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5 ***Escherichia coli* adhesion to surfaces – a thermodynamic assessment**

6 **Joana M.R. Moreira, Manuel Simões, Luís F. Melo, Filipe J. Mergulhão**

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8 LEPABE – Department of Chemical Engineering, Faculty of Engineering, University of Porto, Rua Dr.

9 Roberto Frias s/n 4200-465 Porto, Portugal

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29 Correspondence: Filipe J. M. Mergulhão, Chemical Engineering Department, Faculty of Engineering

30 University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal. Phone: (+351) 225081668. Fax:

31 (+351) 5081449. E-mail: filipem@fe.up.pt.

32 **Abstract**

33 Several studies have tried to correlate bacterial adhesion with the physicochemical properties of the surface
34 with limited success. Most often, the obtained correlations seem to be only applicable to a particular set of
35 experimental conditions making it difficult to obtain guidelines for the design of antibiofouling surfaces.
36 The ratio between Lifshitz van der Waals apolar component and the electron donor component (γ^{LW}/γ^r) was
37 recently shown to correlate with bacterial adhesion to the surfaces of ship hulls and heat exchangers. In this
38 work, four materials with biomedical application (polystyrene, poly-L-lactide, cellulose acetate and
39 polydimethylsiloxane) and glass were characterized and *Escherichia coli* adhesion to those materials was
40 assayed with a parallel plate flow chamber operating in physiological shear stress conditions. Adhesion
41 was correlated with the γ^{LW}/γ^r ratio further extending the application range tested on the original study.
42 Additionally, results from other studies were also evaluated to confirm the applicability of this correlation
43 to other surfaces, microorganisms and experimental conditions. Results show that bacterial adhesion is
44 reduced in surfaces with lower γ^{LW}/γ^r and enhanced otherwise. This finding may be helpful in the design of
45 new coatings by controlling γ^{LW}/γ^r or in the selection of existing materials according to the desired
46 application.

47
48 **Keywords:** Adhesion, *Escherichia coli*, surface properties, thermodynamics, contact angle

49
50 **Introduction**

51 Microorganisms have a natural tendency to adhere to surfaces and form biofilms [1]. Beneficial biofilms
52 can be found in bioremediation processes, wastewater treatment and in the production of various chemicals
53 [2,3]. However, bacterial adhesion and subsequent biofilm growth is a common problem in industry since
54 it can lead to food spoilage by bioconversion or efficiency loss in heat exchangers [4,5]. In the biomedical
55 field biofilms are responsible for many infections in humans [6] and can cause deterioration of the
56 functionality of medical devices [7]. Therefore, in industry, inhibiting or delaying the onset of detrimental
57 biofilms can represent a reduction in operational costs, since fewer stops are required for sanitation [4,8].
58 In the biomedical field, delaying the onset of biofilms in medical devices may reduce the need for
59 antimicrobial treatment and the costs associated with the replacement of infected implants during revision
60 surgery, which may triple the cost of the primary implant procedure [9].
61 Researchers all over the world are trying to understand bacterial adhesion in order to inhibit or promote
62 biofilm development [10,11]. Several strategies have been evaluated in order to control biofilm

63 development [9,12,13] and one of the most promising is to control bacterial adhesion [8,14-17].

64 Bacterial adhesion begins with the attraction between cells and surfaces, followed by adsorption and
65 attachment [18]. The physicochemical forces involved in the initial approach of cells to surfaces are
66 primarily van der Waals, electrostatic, hydration and hydrophobic interactions [18]. Therefore, the correct
67 selection of materials to be used in industrial and biomedical settings can be determinant to the onset of
68 bacterial biofilms on these surfaces.

69 Researchers are trying to define criteria for selection of new materials according to their surface properties
70 [16,17,19]. This methodology has been used intensively since accessible and fast methods such as contact
71 angle measurements are available enabling time and cost reduction in the laboratory [20-22]. However,
72 finding a correlation between surface properties and bacterial adhesion rates has been challenging [23-25].

