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Review

Interaction Parameters for the Formation of Mixed Micelles and Partitioning of Solutes in Them: A Review

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Abstract: When two or more surfactants are mixed, the critical micelle concentration and solubilization capability are changed, and a careful selection of the combination promotes the micelle formation and enhances the solubilizing capability. Thus, understanding the mechanism behind the phenomena is essential for controlling the physical properties of the mixed micelle. The interaction parameters β and B that describe the formation of mixed micelles and their partitioning of solutes, respectively, were proposed by Treiner four decades ago. In this work, data on the formation and partitioning in binary surfactant systems were collected. Although the data on the parameters β and B for polar solutes and theoretical development are still insufficient, the directions of research to acquire an in-depth understanding of the formation and partitioning of the mixed micelle are proposed.

Keywords: surfactant; mixed micelle; regular solution theory

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1. Introduction

Spherical micelles and vesicles can be applied to drug delivery, cancer therapy, and the pseudostationary phase for separation [1–4]. When two or more surfactants are mixed, the critical micelle concentration (CMC) and solubilization capability are changed, and a careful selection of the combination promotes the micelle formation and enhances the solubilizing capability [5–9]. Thus, understanding the mechanism behind the phenomena is essential for controlling the physical properties of the mixed micelle.

The micelle formation of the mixed micelle can be described by the regular solution theory [5,6], more appropriately called the simple mixture model, established by Rubingh and coworkers [10]. The theory uses the interaction parameter β for a binary mixture of surfactants. A negative value β means synergistic interaction, while a positive value indicates an antagonistic interaction. The CMC of the binary mixed micelle at an arbitrary composition can be appropriately described using the CMC of the pure surfactant and β [11].

An equation can also describe the partition coefficients of the mixed micelle based on the regular solution theory. O'Connell and Prausnitz originally derived the equation to calculate the solubility of inert gases in a binary solvent [12,13], and Treiner and coworkers applied the theory to the mixed micelle system with minor modifications [14–16]. The equations have been used for various mixed micelle systems [5,6,15–24].

Treiner and coworkers carefully categorized binary surfactants and solute systems and concluded a linear relation between the interaction parameters β and B when an appropriate combination of binary surfactants (nine examples) and polar solute was used [16,21]. The exceptions are nonpolar solutes, nonionic surfactants with long oxyethylene chains, and amphiphilic compounds such as yellow OB that undergo aggregation [21]. It has been four decades since Treiner reported the relation and many binary mixed micelle systems partitioning various solutes. In addition, there has been progress in modeling mixed micelle [10,25–28] and mixed solvent solubility [29,30]. These progresses may enforce the claim made by Treiner or help to develop a more general model that explains the relation between β and B.

In this work, data on the formation and partitioning in binary surfactant systems were collected. The summarized data set may reveal the relationship between the two parameters.

2. Theories for the Formation and Partitioning of the Mixed Micelle

According to the regular solution theory, the CMC of the binary mixed micelle of surfactants 1 and 2 can be estimated using an interaction parameter (β) and the CMC of pure surfactants [5,6]. The parameter β is related to net (pairwise) interactions among surfactants 1 and 2 in the binary mixed micelle as $\beta = N(W_{11} + W_{22} - 2W_{12})/RT$, where N is Avogadro's number, W_{11}, W_{22}, and W_{12} are the pairwise interaction energies between surfactants 1 and 2 in the micelles, R is the gas constant, and T is the temperature. If $\beta > 0$, then a repulsive (antagonistic) interaction is taking place between two surfactants, while $\beta < 0$ indicates an attractive (synergistic) interaction between the surfactants. The value of $\beta = 0$ indicates the ideal mixture of surfactants. Experimentally, only the mole fraction of total surfactant 1 (a_1) and surfactant 2 (a_2) are known, where $a_1 + a_2 = 1$ (see Figure 1).

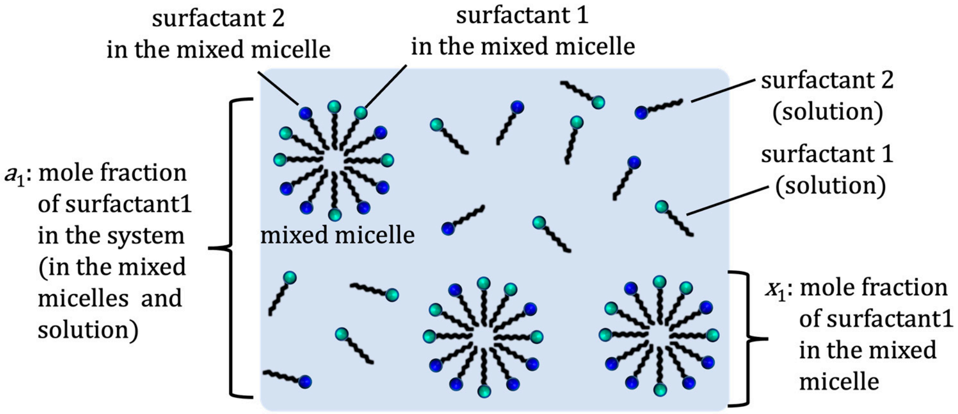

Figure 1. Definition of mole fractions a_1 and x_1.

