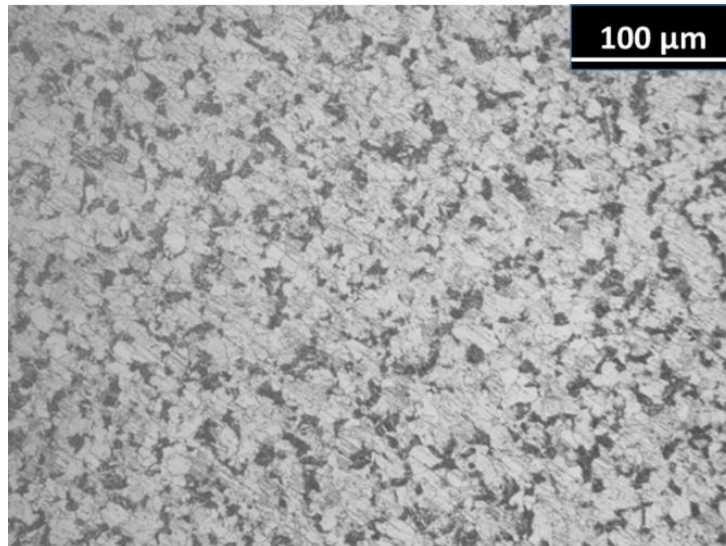
Potentiodynamic polarisation measurements after Fe<sub>3</sub>C layer formation experiments (Method I), shown in Figure 3 in the main manuscript, are shown in **Figure S4**.

OCP measurements in a CO<sub>2</sub>-saturated, 1 wt.% NaCl, pH 5 solution at 50 °C of a bare carbon steel, an Fe<sub>3</sub>C-layered X65 carbon steel coupon and an FeCO<sub>3</sub>-layered X65 carbon steel coupon (Method I), shown in Figure 6 in the main manuscript, are shown in **Figure S5**.

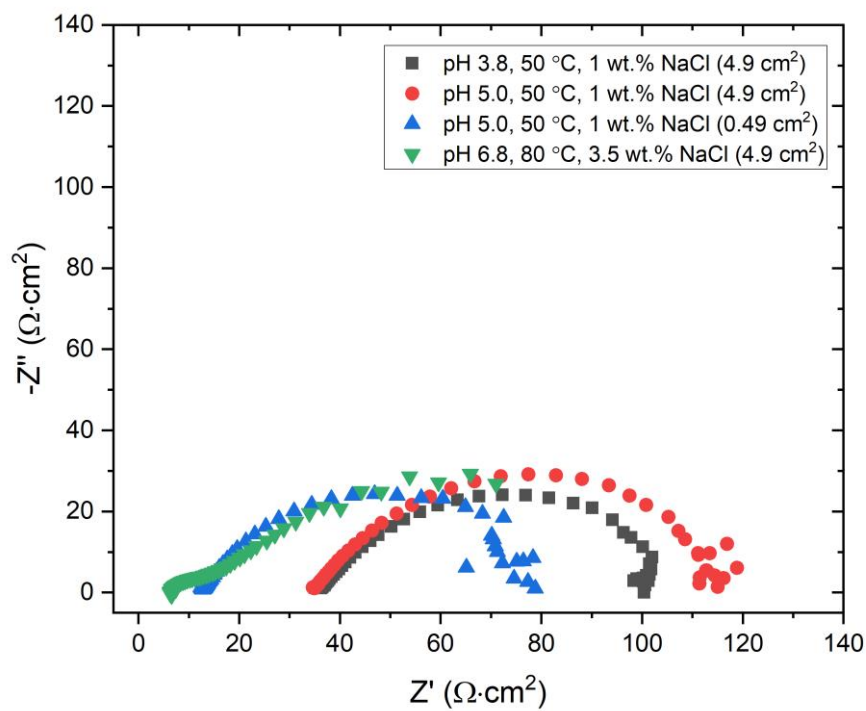
Potentiodynamic polarisation measurements after corrosion experiments in a CO<sub>2</sub>-saturated, 1 wt.% NaCl, pH 5 solution at 50 °C (Method IV), shown in Figure 6 in the main manuscript, are shown in **Figure S6**.

SEM images after galvanic corrosion in a CO<sub>2</sub>-saturated, 1 wt.% NaCl, pH 5 solution at 50 °C of bare carbon steel coupled to Fe<sub>3</sub>C-layered X65 carbon steel coupons are shown in **Figure S7**.

