

# Impact of polyglycerol polyricinoleate (PGPR) type on its interfacial characteristics in W/O emulsions

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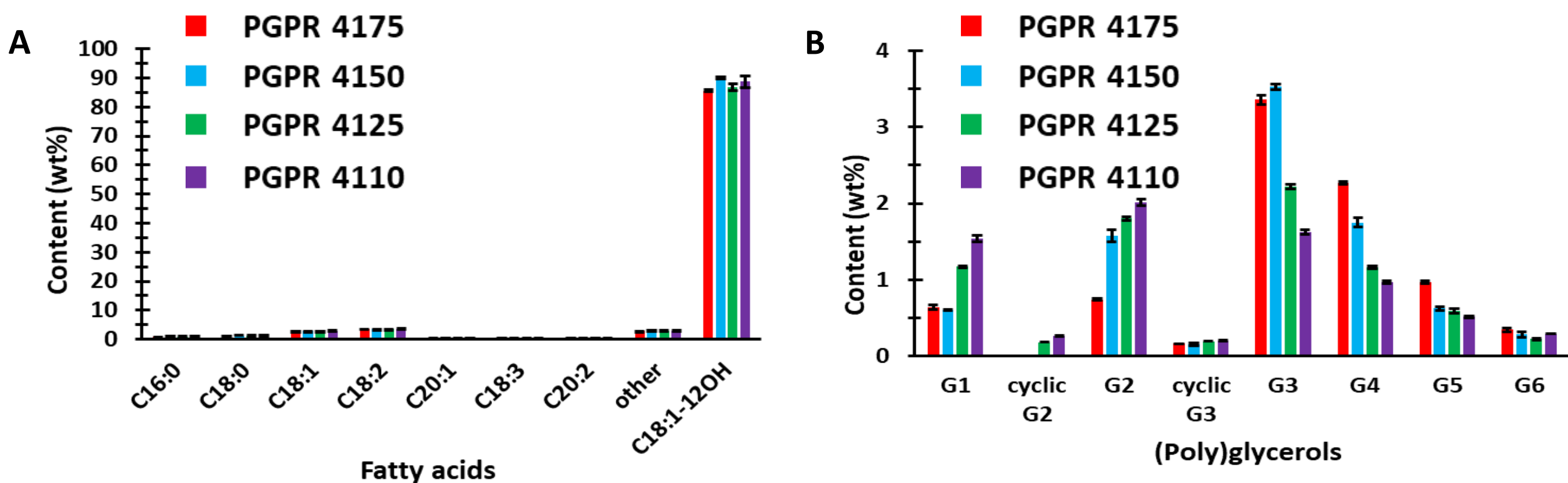
## INTRODUCTION

There are many commercially available PGPR products, and the different products vary in their impact on the interfacial characteristics [1, 2]. To understand the reasons for these differences, this study investigated the molecular properties and the interfacial characteristics of four PGPRs (4175, 4150, 4125, 4110; Palsgaard) to link the structure of PGPR with its surface activity.

## METHODS

- (1) **Molecular properties**
  - Gas chromatography (GC): fatty acids and (poly)glycerol composition.
- (2) **Interfacial characteristics**
  - Drop shape tensiometry: interfacial tension and dilatational rheology at the glyceryl triheptanoate-water interface.
  - Quartz crystal microbalance with dissipation monitoring (QCM-D): mass and viscosity during adsorption on a model hydrophilic surface (a golden sensor modified by mercaptoethanol).
  - GC: adsorbed concentration of PGPR (based on ricinoleic acid content) and polyglycerols in W/O (50/50, m/m) emulsions.
    - Emulsion composition: water phase contains 0.1 M NaCl; oil phase contains 2.5 wt% PGPR in glyceryl triheptanoate; Emulsion preparation: homogenization by an Ultra-Turrax (S25-10G, Germany) using 17500 rpm at 60 °C for 5 min.
    - Adsorbed amount was determined by depletion method (i.e. adsorption follows from original and residual concentration in the oil phase), whereby the continuous oil phase was obtained by ultracentrifugation at 45000g for 2 h (Beckmann Coulter, US).