73 Li and Logan [26] studied the contribution of surface charge and hydrophobicity on the adhesion of three
74 *Escherichia coli* strains, two *Pseudomonas aeruginosa* strains and two *Burkholderia cepacia* strains on
75 metal oxide-coated and uncoated glass surfaces. These authors observed that adhesion was not significantly
76 correlated with bacterial charge and contact angle. Liu and Zhao [27] used the ratio between apolar Lifshitz
77 van der Waals components (γ^{LW}) and electron donor components (r) of modified stainless steel (Ni-P-TiO₂-
78 PTFE nanocomposite coatings) as a surface property parameter to correlate with *Pseudomonas fluorescens*,
79 *Cobetia marina* and *Vibrio alginolyticus* adhesion under static and dynamic conditions. Their results
80 demonstrated that coatings with the lowest γ^{LW}/r had the lowest bacterial adhesion values, and increasing
81 γ^{LW}/r led to higher bacterial adhesion. That study was conducted with surfaces that may be used in ship
82 hulls and heat exchangers but the authors suggested that their results are transferable to the biomedical
83 field. This hypothesis was tested on this work by using four polymeric surfaces (polystyrene (PS), poly-L-
84 lactide (PLLA), cellulose acetate (CA) and polydimethylsiloxane (PDMS)) which can be used in
85 biomedical devices in the human body [18,28-30] and glass. Thermodynamic surface properties were
86 evaluated in order to find if they could be correlated with bacterial adhesion. The hydrodynamic conditions
87 used are similar to those found in the bladder, urinary tract and reproductive system [31,32] where
88 biomedical devices constructed with the selected materials are used [28,29,33,34] and where *E. coli* is the
89 major cause for infection [35,36]. These surfaces were also selected due to their different γ^{LW}/r values
90 which extend the range tested by Liu and Zhao [27]. The applicability of this correlation was also tested
91 using data from other authors studying bacterial adhesion or protein adsorption to different materials (soil
92 minerals, synthetic materials, plasma treated surfaces and metallic materials) in different systems and
93 operational conditions. Thus, the rationale for this work was to find out a selection/design criteria to predict

94 bacterial adhesion to materials used in the industrial and biomedical fields.

95

96 **Materials and methods**

97 ***Bacteria and culture conditions***

98 A starter culture of *E. coli* JM109(DE3) was obtained by inoculation of 500 μ L of a glycerol stock (kept at
99 -80 °C) to a total volume of 0.2 L of inoculation media with 5.5 g L⁻¹ glucose, 2.5 g L⁻¹ peptone, 1.25 g L⁻¹
100 yeast extract in phosphate buffer (1.88 g L⁻¹ KH₂PO₄ and 2.60 g L⁻¹ Na₂HPO₄) at pH 7.0 [37]. This culture
101 was grown in a 1 L shake-flask, incubated overnight at 30 °C with orbital agitation (120 rpm). A volume
102 of 60 mL from the overnight grown culture was used to harvest cells by centrifugation (10 min, 3202 g).
103 Cells were washed twice with citrate buffer 0.05 M [38], pH 5.0 and the pellet was resuspended and diluted
104 in the same buffer in order to reach a cell concentration of 7.6×10^7 cell.mL⁻¹.

105

106 ***Surface preparation***

107 Five materials, PS, glass, PLLA, CA and PDMS were prepared for adhesion assays. PS surface and
108 microscope glass slides (VWR) were firstly washed with a commercial detergent (Sonasol Pril, Henkel
109 Ibérica S A) and immersed in sodium hypochlorite (3%). After rinsing with distilled water, part of the
110 microscope glass slides were coated with the polymers. These were prepared by mixing the polymer in
111 solid form with solvents. Dichloromethane was added to PLLA at 5% (w/w), acetone was added to CA at
112 8% (w/w) and a curing agent (Sylgard 184 Part B, Dow Corning) was added to PDMS (at a 1:10 ratio)
113 (polymers from Sigma, solvents from Normapur). This mixture was prepared in a beaker where it was
114 manually stirred with a glass rod to homogenize the two components without introducing bubbles. The
115 polymers were then deposited as a thin layer on top of glass slides by spin coating (Spin150 Polos™), for
116 PDMS at 2000 rpm for 60 seconds and for the other surfaces at 5000 rpm for 50 seconds.