The regular solution theory relates the mole fraction of surfactant 1 (x_1) in the binary mixed micelle (Figure 1), the CMC of the pure surfactant 1 (C_1) and binary mixed micelle (C_{12}), and the mole fraction of surfactant 1 in total mixed surfactants (a_1) as follows.

$$\beta = \frac{\ln[a_1 C_{12}/(x_1 C_1)]}{(1-x_1)^2} \tag{1}$$

Since x_1 is the mole fraction of surfactant in the binary mixed micelle 1, $x_2 = 1 - x_1$ holds. If C_1, the CMC of the pure surfactant 2 (C_2), C_{12}, and a_1 are known, the unknown variable x_1 can be calculated by iteratively solving the following equation for x_1.

$$x_1{}^2 \ln\left[\frac{a_1 C_{12}}{x_1 C_1}\right] = (1-x_1)^2 \ln\left[\frac{a_2 C_{12}}{(1-x_1)C_2}\right] \tag{2}$$

Experimentally, the x_1 value is determined using ultrafiltration [31–34], small angle neutron scattering [35,36], or a combination of ion-selective electrodes and ultraviolet-visible absorption spectroscopy (UV–Vis) [37]. The theoretical x_1 value obtained using the regular solution theory qualitatively agrees with the experiment [33,35]. According to the theory, x_1 (= $1 - x_2$) at the CMC can be estimated by the following equation:

$$x_1 = \frac{a_1 C_2 f_2 x_2}{a_2 C_1 f_1} \tag{3}$$

$$f_1 = \exp\left[\beta(1-x_1)^2\right] \tag{4}$$

$$f_2 = \exp\left[\beta {x_1}^2\right] \tag{5}$$

where f_1 and f_2 are activity coefficients of the surfactants 1 and 2, respectively.

The values of x_1 and x_2 (= $1 - x_1$) above the CMC can be calculated using the following equation:

$$x_1 = \frac{a_1 C_{\text{total}}}{C_1 f_1 - C_2 f_2 + a_2 C_{\text{total}} / x_2} \tag{6}$$

where C_{total} is the total surfactant concentration. The concentration of surfactants 1 and 2 in the mixed micelles ($C_{1,\text{M}}$ and $C_{2,\text{M}}$, respectively) with the CMC of C_{12} can be expressed as follows:

$$C_{1,\text{M}} = x_1 (C_{\text{total}} - C_{12}) \tag{7}$$

$$C_{2,\text{M}} = x_2 (C_{\text{total}} - C_{12}) \tag{8}$$

By defining synergism as "micelle formation at total mixed surfactant concentrations in the solution phase lower than the CMCs of both surfactants in the mixture", Hua and Rosen derived the second condition for synergism ($|\ln(C_1/C_2)| < |\beta|$) in addition to the first condition of $\beta > 0$ [38]. Likewise, they derived the second condition of antagonism as $|\ln(C_1/C_2)| < \beta$ in addition to the first criterion ($\beta > 0$).

O'Connell and Prausnitz derived the regular solution theory for Henry's coefficient of the gas in the mixed solvent [12,13]. Treiner and coworkers applied the theory to the partition coefficients of the mixed micelle for solutes with slight modification as follows [14,15,20,21]:

$$\ln K_{12} = x_1 \ln K_1 + (1 - x_1) \ln K_2 + B x_1 (1 - x_1) \tag{9}$$

where K_{12}, K_1, and K_2 are partition coefficients of a binary mixed micelle of surfactant composed of 1 and 2, pure micelle composed of surfactant 1, and that of surfactant 2, respectively. The value B is the interaction parameter and should be the same as β; however, they point out that assuming $\beta = B$ is quantitatively not entirely satisfactory, even in the case of nonpolar gases in simple liquid mixtures, because of the omission of the contribution of the solute in the interaction parameter [15]. Therefore, they determined B separately from β, including all surfactant/surfactant and solute/surfactant interaction terms. Using the experimental data of the partition coefficient and CMC value, Treiner and colleagues proposed the following empirical equation to relate the values of β and B [16]:

$$B = 0.194 + 0.343\beta \tag{10}$$

However, the meaning of B remained ambiguous. Recently, the author and coworkers proposed a relation [28].

$$B = \beta - \beta_{123} \tag{11}$$

β_{123} is an additional interaction parameter involving the two surfactants (surfactant 1 and 2) and one solute (regarding the solute as the third component, surfactant 3, of the mixed micelle). The parameter can be determined by measuring the CMC of the ternary system (two surfactants and a solute), the method of which is presented in the reference [28]. The information gives the change in the interaction parameter specific to the ternary system. Still, the evaluation of parameter β_{123} requires evaluating the CMC in a ternary system and determining partitioning constants, treating one of the components as the solute. Such an example is limited so far [28], and further investigation is required to verify and find a relation between β_{123} and β.