## RESULTS



**Figure 1.** Fatty acids (A) and (poly)glycerols (B) composition (in wt%) of four PGPRs. G1: glycerol; G2: diglycerol; G3: triglycerol; G4: tetraglycerol; G5: pentaglycerol; G6: hexaglycerol.

**Table 1.** Dynamic interfacial tension ( $\gamma$ ), dilatational elastic modulus ( $\epsilon_d$ ) and viscous modulus ( $\eta_d$ ) of 4 types of PGPR in glyceryl triheptanoate at adsorption equilibrium. Amplitude and frequency are 15 % and 0.1 Hz, respectively.

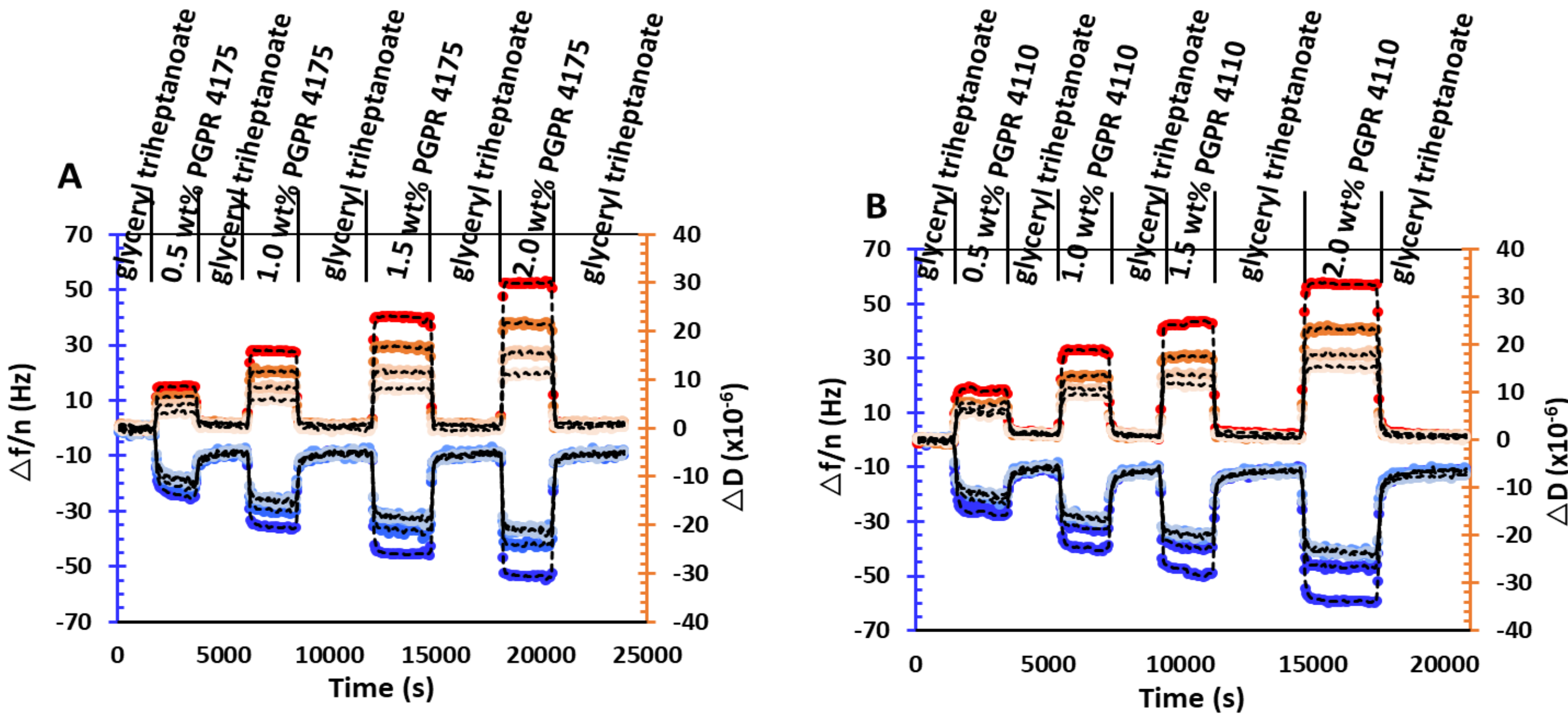
Sample	$\gamma$ (mN/m)	$\epsilon_d$ (mN/m)	$\eta_d$ (mN/m)
0.2 wt% PGPR 4175	6.04 ± 0.14 <sup>a</sup>	13.71 ± 0.27 <sup>a</sup>	3.72 ± 0.12 <sup>a</sup>
0.2 wt% PGPR 4150	6.60 ± 0.19 <sup>a</sup>	13.07 ± 0.26 <sup>a</sup>	3.73 ± 0.29 <sup>a</sup>
0.2 wt% PGPR 4125	7.45 ± 0.27 <sup>b</sup>	13.59 ± 0.53 <sup>a</sup>	3.40 ± 0.12 <sup>a</sup>
0.2 wt% PGPR 4110	8.36 ± 0.40 <sup>c</sup>	13.44 ± 0.50 <sup>a</sup>	3.49 ± 0.15 <sup>a</sup>