117

118

119 ***Surface characterization***

120 The surface charge of bacteria and material surfaces was characterized by zeta potential and surface
121 hydrophobicity using the contact angle method. One *E. coli* suspension was prepared as described before,
122 and particle suspensions of each material [39] were also prepared in order to measure the electrophoretic
123 mobility, using a Nano Zetasizer (Malvern Instruments, UK). The hydrophobicity of bacteria and surfaces
124 was evaluated considering the Lifshitz van der Waals acid base approach [40]. Contact angles were

125 determined automatically by the sessile drop method in a contact angle meter model (OCA 15 Plus;
 126 Dataphysics, Filderstadt, Germany) using water, formamide and α -bromonaphtalene (Sigma) as reference
 127 liquids with surface tension components taken from literature [41]. For each surface (PLLA, PS, CA, PDMS
 128 and glass), at least 10 measurements with each liquid were performed at 25 ± 2 °C. One *E. coli* suspension
 129 was prepared in the same conditions as for the adhesion assay and its physicochemical properties were also
 130 determined by sessile drop contact angle measurement as described by Busscher et al. [42].

131 According to van Oss [40], the total surface energy (γ^{Tot}) of a pure substance is the sum of the apolar
 132 Lifshitz-van der Waals components of the surface free energy (γ^{LW}) and polar Lewis acid-base components
 133 (γ^{AB}):

$$134 \quad \gamma^{TOT} = \gamma^{LW} + \gamma^{AB} \quad (1)$$

135 The polar AB component comprises the electron acceptor γ^+ and electron donor γ^- parameters, and is given
 136 by:

$$137 \quad \gamma^{AB} = 2\sqrt{\gamma^+ \gamma^-} \quad (2)$$

138 The surface energy components of a solid or bacterial surface (s) are obtained by measuring the contact
 139 angles (θ) with the three different liquids (l) with known surface tension components, followed by the
 140 simultaneous resolution of three equations of the type:

$$141 \quad (1 + \cos \theta)\gamma_l = 2\left(\sqrt{\gamma_s^{LW} \gamma_l^{LW}} + \sqrt{\gamma_s^+ \gamma_l^-} + \sqrt{\gamma_s^- \gamma_l^+}\right) \quad (3)$$

142 The degree of hydrophobicity of a given surface (solid and bacterial surface) is expressed as the free energy
 143 of interaction (ΔG mJ.m⁻²) between two entities of that surface immersed in polar liquid (such as water (w)
 144 as a model solvent).

145 If the interaction between the two entities is stronger than the interaction of each entity with water, $\Delta G < 0$
 146 mJ.m⁻², the material is considered hydrophobic, if $\Delta G > 0$ mJ.m⁻², the material is hydrophilic. ΔG was
 147 calculated from the surface tension components of the interacting entities, using the equation:

$$148 \quad \Delta G = -2\left(\sqrt{\gamma_s^{LW}} - \sqrt{\gamma_w^{LW}}\right)^2 + 4\left(\sqrt{\gamma_s^+ \gamma_w^-} + \sqrt{\gamma_s^- \gamma_w^+} - \sqrt{\gamma_s^+ \gamma_s^-} - \sqrt{\gamma_w^+ \gamma_w^-}\right); \quad (4)$$

149 When studying the interaction (free energy of adhesion) between surface (s) and bacteria (b) that are
 150 immersed in water, the total interaction energy, ΔG^{Adh} , can be expressed as:

$$151 \quad \Delta G^{Adh} = \gamma_{sb}^{LW} - \gamma_{sw}^{LW} - \gamma_{bw}^{LW} + 2\left[\sqrt{\gamma_w^+}\left(\sqrt{\gamma_s^-} + \sqrt{\gamma_b^-} - \sqrt{\gamma_w^-}\right) + \sqrt{\gamma_w^-}\left(\sqrt{\gamma_s^+} + \sqrt{\gamma_b^+} - \sqrt{\gamma_w^+}\right) - \sqrt{\gamma_s^+ \gamma_b^-} - \sqrt{\gamma_s^- \gamma_b^+}\right] \quad (5)$$

152 Thermodynamically, if $\Delta G^{Adh} < 0$ mJ.m⁻² adhesion is favoured, while adhesion is not expected to occur if

153 $\Delta G^{\text{Adh}} > 0 \text{ mJ.m}^{-2}$.