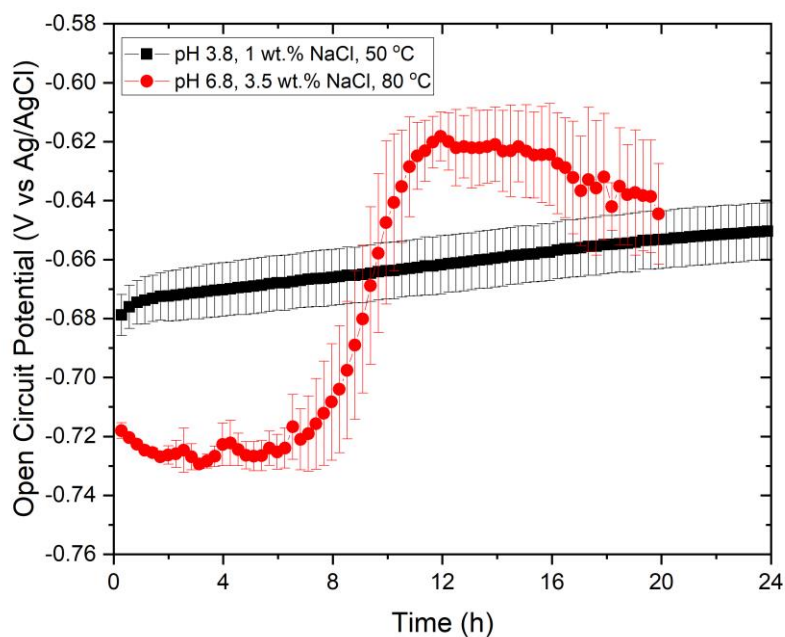
FIB-SEM images after galvanic corrosion in a CO<sub>2</sub>-saturated, 1 wt.% NaCl, pH 5 solution at 50 °C of bare carbon steel coupled to Fe<sub>3</sub>C-layered X65 carbon steel coupons are shown in **Figure S8**.



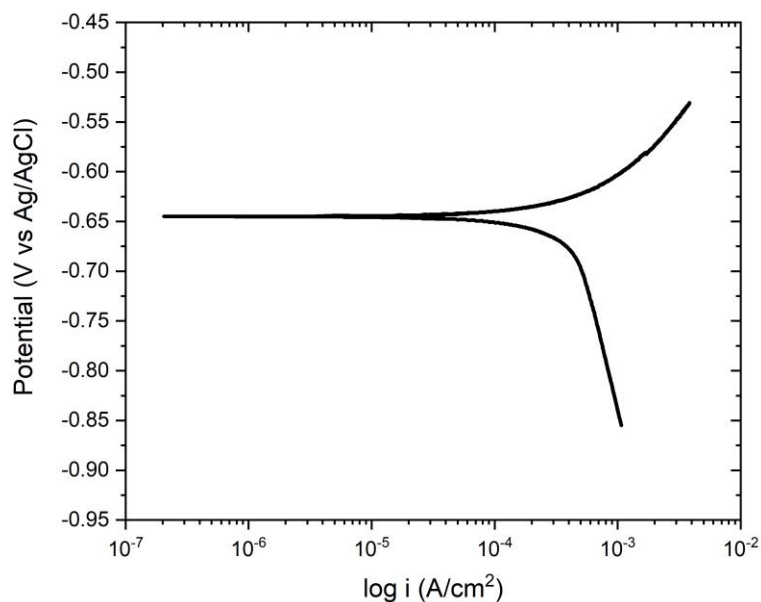
**Figure S1:** Microscope image of X65 carbon steel after etching with 2% Nital solution, showing a microstructure consisting of ferrite (light grey) and pearlite (dark grey) phases