3. Results

Figure 2 summarizes the solute molecules used for determining the partition coefficients of the binary mixed micelles. Most of the solutes have a chromophore such as an aromatic ring. This could be because UV–Vis measurement is employed to evaluate the partition coefficient. In the case of UV–Vis measurement, the solute is the chromophore used in the spectroscopy assays. Since the relationship between B and β depends on the polarity of the solutes, the values of the octanol–water partition coefficient (log$P$) are estimated using ChemDraw 21.0.0.28.

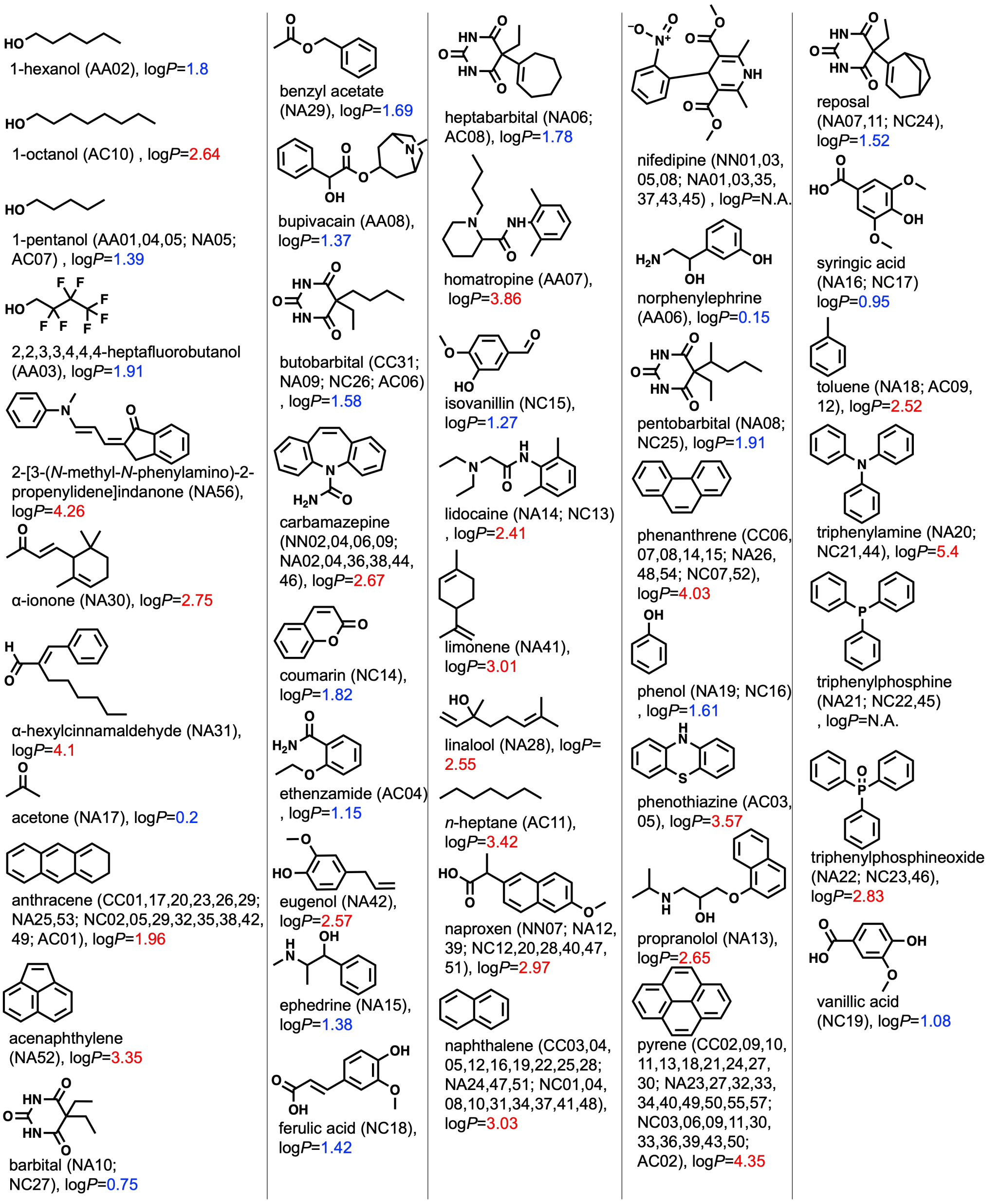

Figure 2. Chemical structures of solutes in alphabetical order. The system IDs are shown in parentheses. Values of log$P$ were estimated using ChemDraw 21.0.0.28 and colored with blue if log$P \geq 1.96$ (=log$P$ of anthracene) and red if $1.96 >$ log$P$. See “Discussion“ for the reason to choose the value.