154

155 ***Flow chamber experiments***

156 A PPFC with dimensions of $25.4 \times 1.6 \times 0.8 \text{ cm}$ (L x W x H) was connected to a centrifugal pump by a
157 tubing system. It contained a bottom and a top opening at the exit for the introduction of the test surfaces.
158 The PPFC was mounted in a microscope (Nikon Eclipse LV100, Japan) to monitor *E. coli* attachment to
159 each surface for 30 min. The cellular suspension was circulated at 2 ml.s^{-1} and images were acquired with
160 a camera (Nikon digital sight DS-RI 1, Japan) connected to the microscope. The hydrodynamic conditions
161 were simulated by computational fluid dynamics and the results have shown that in the viewing point, the
162 conditions are of steady flow and the average shear stress was of 0.01 Pa (not shown). Approximate shear
163 stresses can be found in the bladder, urinary tract and reproductive system [31,32]. Temperature was kept
164 constant at $37 \text{ }^\circ\text{C}$ using a recirculating water bath. All adhesion experiments were performed in triplicate
165 for each surface.

166 The microscopy images recorded during the cell adhesion assays were analyzed with the program ImageJ
167 (v1.46r). The number of adhered cells after 30 min was then divided by the surface area of the field of view
168 to obtain the density of bacteria per square centimeter.

169

170 ***Statistical analysis***

171 Paired *t*-test analyses were performed to estimate whether or not there was a significant difference between
172 the results obtained on each surface. Results were evaluated individually using the three independent results
173 obtained with one surface and the three individual results obtained with other surface. Results were
174 considered statistically different when a confidence level greater than 95% was reached ($P < 0.05$). Standard
175 deviation between the 3 values obtained from the independent experiments was also calculated.

176

177 ***Re-plotted data***

178 Relevant works, where some authors had tried to find a correlation between surface properties of different
179 materials and bacterial adhesion (as well as protein adsorption to those surfaces) were selected and data
180 was re-plotted in this work in order to compare with the new data here presented. Bacterial adhesion and
181 protein adsorption data were represented as a function of the ratio between the Lifshitz-van der Waals
182 component and the Lewis acid-base electron donor γ -component ($\gamma^{\text{LW}}/\gamma$) for each tested surface.

183

184 **Results and discussion**

185 In this work, five materials (PLLA, PDMS, PS, CA and glass) were tested in order to evaluate *E. coli*
186 adhesion after determination of thermodynamic surface properties. Table 1 shows the contact angle
187 measurements for each surface, the thermodynamic surface energy properties, the zeta potential values and
188 the cell adhesion results.

189 Based on contact angle values, surfaces can be classified into hydrophilic or hydrophobic if the contact
190 angle of water with the surfaces is, respectively, lower or higher than 65° [43]. From the results in Table 1
191 it is possible to anticipate that glass and *E. coli* have hydrophilic surfaces and the other surfaces are
192 hydrophobic. Regarding the values determined for the van der Waals forces apolar component (γ^{LW}) [44],
193 it is possible to observe that CA has the highest attractive apolar component value and PDMS the lowest.
194 In what concerns the polar surface components ($\gamma^+ \gamma^-$), results showed that PLLA, PDMS, PS and *E. coli* are
195 monopolar surfaces, being electron donors (Table 1). Conversely, CA and glass are polar surfaces, being
196 electron donors and acceptors. From the total free energy results, it is also possible to observe that PLLA,
197 PDMS, PS, and CA are hydrophobic surfaces ($\Delta G < 0 \text{ mJ.m}^{-2}$) whereas glass and *E. coli* are hydrophilic
198 ($\Delta G > 0 \text{ mJ.m}^{-2}$). Therefore, results obtained with the determination of surface properties support the
199 preliminary evaluation made by water contact angle measurement.

200 From the cell adhesion results (Table 1) it is possible to observe that a higher number of adhered cells was
201 obtained on the PLLA surface (the most hydrophobic) and a lower bacterial adhesion value was observed
202 on glass ($P < 0.05$) (the most hydrophilic). Previous studies have shown that *E. coli* adhesion is enhanced
203 in hydrophobic surfaces and decreased in hydrophilic materials [45,46]. However, if hydrophobicity was
204 the only relevant factor, an increase in the ΔG values should have led to a consistent decrease in bacterial
205 adhesion and this was not observed for PDMS. Thus, a correlation between surface hydrophobicity and
206 bacterial adhesion was not found.