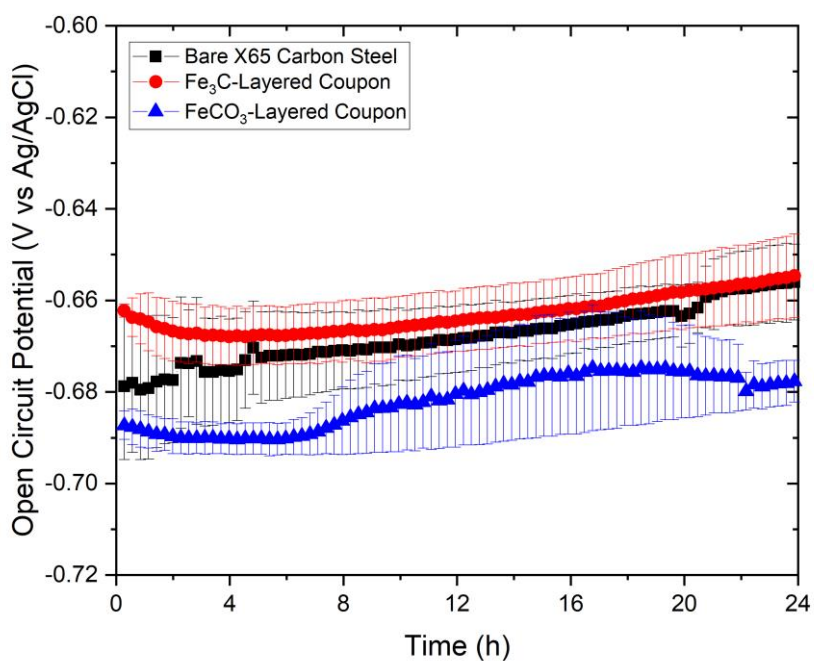
**Figure S2:** Nyquist plot to determine solution resistance ( $R_s$ ) in  $\text{CO}_2$ -saturated conditions at different pH, wt.% NaCl and temperatures to reflect the different conditions employed throughout the study



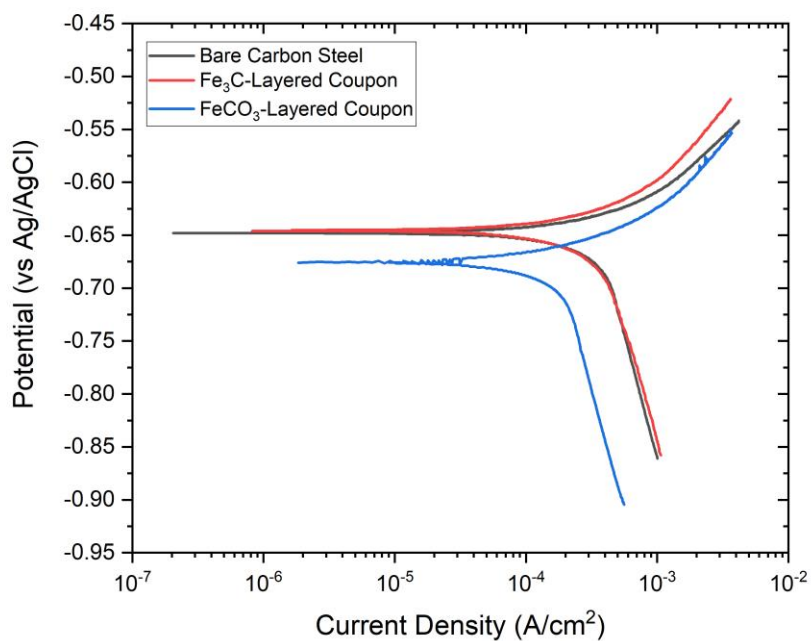
**Figure S3:** OCP measurements of  $\text{Fe}_3\text{C}$  layers on X65 steel (Method I) in  $\text{CO}_2$ -saturated, pH 3.8, 1 wt.% NaCl conditions at 50 °C and  $\text{FeCO}_3$  layers on X65 steel (Method II) in  $\text{CO}_2$ -saturated, pH 6.8, 3.5 wt.% NaCl conditions at 80 °C