Figures 3–6 show the chemical structures of surfactants used for studying the formation and partitioning of mixed micelles from the surfactants reported by researchers. The anionic surfactants include perfluoroalkyl surfactant LFOS, which shows an antagonistic interaction with surfactants bearing alkyl groups, resulting in a positive β value. For the Gemini surfactants [39] and bile salts [40], reviews on the interaction parameter β have recently been reported. Some compounds were excluded due to their undefined structure [41].

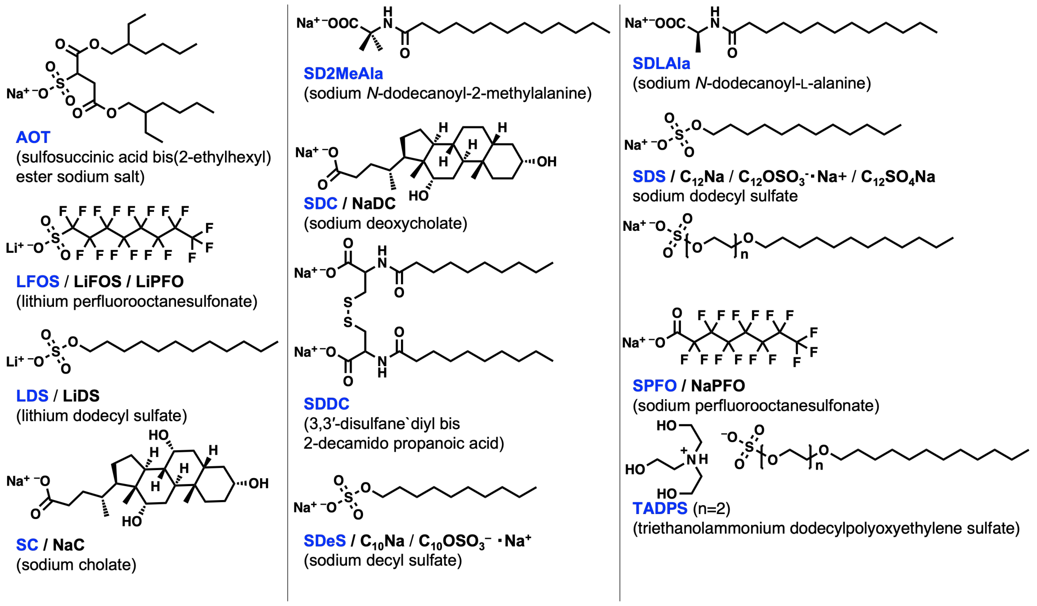

Figure 3. Chemical structures of anionic surfactants. The compound names used in this work are colored in blue. The other abbreviations appear in the literature.