207 The thermodynamic theory indicates that a system with a lower interacting energy (ΔG^{Adh}) usually leads to
208 a higher affinity between bacteria and surfaces [21]. Therefore, based on the results in Table 1 *E. coli* should
209 have adhered more to CA and PLLA and have a lower affinity to glass. Thus, it seems that cell adhesion is
210 also not directly correlated with ΔG^{Adh} . Other authors have also tried to find a correlation between bacterial
211 adhesion and surface hydrophobicity or surface free energy of adhesion without success. In a study by
212 Oliveira et al. [24], a correlation between the hydrophobicity of materials (polyethylene, polypropylene,
213 and granite) used in kitchens and the adhesion of four *Salmonella enteritidis* strains was also not found.

214 Barton et al. [47] were also not successful in finding a correlation between the free energy of adhesion of
215 orthopedic implant polymers (poly(orthoester), poly(L-lactic acid), polysulfone, polyethylene, and
216 poly(ether-ether ketone)) and *S. epidermidis* or *E. coli* adhesion.

217 In this work, a correlation between electron donor character (γ^r) and bacterial adhesion was also not
218 observed particularly for glass which showed a very high value of γ^r (52.43 mJ.m⁻²) compared to the other
219 surfaces (Table 1). Additionally, for the zeta potential data, negative values indicate electrical repulsion
220 between negative charged bacteria and surfaces [48] but a correlation was not found for this parameter
221 either.

222 Several studies have been performed by other research groups in order to find a good correlation between
223 bacterial adhesion (and adsorption of organic/inorganic particles) and some physicochemical parameter
224 from the surface. A literature survey was performed in order to find such works where complete information
225 about the thermodynamic properties was included or where these properties could be calculated from
226 reported data (Table 2). Hong et al. [49] studied the role of surface properties in the adhesion of *Bacillus*
227 *subtilis* to soil minerals. These authors observed a significant correlation between adhesion capacity and
228 the specific external surface area of the minerals, but they did not find a correlation between surface
229 hydrophobicity (ranging from -32. 2 and 33.2 mJ.m⁻²) and adhesion. Katsikogianni et al. [50] studied the
230 role of the free energy of adhesion (from -10.5 to 17.2 mJ.m⁻²) in the attachment of *Staphylococcus*
231 *epidermidis* to plasma modified PET films under quasi-static (5 s⁻¹) and dynamic conditions (50 and 200 s⁻¹).
232 A strong correlation between the thermodynamic predictions and the measured values of bacterial
233 adhesion under quasi-static conditions was observed. Moreover, the authors reported that the polar acid–
234 base interactions dominated the interactions of bacteria with the substrates in aqueous media. However,
235 under flow conditions, the increase in the shear rate reduced the predictability of the thermodynamic
236 models. Cunliffe et al. [51] used synthetic materials with energies ranging from 15 to 42 mJ.m⁻² for bacterial
237 adhesion and adsorption of bovine serum albumin (with a net negative charge) and cytochrome c (with a
238 positive charge). Protein adsorption and *Listeria monocytogenes* adhesion also showed some correlation
239 with the chemistry of the surfaces. Liu and Zhao [27] have suggested a ratio between Lifshitz van der Waals
240 apolar component and the electron donor component (γ^{LW}/γ^r) as a good correlation factor for cell adhesion.
241 These authors have used *P. fluorescens*, *C. marina*, and *V. alginolyticus* and Ni-P-TiO₂-PTFE coatings in
242 different hydrodynamic conditions (Table 2). This ratio was also tested for the adhesion values obtained in
243 the present work as well as for the results reported by other groups comprising 29 different surfaces, 7
244 organisms, 2 proteins and different shear stress conditions (Table 2). The (γ^{LW}/γ^r) range covered in each

245 study as well as the identification of the tested surfaces is provided in Fig. 1.

246 In the present work, surfaces with the highest γ^{LW}/r values had the highest bacterial adhesion (Fig. 2a). This
247 may be due to a lower surface electron donor component (γ^r , repulsive) or a high apolar component (γ^{LW} ,
248 attractive) [44]. The highest adhesion value was observed for PLLA ($P < 0.05$) which has the lowest
249 repulsive forces (lower γ^r , Table 1) when compared with the adhesion values observed for PS, CA, and
250 PDMS. Regarding PDMS, it is possible to note that a similar γ^r value was observed for this surface and
251 PLLA. However, PDMS exhibited the lowest apolar attractive forces value (γ^{LW}) and this may have led to
252 a lower adhesion than observed for CA and PS (with higher γ^r , Table 1). Glass, has the strongest repulsive
253 force value (γ^r) which can explain the lowest adhesion.

