Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supporting Information

Engineering the surface of titanium to improve its bioactivity and antibacterial activity through a multi-functional coating approach

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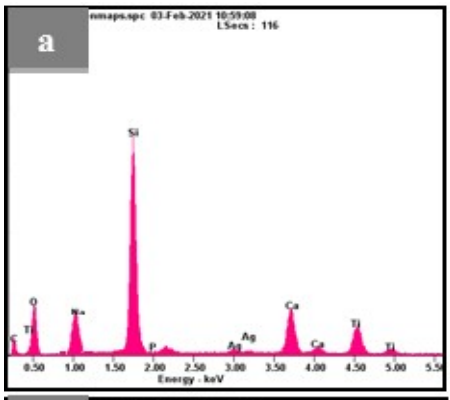
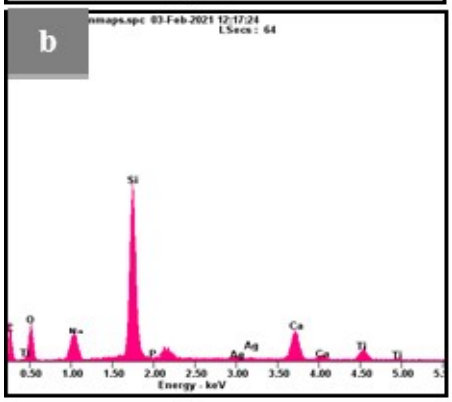
Synthesis of graphene oxide (GO), reduced graphene oxide (rGO) and silver functionalized reduced graphene oxide (Ag-rGO)

Graphite oxide was synthesized by modified Hummers' method with some improvements. Briefly, 1 g of graphite powder (particle diameter ≤ 40 mm, Sigma Aldrich), 0.75 g of $NaNO_3$ and 69 mL of H_2SO_4 were mixed in a beaker and kept in ice bath (0 °C). Then, 5 g of $KMnO_4$ was added to this mixture in a stepwise manner. After the complete addition of $KMnO_4$, the mixture was stirred at 35 °C for 24 h to ensure the complete oxidation of graphite. Next, 100 mL of water was added to the brownish grey paste and the temperature was kept at 95 °C for 15 min. Finally, 175 mL of deionized water and 10 mL of 30 % H_2O_2 were added to reduce the residual permanganate and the suspension turned to yellowish brown in colour. The solution was allowed to settle down overnight and washed with 5 % HCl and deionized water, respectively, until the pH of the solution becomes neutral. The product was separated using centrifugation at 4000 rpm and the collected product was dried at 60 °C for 24 h in a vacuum oven to finally get graphite oxide (GO).

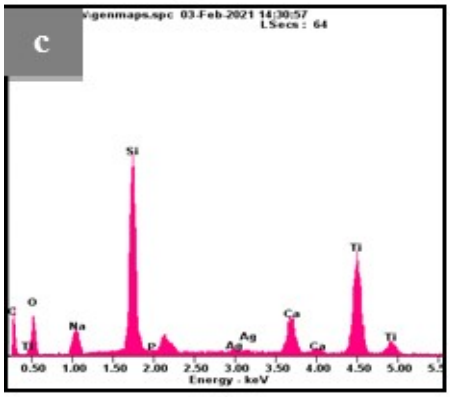
To synthesize silver functionalized reduced graphene oxide (Ag-rGO), GO (0.5 wt. %) was dispersed in deionized water. Further, 0.25 wt. % of $AgNO_3$ was added to the GO dispersion and the mixture was stirred well followed by sonication for 1 h. Subsequently, 5 ml of 50-60% hydrazine hydrate was added and the mixture was refluxed at 70 °C for 6 h. The AgrGO was filtered, washed with deionized water and dried at 60 °C for 12 h in a vacuum oven. For preparing pure rGO, the same procedure was performed in the absence of $AgNO_3$.



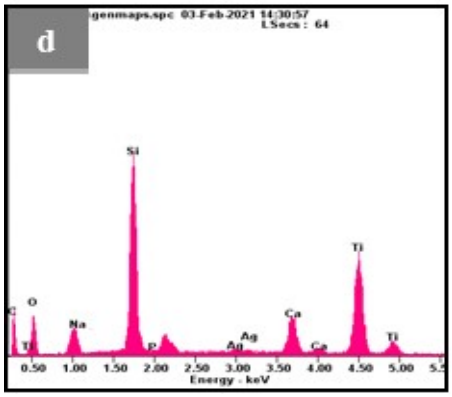
Element	At %
C K	33.75
O K	34.89
Na K	08.91
Si K	11.07
P K	01.01
Ag L	01.03
Ca K	07.51
Ti K	01.83
Total	100.00