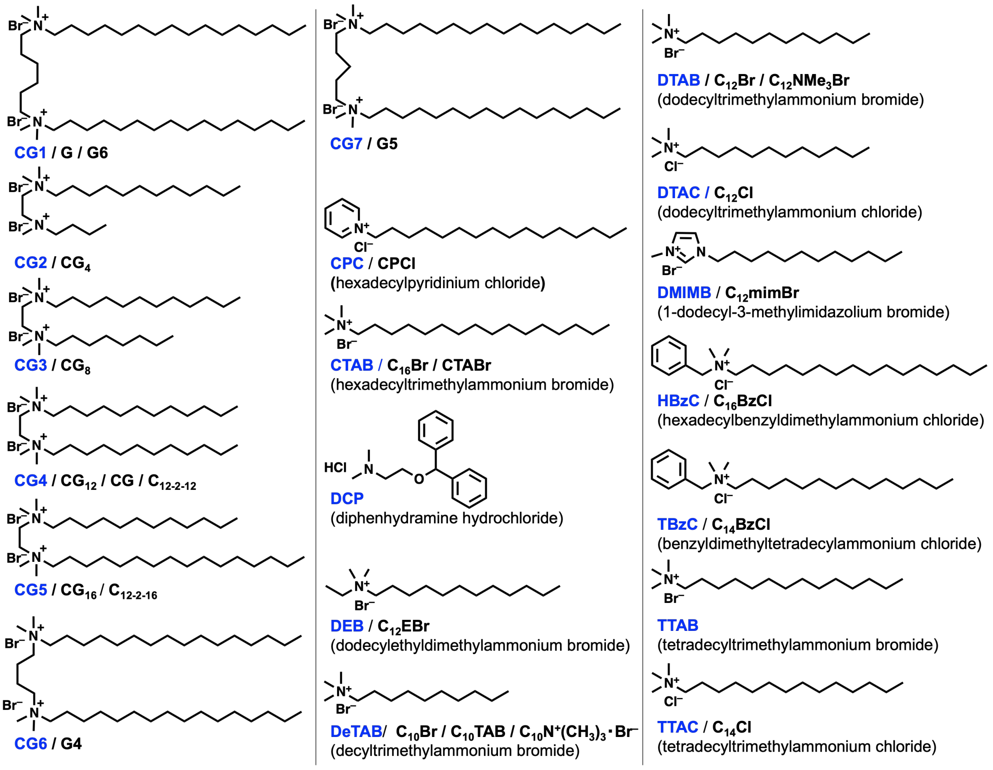

Figure 4. Chemical structures of cationic surfactants. The compound names used in this work are colored in blue. The other abbreviations appear in the literature.

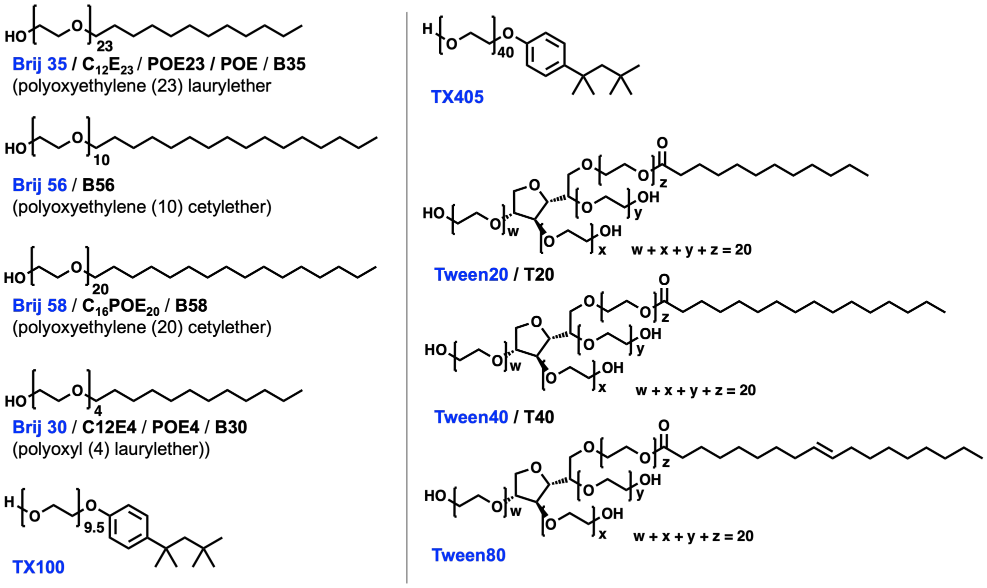

Figure 5. Chemical structures of nonionic surfactants. The compound names used in this work are colored in blue. The other abbreviations appear in the literature.

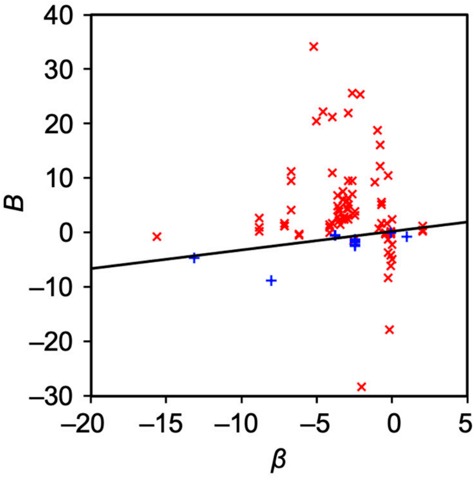

Figure 6. The relation between β and B. Red cross indicates nonpolar solute with $\log P > 1.96$, while blue cross indicates polar solutes with $1.96 \geq \log P$. The black line is drawn based on Equation (10).