254 In the work of Liu and Zhao [27] the second order equation $y = a + bx + cx^2$ was used to correlate
255 experimental data and the obtained correlation coefficients varied between 0.8123 and 0.9247 (Figs. 2b and
256 c). In this work, the same equation was applied to the adhesion results and a correlation factor of 0.9917
257 was obtained (Fig. 2a). Additionally, results from all these works from the literature survey (Table 2 and
258 Fig. 1) were re-plotted in Fig. 2, where it is possible to see that the γ^{LW}/r parameter has a strong correlation
259 with bacterial adhesion results from the work of Katsikogianni et al. [50] (Fig. 2d), Hong et al. [49] (Fig.
260 2e) and Cunliffe et al. [51] (Fig. 2f) and with the values obtained for protein adsorption by the same author
261 (Figs. 2g and h).

262 Liu and Zhao [27] were able to correlate cell adhesion to the γ^{LW}/r ratio and their working range was
263 between 1.21 and 6.74 (Fig. 1). Although these authors have tested metallic surfaces that can be used in
264 heat exchangers and ship hulls, they have suggested that their results could also be applied to biomedical
265 surfaces. With the results obtained in the present work, this hypothesis was confirmed since a good
266 correlation between *E. coli* adhesion to biomedical polymers and the γ^{LW}/r surface parameter was found for
267 an extended γ^{LW}/r range. Additionally, and considering data obtained from other works, it was possible to
268 observe the validity of this correlation under diversified conditions.

269 Therefore, the available data seem to indicate that the γ^{LW}/r ratio can be a good parameter for rapid material
270 selection that can be used either to promote (higher γ^{LW}/r values) or to decrease bacterial adhesion (lower
271 γ^{LW}/r values). These results may also be helpful in the design of new materials by controlling the ratio γ^{LW}
272 $/r$ according to the desired application.

273

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Table 1 Surface thermodynamic properties and cell adhesion results

Surface	Contact angle / (°)			Surface properties					<i>E. coli</i> - surface interaction	Zeta potential / mV	Adhered cells.cm ⁻²
	Water	Formamide	α -bromonaphtalene	$\gamma^{LW}/$ (mJ.m ⁻²)	$\gamma^+ /$ (mJ.m ⁻²)	$\gamma^- /$ (mJ.m ⁻²)	$\Delta G/$ (mJ.m ⁻²)	$\Delta G^{Adh}/$ (mJ.m ⁻²)			
PLLA	88.03± 1.01	68.49± 0.95	25.59± 1.54	40.15	0.000	4.374	-65.32	29.90	-27.90	1.82×10 ⁶ ±2.76×10 ⁴	
PDMS	113.6± 0.62	111.2± 0.61	87.62± 1.77	12.04	0.000	4.544	-61.82	32.60	-29.30	1.29×10 ⁶ ±3.79×10 ⁵	
PS	80.81± 0.68	64.33±1.24	24.64± 1.11	40.45	0.000	8.290	-49.56	37.80	-29.80	1.36×10 ⁶ ±1.35×10 ⁵	
CA	65.24± 0.49	36.63± 2.05	22.47± 1.05	41.09	1.441	9.629	-37.58	25.50	-23.40	1.35×10 ⁶ ±1.32×10 ⁵	
Glass	16.38±0.35	17.19± 0.35	44.48± 0.71	32.59	2.586	52.43	27.99	62.90	-37.00	1.18×10 ⁶ ±7.47×10 ⁴	
<i>E. coli</i>	19.13± 0.88	73.34± 0.65	58.54± 2.01	25.71	0.000	123.2	121.90	n/a	-17.00		

PS - polystyrene, PLLA - poly-L-lactide, CA - cellulose acetate, PDMS - polydimethylsiloxane; γ^{LW} - apolar component, γ^+ and γ^- - surface tension parameters, ΔG - free surface energy, ΔG^{Adh} - free energy of interaction between *E. coli* and each surface; n/a – not applicable.