Element	At %
C K	35.44
O K	36.29
Na K	07.30
Si K	10.52
P K	00.89
Ag L	00.93
Ca K	06.87
Ti K	01.77
Total	100.00



Element	At %
C K	37.87
O K	34.22
Na K	07.53
Si K	10.08
P K	00.96
Ag L	00.85
Ca K	06.34
Ti K	02.15
Total	100.00

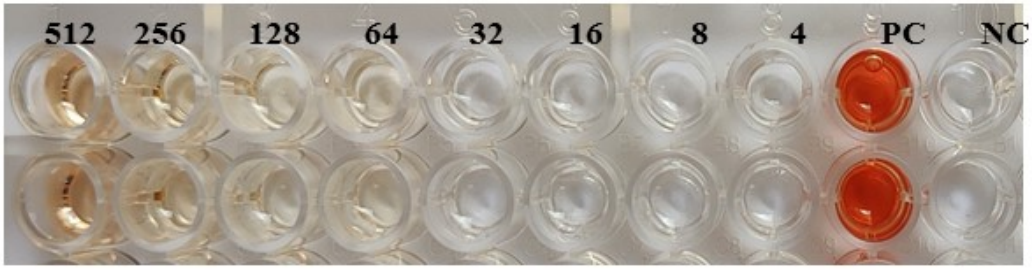


Element	At %
C K	36.22
O K	32.93
Na K	08.08
Si K	10.93
P K	00.90
Ag L	01.94
Ca K	07.29
Ti K	01.71
Total	100.00

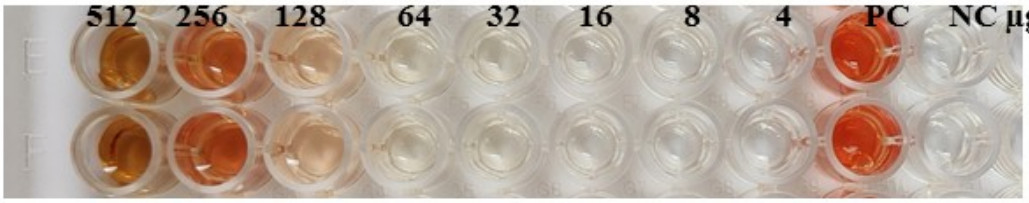
Fig. S1 EDS spectra and elemental composition of: (a) AgBG/PCL; (b) AgBG-GO/PCL; (c) AgBG-rGO/PCL and (d) BG-AgrGO/PCL composite coatings deposited over TNT-Ti.

Table S1: Minimum Inhibitory Concentration (MIC) of AgBG, AgBG-GO, AgBG-rGO and BG-AgrGO composites against *S. aureus* and *E. coli* incubated at 37°C for 16 h.

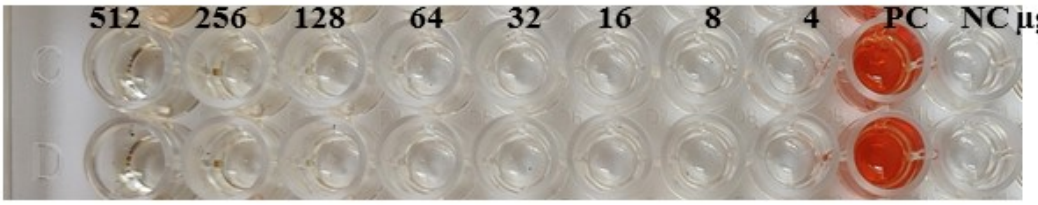
Sample Name	Minimum Inhibitory Concentration (MIC) (mg/L)	
	Gram-positive bacterium	Gram-negative bacterium
	S. aureus	*E. coli*
AgBG	32	32
AgBG-GO	16	16
AgBG-rGO	32	16
BG-AgrGO	64	32
BG	Not determined	Not determined
Positive control	4	2