Tables 1–6 are the previously reported mixed micelle systems. Some studies of partitioning solutes using binary micelles did not provide information on the interaction parameter B. Such cases may require a further analysis based on their data or reproducing the experiment may be beneficial to note. In the literature, various methods were employed to analyze the micellar formation and partitioning of the solutes: surface tension (ST); fluorescence (FL); nuclear magnetic resonance (NMR); conductance (CD); gas chromatography (GC); UV–Vis; micellar enhanced ultrafiltration (MEUF); micellar liquid chromatography (MLC); high-performance liquid chromatography (HPLC); and micellar electrokinetic chromatography (MEKC). The most popular method to determine is ST, while the most popular method to determine B is UV–Vis measurement. MEKC was recently introduced and related to the regular solution theory to analyze the mixed micelle [27]. The method can determine not only the partition coefficients [42,43] but also the kinetics of the solute permeation of the micelles [44,45]. Various methods were used to analyze the interaction parameters, but comparative studies of the different methods to determine the interaction parameter for the partitioning of solutes using the same surfactants and solute combination have not been reported to the best of the author's knowledge. The systems of the binary micelles are categorized with the types of composing surfactants: anionic/anionic (AA); cationic/cationic (CC); nonionic/nonionic (NN); nonionic/anionic (NA); nonionic/cationic (NC); and anionic/cationic (AC) surfactants. In general, the interaction between the two surfactants is mainly due to electrostatic forces, and the strength of the attractive electrostatic interaction parameter β decreases in the order of AN > NA ≈ NC > AA ≈ CC ≈ NN surfactants [11,27].

Table 1. The interaction parameters of mixed micelles composed of anionic/anionic surfactants.

		Measurement of β				**Measurement of B**				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
AA01	**LFOS/LDS**	2.2	23	NMR	[19]	1-pentanol	–	25	GC	[20]
AA02	**LFOS/LDS**	2.2	23	ST	[46]	1-hexanol	–	–	GC	[46]
AA03	**LFOS/LDS**	2.2	23	ST	[46]	2,2,3,3,4,4,4-heptafluorobutanol	–	–	GC	[46]
AA04	**SPFO/SDeS**	1.8	23	NMR	[19]	1-pentanol	–	25	GC	[14]
AA05	**SPFO/SDS**	2.1	23	NMR	[19]	1-pentanol	–	25	GC	[14]
AA06	**SD2MeAla/SDLAla**	−0.090	25	ST	[27]	norphenylephrine	0.20	25	MEKC	[27]
AA07	**SD3MeAla/SDLAla**	−0.090	25	ST	[27]	homatropine	0.16	25	MEKC	[27]
AA08	**SD4MeAla/SDLAla**	−0.090	25	ST	[27]	bupivacaine	0.21	25	MEKC	[27]

Table 2. The interaction parameters of mixed micelles composed of cationic/cationic surfactants.

		Measurement of β				**Measurement of B**				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
CC01	**CG1/CPC**	−0.8	25	ST	[47]	anthracene	12.25	25	UV–Vis	[47]
CC02	**CG1/CPC**	−0.8	25	ST	[47]	pyrene	16.28	25	UV–Vis	[47]
CC03	**CG4/CG2**	−0.08	25	ST	[24]	naphthalene	−4.71	25	UV–Vis	[24]
CC04	**CG4/CG2**	−0.08	25	ST	[24]	naphthalene	2.58	25	UV–Vis	[24]
CC05	**CG4/CG2**	−0.08	25	ST	[24]	naphthalene	−2.13	25	UV–Vis	[24]
CC06	**CG4/CG3**	−0.14	25	ST	[24]	phenanthrene	−5.92	25	UV–Vis	[24]
CC07	**CG4/CG3**	−0.14	25	ST	[24]	phenanthrene	0.25	25	UV–Vis	[24]
CC08	**CG4/CG3**	−0.14	25	ST	[24]	phenanthrene	−4.01	25	UV–Vis	[24]
CC09	**CG4/CG5**	−0.29	25	ST	[24]	pyrene	−3.67	25	UV–Vis	[24]
CC10	**CG4/CG5**	−0.29	25	ST	[24]	pyrene	−8.22	25	UV–Vis	[24]
CC11	**CG4/CG5**	−0.29	25	ST	[24]	pyrene	10.5	25	UV–Vis	[24]
CC12	**CG4/CG5**	−0.38	25	ST	[48]	naphthalene	–	25	UV–Vis	[48]
CC13	**CG4/CG5**	−0.38	25	ST	[48]	pyrene	–	25	UV–Vis	[48]
CC14	**CG4/CPC**	−0.32	25	ST	[47]	phenanthrene	−1.06	25	UV–Vis	[49]
CC15	**CG4/DTAB**	−0.22	25	ST	[47]	phenanthrene	−17.6	25	UV–Vis	[49]
CC16	**CG6/CTAB**	−8.91	30	ST	[50]	naphthalene	1.04	30	UV–Vis	[50]
CC17	**CG6/CTAB**	−8.91	30	ST	[50]	anthracene	0.45	30	UV–Vis	[50]
CC18	**CG6/CTAB**	−8.91	30	ST	[50]	pyrene	2.73	30	UV–Vis	[50]
CC19	**CG7/CTAB**	−7.2	30	ST	[50]	naphthalene	1.26	30	UV–Vis	[50]
CC20	**CG7/CTAB**	−7.2	30	ST	[50]	anthracene	1.92	30	UV–Vis	[50]
CC21	**CG7/CTAB**	−7.2	30	ST	[50]	pyrene	1.39	30	UV–Vis	[50]
CC22	**CG1/CTAB**	−6.76	30	ST	[50]	naphthalene	4.17	30	UV–Vis	[50]
CC23	**CG1/CTAB**	−6.76	30	ST	[50]	anthracene	9.73	30	UV–Vis	[50]
CC24	**CG1/CTAB**	−6.76	30	ST	[50]	pyrene	11.21	30	UV–Vis	[50]
CC25	**CTAB/HBzC**	−0.47	25	ST	[51]	naphthalene	−0.03	25	UV–Vis	[51]
CC26	**CTAB/HBzC**	−0.47	25	ST	[51]	anthracene	0.01	25	UV–Vis	[51]
CC27	**CTAB/HBzC**	−0.47	25	ST	[51]	pyrene	−0.1	25	UV–Vis	[51]
CC28	**DTAB/DEB**	−0.14	25	ST	[51]	naphthalene	0.04	25	UV–Vis	[51]
CC29	**DTAB/DEB**	−0.14	25	ST	[51]	anthracene	−0.25	25	UV–Vis	[51]
CC30	**DTAB/DEB**	−0.14	25	ST	[51]	pyrene	−0.2	25	UV–Vis	[51]
CC31	**TTAC/TBzC**	0.92	–	CD	[21]	butobarbital	−0.60	25	UV–Vis	[21]