Table 2 Summary of the experimental conditions used by other authors and in the present study

Organism/compound	Surface material	Platform	T / °C	Hydrodynamics	Assay time / h	Correlated parameter	Reference
<i>Bacillus subtilis</i>	Soil minerals	Conical flask	25	Shaking at 1.2 g	2	SESA	[49]
<i>Staphylococcus epidermis</i>	Helium plasma treated PET ^b	Well - tissue culture plates and a radial flow chamber	37	Shear rate: 5, 50 and 200 s ⁻¹	2.5	ΔG^{Adh}	[50]
<i>Listeria monocytogenes</i> Bovine serum albumine Cytochrome c	Synthetic	Capped bottles	37	Gentle shaking	24 and 1 ^c	Surface chemistry	[51]
<i>Pseudomonas fluorescens</i> <i>Cobetia marina</i> <i>Vibrio alginolyticus</i>	Ni – P coatings with TiO ₂ and PTFE, stainless steel	Static tank and dynamic PPFC	28	Static, dynamic – shear stress: 0.98, 0.46, 0.21 mPa	6 and 24 ^d	$\gamma^{\text{LW}}/\gamma^-$	[27,52]
<i>Escherichia coli</i>	Polymeric coatings, glass	PPFC	37	Shear stress: 0.01 Pa	0.5	$\gamma^{\text{LW}}/\gamma^-$	This work

^a SESA – Specific external surface area, ^b PET - polyethylene terephthalate,

^c Referent to microorganism adhesion and proteins adsorption, respectively

^d Referent to static and dynamic conditions, respectively

Reference	$\gamma^L W / \gamma^S$ range										
	0	1	2	3	4	5	6	7	8	9	10
[49]		A1 A2	A3	A4		A5					
[50]		B1B2	B3 B4	B5							
[51]		C1	C2	C3	C4						
[27, 52]			D1 D2 D3 D4 D5 D6	D7 D8	D9		D10				
This work		E1	E2		E3	E4			E5		

A1 - mica, A2 - quartz, A3 - kaolinite, A4 - montmorillonite, A5 - sirmessite

B1 - 1 h helium plasma treated polyethylene terephthalate (PET), B2 - 8 days helium plasma treated PET, B3 - 17 days helium plasma treated PET, B4 - 58 days helium plasma treated PET, B5 - 30 days helium plasma treated PET

C1 - SiO-(CH₂)₃NH-CO-PEO-5000-OCH₃, C2 - SiO-(CH₂)₃NH-CO-NH₂, C3 - SiO-(CH₂)₃NH-CO-PNMeAm, C4 - SiO-(CH₂)₃NH-CO-(CF₂)CF₃

D1 - Ni-P-TiO₂-PTFE 3, D2 - Ni-P-TiO₂-PTFE 4, D3 - Ni-P-TiO₂-PTFE 1, D4 - Ni-P-TiO₂-PTFE 2, D5 - Ni-P-TiO₂-PTFE 2, D6 - Ni-P-TiO₂-PTFE 2, D7 - Ni-P-TiO₂-PTFE 1, D8 - Ni-P-TiO₂-PTFE 2, D9 - Ni - P, D10 - stainless steel

E1 - glass, E2 - polydimethylsiloxane, E3 - cellulose acetate, E4 - polystyrene, E5 - poly-L-lactide

Fig. 1 Surfaces used and $\gamma^L W / \gamma^S$ tested in different works attempting to find a correlation between adhesion and thermodynamic properties.