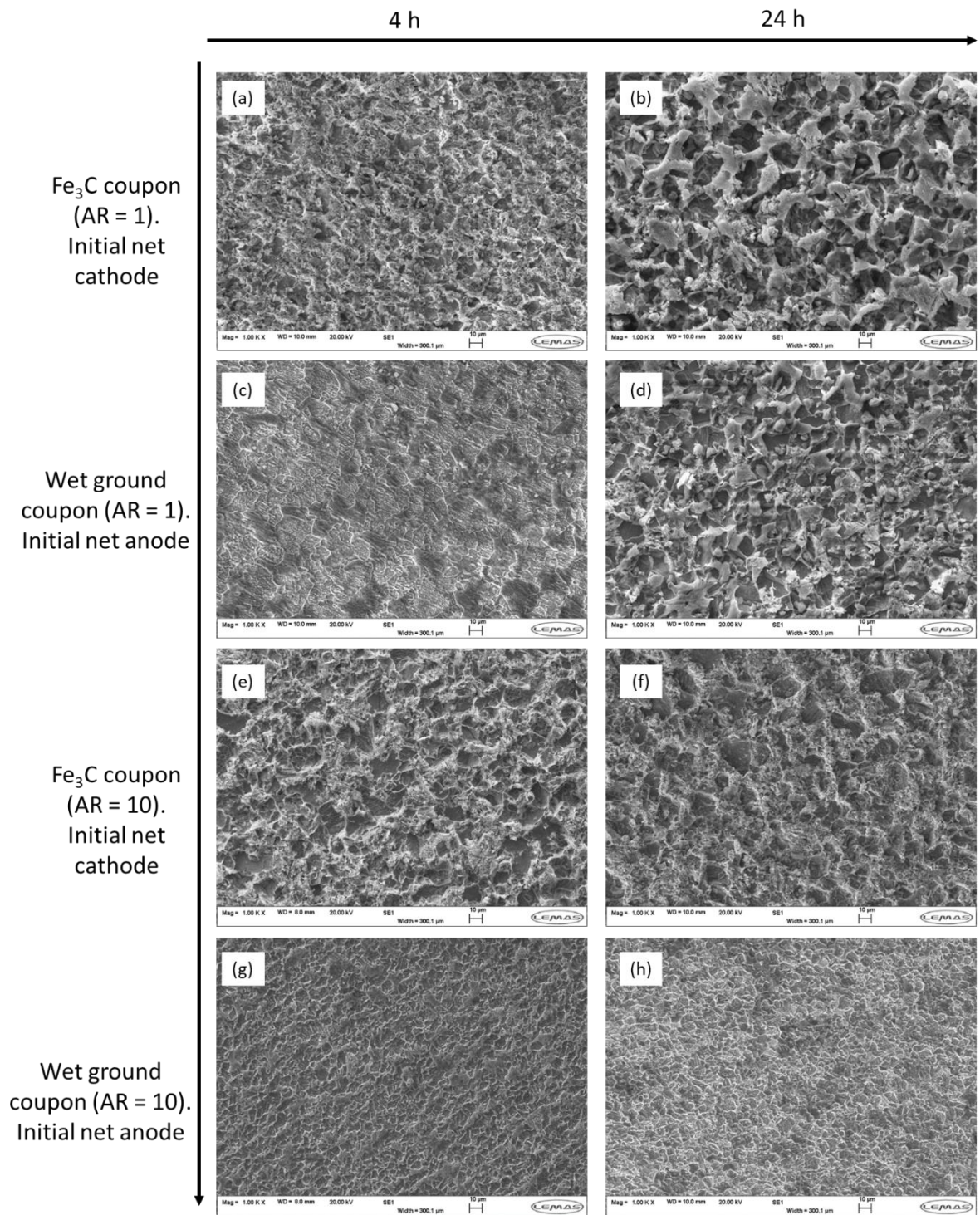
**Figure S4:** Potentiodynamic polarisation measurements of  $\text{Fe}_3\text{C}$  layers on X65 steel (Method I) in  $\text{CO}_2$ -saturated, pH 3.8, 1 wt.% NaCl conditions at 50 °C after 24 h of exposure



**Figure S5:** OCP measurements of bare X65 carbon steel and layered coupons in a CO<sub>2</sub>-saturated, pH 5, 1 wt.% NaCl solution at 50 °C