Table 3. The interaction parameters of mixed micelles composed of nonionic/nonionic surfactants.

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
NN01	**Brij30/Tween20**	−0.59	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NN02	**Brij30/Tween20**	−0.59	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NN03	**Brij35/Tween20**	−2.67	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NN04	**Brij35/Tween20**	−2.67	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NN05	**Brij56/Tween40**	−1.36	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NN06	**Brij56/Tween40**	−1.36	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NN07	**Brij58/Brij35**	−1.23	25	ST	[53]	naproxen	9.39	25	UV–Vis	[53]
NN08	**Brij58/Tween40**	−2.67	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NN09	**Brij58/Tween40**	−2.67	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]

Table 4. The interaction parameters of mixed micelles composed of nonionic/anionic surfactants.

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
NA01	**Brij30/SC**	−8.77	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA02	**Brij30/SC**	−8.77	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA03	**Brij35/SC**	−7.97	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA04	**Brij35/SC**	−7.97	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA05	**Brij35/SDS**	−2.5	25	ST	[18]	1-pentanol	–	25	GC	[20]
NA06	**Brij35/SDS**	−2.5	25	ST	[18]	heptabarbital	−2.1	25	UV–Vis	[21]
NA07	**Brij35/SDS**	−2.5	25	ST	[18]	reposal	−2.3	–	UV–Vis	[15]
NA08	**Brij35/SDS**	−2.5	25	ST	[18]	pentobarbital	−1.4	–	UV–Vis	[15]
NA09	**Brij35/SDS**	−2.5	25	ST	[18]	butobarbital	−1.7	–	UV–Vis	[15]
NA10	**Brij35/SDS**	−2.5	25	ST	[18]	barbital	−1.1	–	UV–Vis	[15]
NA11	**Brij35/SDS**	−2.5	25	ST	[18]	reposal (0.2 M NaCl)	−1.7	–	UV–Vis	[15]
NA12	**Brij35/SDS**	−2.71	25	ST	[53]	naproxen	25.8	25	UV–Vis	[53]
NA13	**Brij35/SDS**	−2.71	25	ST	[53]	propranolol	–	25	MEUF	[54]
NA14	**Brij35/SDS**	−2.71	25	ST	[53]	lidocaine	–	25	MEUF	[54]
NA15	**Brij35/SDS**	−2.71	25	ST	[53]	ephedrine	–	25	MEUF	[54]
NA16	**Brij35/SDS**	−2.71	25	ST	[53]	syringic acid	–	25	MEUF	[54]
NA17	**Brij35/SDS**	−2.71	25	ST	[53]	acetone	–	25	MEUF	[54]
NA18	**Brij35/SDS**	−2.71	25	ST	[53]	toluene	–	25	MEUF	[54]
NA19	**Brij35/SDS**	−2.71	25	ST	[53]	phenol	–	25	MEUF	[54]
NA20	**Brij35/SDS**	−2.71	25	ST	[53]	triphenylamine	7.09	25	UV–Vis	[23]
NA21	**Brij35/SDS**	−2.71	25	ST	[53]	triphenylphosphine	30.1	25	UV–Vis	[23]
NA22	**Brij35/SDS**	−2.71	25	ST	[53]	triphenyl phosphine oxide	9.69	25	UV–Vis	[23]
NA23	**Brij35/SDS**	−2.6	25	ST	[55]	pyrene	−1.34	25	UV–Vis	[55]
NA24	**Brij35/SDS**	−2.71	25	ST	[53]	naphthalene	–	25	UV–Vis	[56]