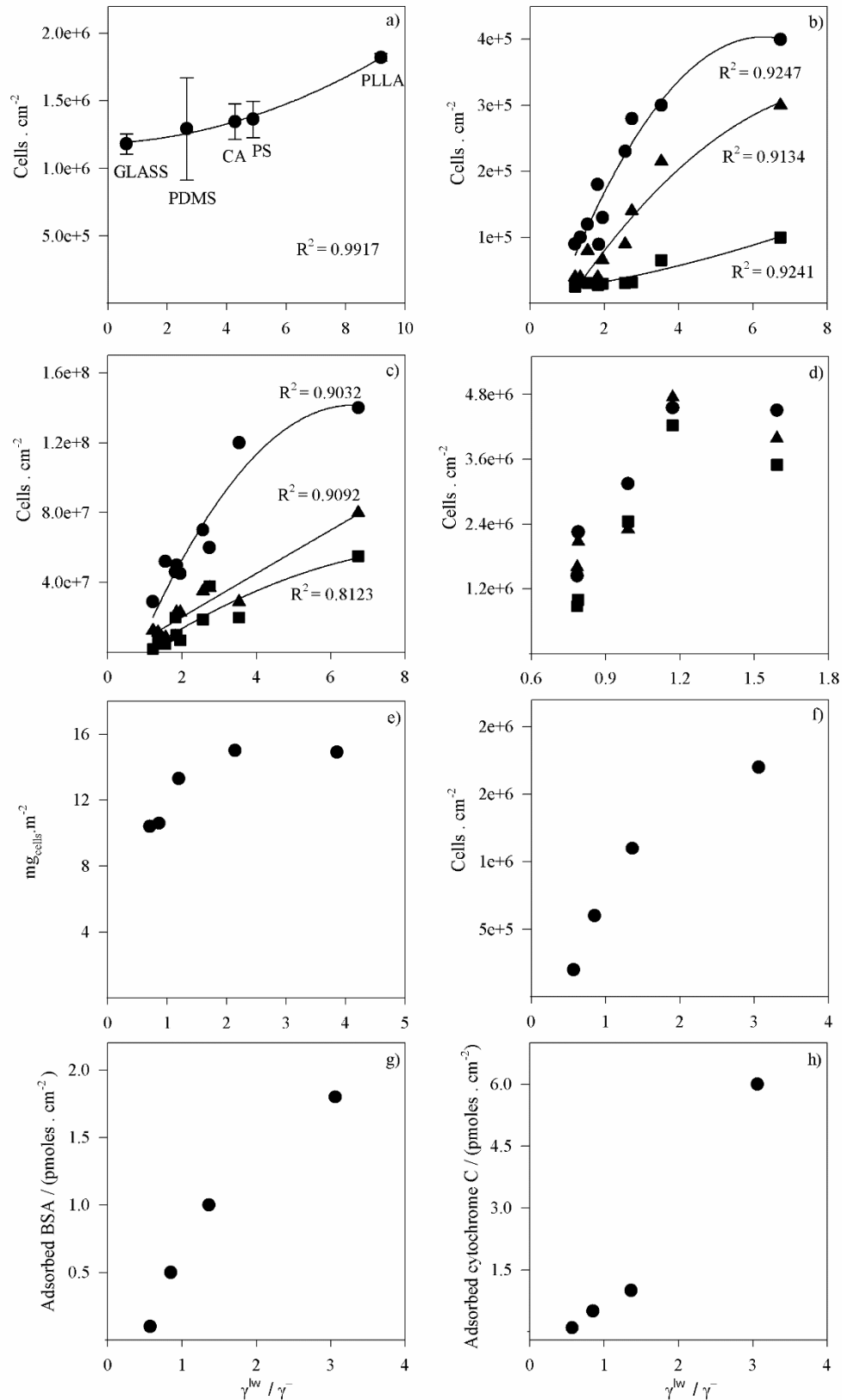


Fig. 2 Relationship between bacterial adhesion or protein adsorption and the ratio between apolar Lifshitz van der Waals components (γ^{LW}) and electron donor component (r). a) *E. coli* adhesion on polymeric and glass surfaces b) *Vibrio* (circle), *Cobetia* (triangle) and *P. fluorescens* (square) adhesion on Ni – P coatings

with TiO₂ and PTFE and stainless steel, re-plotted from Liu and Zhao [27] , c) *Vibrio* adhesion at 0.21 (circle), 0.46 (triangle), and 0.98 (square) mPa on Ni – P coatings with TiO₂ and PTFE and stainless steel, re-plotted from Liu and Zhao [27], d) *Staphylococcus epidermis* adhesion at 5 (circle), 50 (triangle) and 200 s⁻¹ (square) on helium plasma treated PET, re-plotted from Katsikogianni et al. [50], e) *Bacillus subtilis* adhesion on soil minerals, re-plotted from Hong et al. [49], f) *Listeria monocytogenes* adhesion on synthetic surfaces, re-plotted from Cunliffe et al. [51], g) Bovine serum albumin adsorption on synthetic surfaces, re-plotted from Cunliffe et al. [51], h) Cytochrome c adsorption on synthetic surfaces, re-plotted from Cunliffe et al. [51]. Whenever a correlation was reported by the original authors it was also represented in this figure and the correlation factor (R²) is indicated (panels a, b and c).