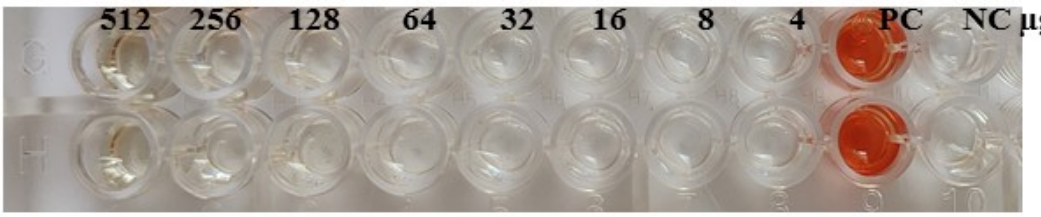
AgBG



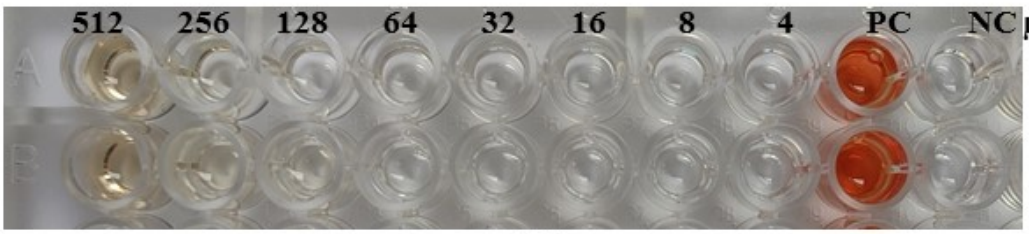
AgBG-GO



BG-AgrGO

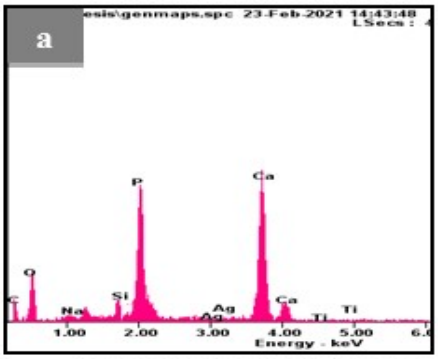


AgBG-rGO

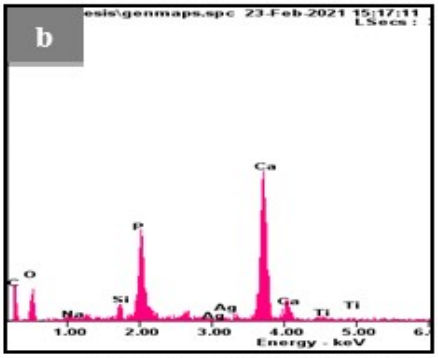


BG

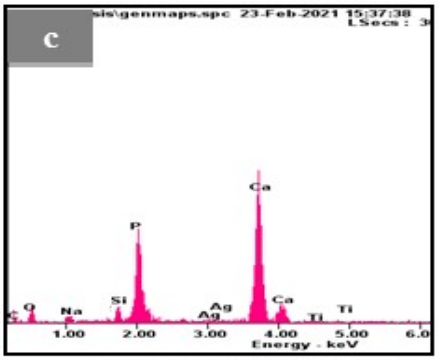
Fig. S2 Hemolytic activity of BG, AgBG, AgBG-GO, AgBG-rGO and BG-AgrGO composites at various concentrations incubated for 1 h at 37 °C.



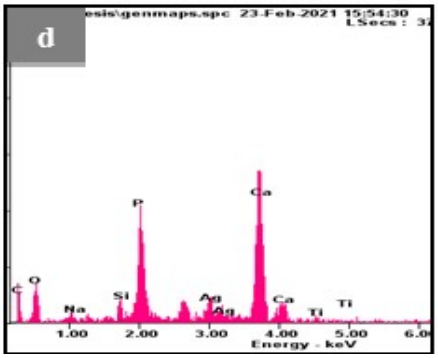
Element	At %
C K	24.34
O K	38.89
Na K	01.25
Si K	02.01
P K	12.90
Ag L	00.15
Ca K	20.32
Ti K	00.14
Total	100.00



Element	At %
C K	26.39
O K	33.59
Na K	02.27
Si K	02.84
P K	13.02
Ag L	00.35
Ca K	21.36
Ti K	00.18
Total	100.00



Element	At %
C K	28.21
O K	33.98
Na K	02.11
Si K	02.45
P K	12.33
Ag L	00.24
Ca K	18.05
Ti K	00.63
Total	100.00

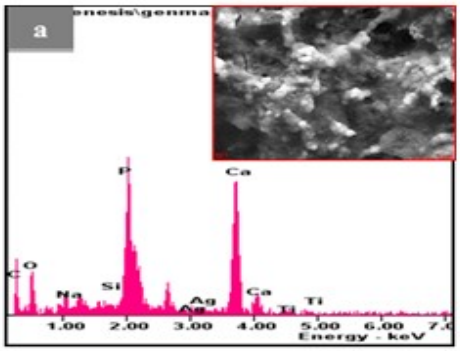


Element	At %
C K	24.63
O K	35.26
Na K	02.95
Si K	02.16
P K	12.71
Ag L	00.98
Ca K	19.52
Ti K	01.79
Total	100.00

