Electronic Supporting Information

Extraction and separation of pigments from *Saccharina latissima* using eutectic solvents

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Section S1: Extraction of fucoxanthin - Analysis of COSMO-RS

Here, the binarity affinities of each ES precursor and those of fucoxanthin, defined as the activity coefficient of the component in an equimolar binary mixture, were estimated using COSMO-RS and are illustrated in **Figure S1**. A value of zero represents thermodynamic ideality, while negative or positive values indicate affinity or lack thereof, respectively. As expected, the results reveal a strong affinity between ChCl and water. Interestingly, there is also a significant affinity between LevA and ChCl. Despite its hydrophobicity, menthol exhibits the greatest repulsion (or least affinity) with ChCl rather than water. Thus, eutectic solvents based on ChCl are expected to be immiscible with those based on menthol. Furthermore, fucoxanthin shows significant repulsion with ChCl, thermodynamic ideality with menthol, and slight affinity with LevA. As such, the extraction of fucoxanthin is expected to be more favorable towards Ment-rich ES, non-existent towards aqueous ChCl, and facilitated by the presence of LevA.

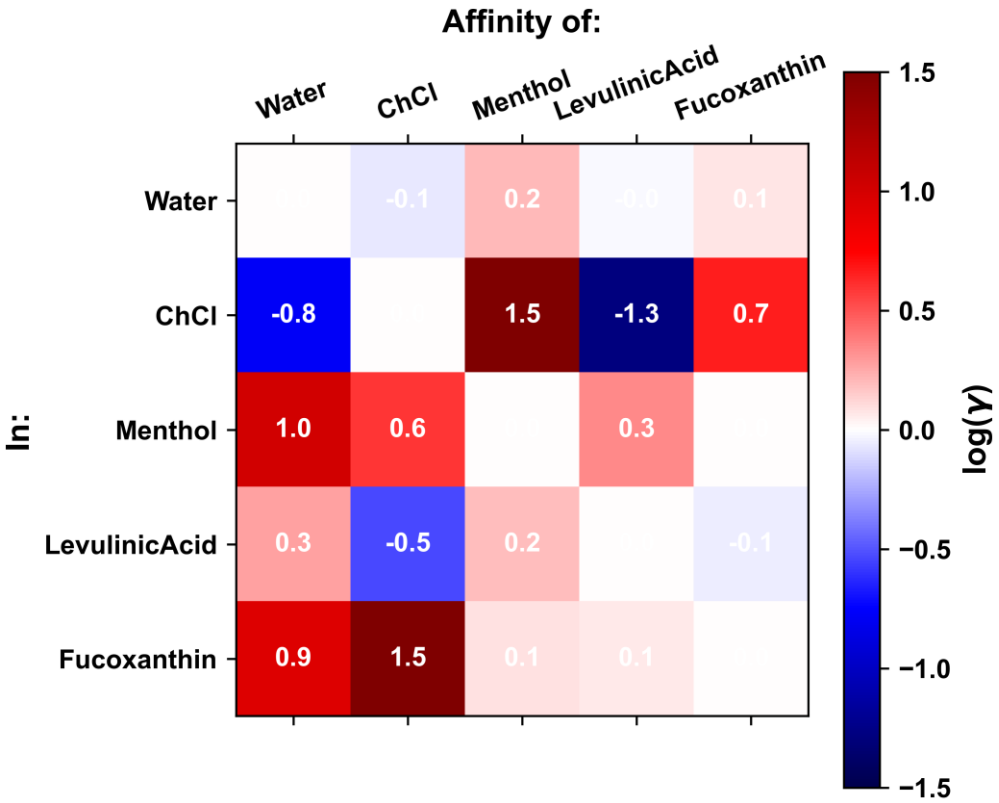

Figure S1. Binary affinities of the ES components used in this work, expressed as the natural logarithm of the COSMO-RS-predicted activity coefficient of the column solutes in the row solvents at equimolar compositions.

Section S2: LLE system and partition of fucoxanthin - Analysis of COSMO-RS

The compositions of the hydrophobic and hydrophilic eutectic solvents after liquid-liquid equilibrium were predicted using COSMO-RS and are reported in Table S1. This considers that the hydrophobic ES (α) is a 1:1 mole ratio mixture of Ment and LevA with 20% (v/v) of added water ($x^{\alpha}_{Water} = 0.65$, $x^{\alpha}_{Ment} = 0.17$, and $x^{\alpha}_{LevA} = 0.17$) and that the hydrophilic ES (β) is a 1:2 mole ratio mixture of ChCl and LevA ($x^{\beta}_{ChCl} = 0.33$ and $x^{\beta}_{LevA} = 0.33$). After equilibrium, note how there is little ChCl in the hydrophobic phase and, likewise, little Ment in the hydrophilic phase. Moreover, water and LevA migrate from the hydrophobic to the hydrophilic phase. Finally, equilibrium is established with a much larger hydrophilic than hydrophobic phase, as a 1:1 v/v ratio leads to a final equilibrium where the hydrophilic phase represents 87% of the system.

Table S1. Compositions of the hydrophobic and hydrophilic ES after thermodynamic equilibrium, predicted using COSMO-RS at 298 K.

Ratio β/α (v/v)	Hydrophobic ES at Equilibrium				Hydrophilic ES at Equilibrium				Fraction of β (%mol)
	x^{α}_{Water}	x^{α}_{ChCl}	x^{α}_{Ment}	x^{α}_{LevA}	x^{β}_{Water}	x^{β}_{ChCl}	x^{β}_{Ment}	x^{β}_{LevA}	
1	0.163	0.002	0.677	0.159	0.489	0.122	0.030	0.358	87%
2	0.108	0.003	0.743	0.146	0.354	0.172	0.040	0.434	93%
3	0.081	0.003	0.774	0.142	0.278	0.201	0.045	0.476	96%
4	0.067	0.003	0.791	0.140	0.229	0.219	0.048	0.503	98%
5	0.055	0.003	0.803	0.139	0.195	0.232	0.050	0.522	>99.9%

The partition coefficients of fucoxanthin were estimated using COSMO-RS across the LLE compositions listed in Table S1 and are reported in Table S2. These partition coefficients remain relatively constant regardless of the initial ES ratio and are favourable towards the hydrophobic ES. Thus, the experimental observation reported in Figure 4 where fucoxanthin progressively partitions into the hydrophilic phase with increasing β/α ratio relates to the migration of water and LevA to the hydrophilic ES. This migration results in an increased volume and enhanced capacity of the hydrophilic phase to solvate fucoxanthin.

Table S2. Logarithm of the partition coefficient of fucoxanthin between the hydrophobic (α) and hydrophilic (β) phases, predicted using COSMO-RS at 298 K.

Initial Ratio β/α (v/v)	**log10(P)**
1	-2.6
2	-2.4
3	-2.3
4	-2.2
5	-2.1