<sup>a</sup> and <sup>b</sup>: different superscript letters in a column indicate significantly different results ( $p$ -value <0.05).

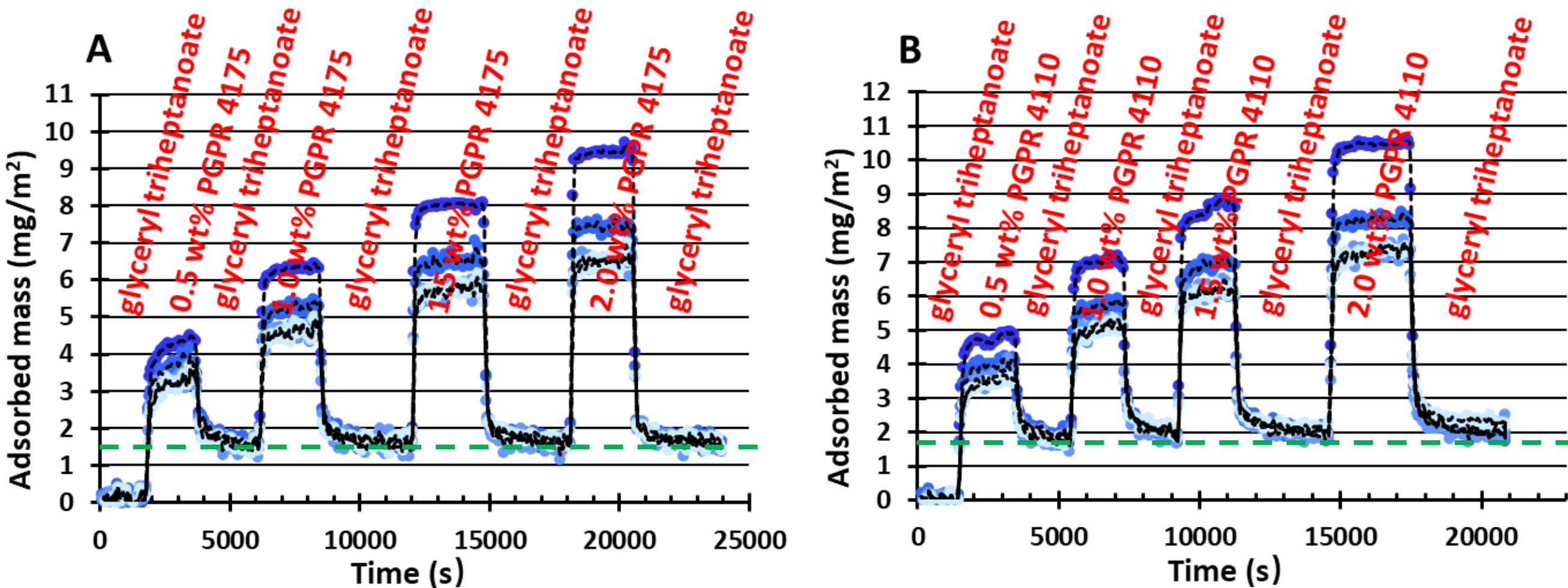
**Table 2.** Adsorbed amount (in wt%) of PGPR (based on ricinoleic acid) and polyglycerols in W/O (50/50, m/m) emulsions, stabilized by 2.5 wt% of PGPR 4175 or PGPR 4110.