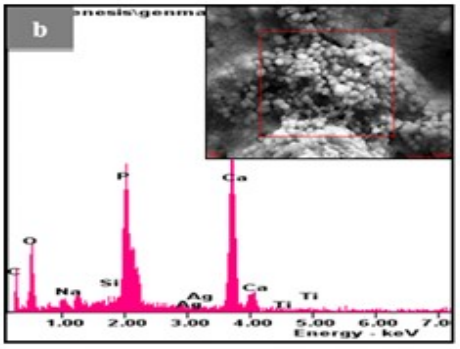
Fig. S3 EDS spectra and elemental composition of: (a) AgBG/PCL; (b) AgBG-GO/PCL; (c) AgBG-rGO/PCL and (d) BG-AgrGO/PCL composite coatings deposited over TNT-Ti after immersion in SBF at 37 ±1° C for 10 days

Table S2: Elemental composition acquired at the surface of AgBG/PCL, AgBG-GO/PCL, AgBG-rGO/PCL and BG-AgrGO/PCL composite coatings deposited over TNT-Ti after immersion in SBF at 37 ± 1 °C for 10 days

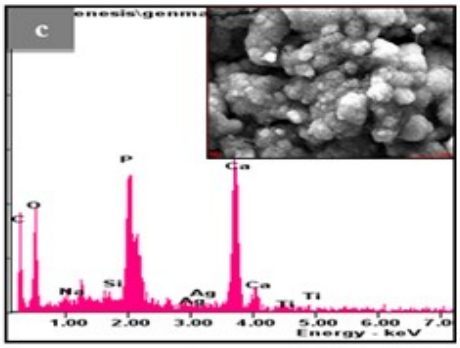
Elemental composition (at. %)	**Type of sample**			
	AgBG/PCL composite coating	AgBG-GO/ PCL composite coating	AgBG-rGO/ PCL composite coating	BG-AgrGO/ PCL composite coating
C	24.34	26.39	28.21	24.63
O	38.89	33.59	35.98	35.26
Na	01.25	02.27	02.11	02.95
Si	02.01	02.84	02.45	02.16
P	12.90	13.02	12.33	12.71
Ag	00.15	00.35	00.24	00.98
Ca	20.32	21.36	18.05	19.52
Ti	00.14	00.18	00.63	01.79
Elemental ratio Ca/P	**1.57**	**1.64**	**1.46**	**1.53**



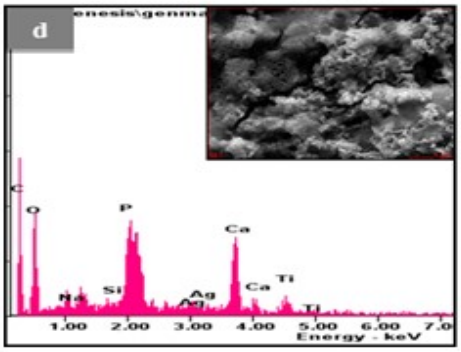
Element	At %
C K	33.05
O K	37.32
Na K	01.69
Si K	00.24
P K	11.31
Ag L	00.19
Ca K	15.78
Ti K	00.42
Total	100.00



Element	At %
C K	32.96
O K	35.86
Na K	01.40
Si K	00.13
P K	12.04
Ag L	00.16
Ca K	17.06
Ti K	00.39
Total	100.00



Element	At %
C K	34.23
O K	36.98
Na K	01.58
Si K	00.18
P K	10.98
Ag L	00.24
Ca K	15.48
Ti K	00.33
Total	100.00



Element	At %
C K	33.65
O K	35.83
Na K	01.63
Si K	00.19
P K	11.45
Ag L	00.63
Ca K	16.07
Ti K	00.55
Total	100.00

Fig. S4 EDS spectra and elemental composition of: (a) AgBG/PCL; (b) AgBG-GO/PCL; (c) AgBG-rGO/PCL and (d) BG-AgrGO/PCL composite coatings deposited over TNT-Ti after immersion in HBSS at 37 ±1° C for 14 days

Elemental composition (at. %)	**Type of sample**			
	AgBG/PCL composite coating	AgBG-GO/ PCL composite coating	AgBG-rGO/ PCL composite coating	BG-AgrGO/PCL composite coating
C	33.05	32.96	34.23	33.65
O	37.32	35.86	36.98	35.83
Na	01.69	01.40	01.58	01.63
Si	00.24	00.13	00.18	00.19
P	11.31	12.04	10.98	11.45
Ag	00.19	00.16	00.24	00.63
Ca	15.78	17.06	15.48	16.07
Ti	00.42	00.39	00.33	0.55
Total	100.00	100.00	100.00	100.00

Table S3: Elemental composition acquired at the surface of AgBG/PCL, AgBG-GO/PCL, AgBG-rGO/PCL and BG-AgrGO/PCL composite coatings deposited over TNT-Ti after immersion in HBSS at 37 ± 1 °C for 14 days