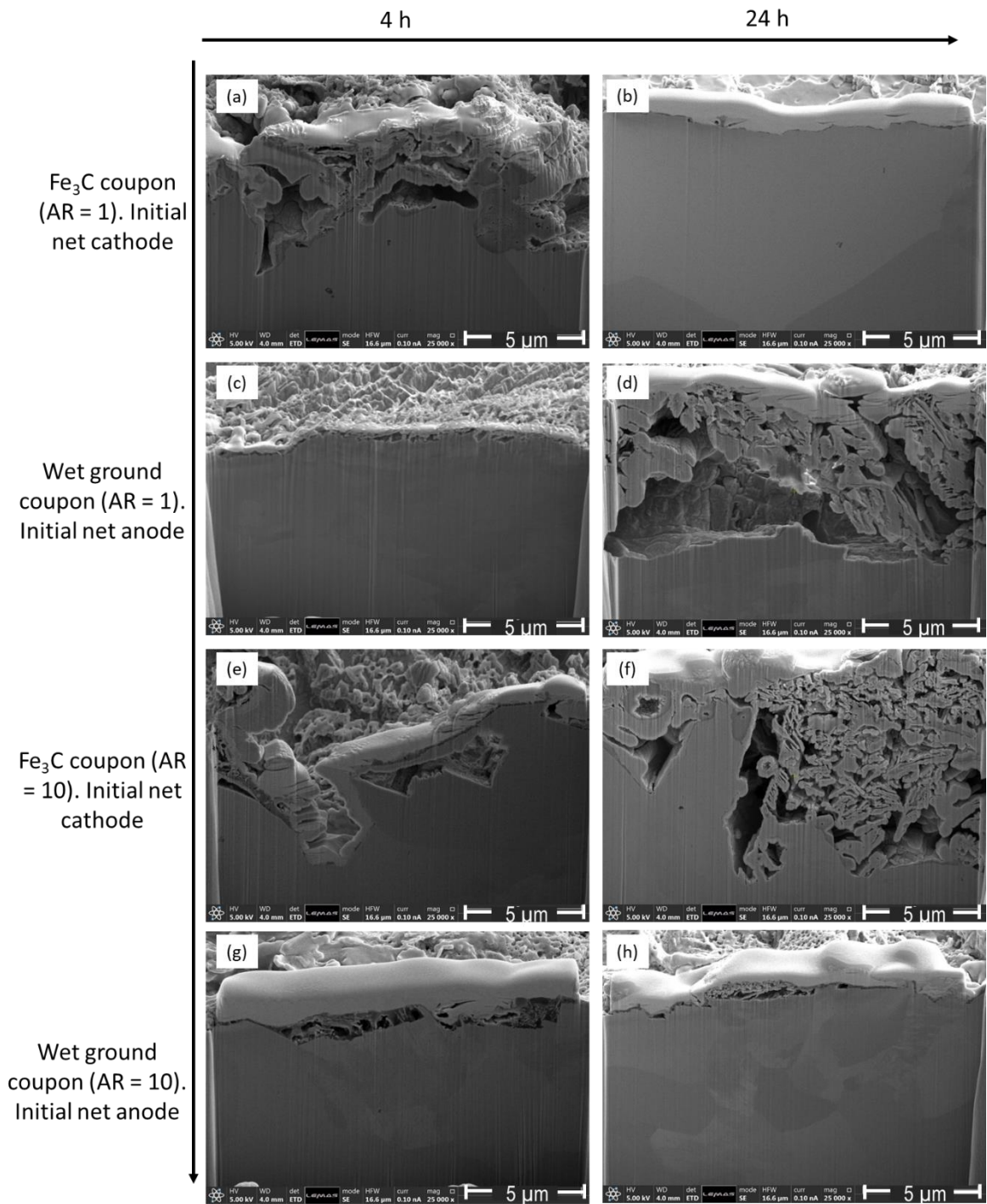


**Figure S6:** Potentiodynamic polarisation measurements of bare X65 carbon steel, Fe<sub>3</sub>C-layered coupons and FeCO<sub>3</sub>-layered coupons in CO<sub>2</sub>-saturated, pH 5, 1 wt.% NaCl conditions at 50 °C after 24 h of exposure



**Figure S7:** SEM images of X65 carbon steel coupons after ZRA measurements completed at AR = 1 (a-d) and AR = 10 (e-h) over 4 h (a, c, e and g) and 24 h (b, d, f and h) in a  $\text{CO}_2$ -saturated, pH 5, 1 wt.% NaCl solution at 50 °C by coupling a wet ground X65 carbon steel coupon to an  $\text{Fe}_3\text{C}$ -layered X65 carbon steel coupon





**Figure S8:** SEM images obtained after FIB milling to show a cross section of the surface layers formed after ZRA measurements completed at an AR = 1 (a-d) and AR = 10 (e-h) over 4 h (a, c, e and g) and 24 h (b, d, f and h) in a  $\text{CO}_2$ -saturated, pH 5, 1 wt.% NaCl solution at 50 °C by coupling a wet ground X65 carbon steel coupon to an  $\text{Fe}_3\text{C}$ -layered X65 carbon steel coupon