Adsorbed concentration (wt%)	PGPR 4175	PGPR 4110
PGPR (based on ricinoleic acid)	0.4225 ± 0.0386 <sup>a</sup>	0.4950 ± 0.0208 <sup>b</sup>
Diglycerol	0.0047 ± 0.0001 <sup>b</sup>	0.0183 ± 0.0010 <sup>a</sup>
Triglycerol	0.0291 ± 0.0016 <sup>b</sup>	0.0164 ± 0.0007 <sup>a</sup>
Tetraglycerol	0.0276 ± 0.0016 <sup>b</sup>	0.0110 ± 0.0007 <sup>a</sup>
Pentaglycerol	0.0163 ± 0.0001 <sup>b</sup>	0.0093 ± 0.0001 <sup>a</sup>

<sup>a</sup> and <sup>b</sup>: different superscript letters in a row indicate significantly different results ( $p$ -value <0.05).



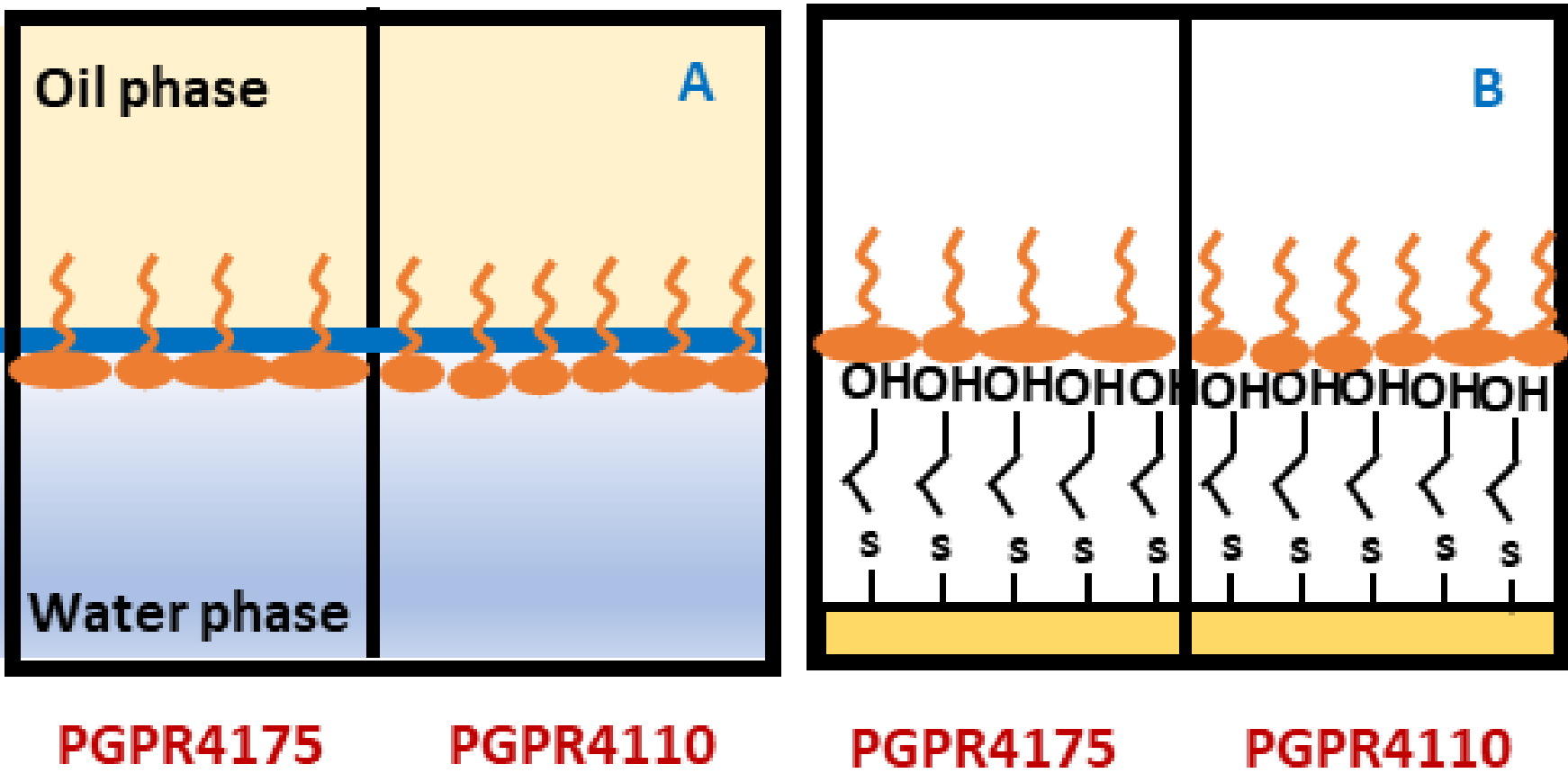
**Figure 2.** Normalised frequency (blue) and dissipation (red) shifts of different concentration of PGPR in glyceryl triheptanoate as a function of time (s) for the 3rd, 5th, 7th, and 9th overtones (with fading colours for increasing overtones) upon contacting a model hydrophilic surface; A: PGPR 4175; B : PGPR 4110.



**Figure 3.** Adsorbed mass of 3rd, 5th, 7th and 9th overtones (with fading colours for increasing overtones) as a function of time for PGPR 4175 (A) and PGPR 4110 (B) upon contacting a model hydrophilic surface. The adsorbed PGPR film on the model hydrophilic surface is a homogeneous rigid film, because the changes of dissipation are smaller than  $1 \times 10^{-6}$  and the changes of frequency and dissipation are independent of overtones (Figure 2) [3, 4]. So, the Sauerbrey model ( $\Delta m = -C \cdot (\Delta f/n)$ ,  $C = 17.7 \text{ ng cm}^{-2} \text{ Hz}^{-1}$ ) was used to calculate adsorbed mass [5, 6].

## DISCUSSION/ CONCLUSION

- No obvious difference of fatty acid composition between four PGPRs (Figure 1A). Higher content of triglycerol, tetraglycerol, pentaglycerol and hexaglycerol in PGPR 4175 and PGPR 4150, while higher content of glycerol and diglycerol in PGPR 4125 and PGPR 4110 (Figure 1B).
- Lower dynamic interfacial tension of the PGPR 4175 and PGPR 4150 films than that of the PGPR 4125 and PGPR 4110 films (Table 1). No significant difference of dynamic dilatational elastic and viscous modulus between four PGPRs (Table 1).
- Lower adsorbed mass of PGPR 4175 from an oily (glyceryl triheptanoate) solution on a model hydrophilic surface (Figure 3).
- Lower adsorbed PGPR concentration in PGPR 4175-stabilized W/O emulsion (Table 2). Higher adsorbed tri-, tetra- and pentaglycerol concentration in PGPR 4175-stabilized W/O emulsion and higher adsorbed di-, and triglycerol concentration in PGPR 4110-stabilized W/O emulsion (Table 2).
- ❖ These results suggest that PGPR 4175 has a stronger interfacial activity than the other three PGPRs. This indicates that PGPR molecular species with a large hydrophilic headgroup possess a higher adsorption capacity (As shown in Figure 4). Thus, the surface activity of PGPR may be improved by optimizing its polyglycerol composition, thereby reducing the required amount of PGPR.



**Figure 4.** Schematic illustration of the adsorption behavior of PGPR 4175 and PGPR 4110 at the oil-water interface (A), and on a mercaptoethanol modified gold surface (B).

## REFERENCES

[1] Price et al. (2022), LWT, 165, 113704; [2] Paximada et al. (2021), Journal of Food Processing and Preservation, 45(9), e15757; [3] Swana et al. (2022), Membranes, 12, 558; [4] Saftics et al. (2021), Advances in Colloid and Interface Science, 294, 102431; [5] Li et al. (2021), Food Hydrocolloids, 113, 106489; [6] Li et al. (2022), Food Hydrocolloids, 122, 107074.