NA25	**Brij35/SDS**	−2.71	25	ST	[53]	anthracene	–	25	UV–Vis	[56]
NA26	**Brij35/SDS**	−2.71	25	ST	[53]	phenanthrene	–	25	UV–Vis	[56]
NA27	**Brij35/SDS**	−2.71	25	ST	[53]	pyrene	–	25	UV–Vis	[56]
NA28	**Brij35/SDS**	−2.71	25	ST	[53]	linalool	–	25	UV–Vis	[56]
NA29	**Brij35/SDS**	−2.71	25	ST	[53]	benzyl acetate	–	25	UV–Vis	[56]
NA30	**Brij35/SDS**	−2.71	25	ST	[53]	α-ionone	–	25	UV–Vis	[56]
NA31	**Brij35/SDS**	−2.71	25	ST	[53]	α-hexylcinnamaldehyde	–	25	UV–Vis	[56]
NA32	**Brij35/SLE1S**	2.01	–	FL	[57]	pyrene	1.33	–	UV–Vis	[57]
NA33	**Brij35/SLE2S**	2.01	–	FL	[57]	pyrene	0.68	–	UV–Vis	[57]
NA34	**Brij35/SLE3S**	2.01	–	FL	[57]	pyrene	0.40	–	UV–Vis	[57]
NA35	**Brij56/SC**	−10.42	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA36	**Brij56/SC**	−10.42	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA37	**Brij58/SC**	−9.11	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA38	**Brij58/SC**	−9.11	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA39	**Brij58/SDS**	−5.07	25	ST	[53]	naproxen	20.6	25	UV–Vis	[53]
NA40	**Brij58/SDS**	−2.1	25	ST	[55,58]	pyrene	−28.3	25	UV–Vis	[55]
NA41	**Brij58/SDS**	−6.18	30	ST	[55,58]	limonene	−0.02	30	UV–Vis	[59]
NA42	**Brij58/SDS**	−6.18	30	ST	[55,58]	eugenol	−0.40	30	UV–Vis	[59]

Table 4. *Cont.*

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
NA43	**Tween20/SC**	−8.08	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA44	**Tween20/SC**	−8.08	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA45	**Tween40/SC**	−8.9	25	ST	[52]	nifedipine	–	25	UV–Vis	[52]
NA46	**Tween40/SC**	−8.9	25	ST	[52]	carbamazepine	–	25	UV–Vis	[52]
NA47	**Tween80/SDDC**	−4.2	30	ST	[60]	naphthalene	–	30	UV–Vis	[60]
NA48	**Tween80/SDDC**	−4.2	30	ST	[60]	phenanthrene	–	30	UV–Vis	[60]
NA49	**Tween80/SDDC**	−4.2	30	ST	[60]	pyrene	–	30	UV–Vis	[60]
NA50	**TX100/SDS**	−1.5	25	ST	[55]	pyrene	−2.81	25	UV–Vis	[55]
NA51	**TX100/SDS**	−1.5	25	ST	[55]	naphthalene	–	25	UV–Vis	[56]
NA52	**TX100/SDS**	−1.5	25	ST	[55]	acenaphthylene	–	25	UV–Vis	[56]
NA53	**TX100/SDS**	−1.5	25	ST	[55]	anthracene	–	25	UV–Vis	[56]
NA54	**TX100/SDS**	−1.5	25	ST	[55]	phenanthrene	–	25	UV–Vis	[56]
NA55	**TX100/SDS**	−1.5	25	ST	[55]	pyrene	–	25	UV–Vis	[56]
NA56	**TX100/SDS**	−4.02	30	FL	[61]	2-[3-(*N*-methyl-*N*-phenylamino)-2-propenylidene] indanone	–	30	UV–Vis	[61]
NA57	**TX405/SDS**	−3.5	25	ST	[55]	pyrene	1.54	25	UV–Vis	[55]

Table 5. The interaction parameters of mixed micelles composed of nonionic/cationic surfactants.

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
NC01	**Brij30/DEB**	−3.66	25	ST	[51]	naphthalene	3.15	25	UV–Vis	[51]
NC02	**Brij30/DEB**	−3.66	25	ST	[51]	anthracene	6.96	25	UV–Vis	[51]
NC03	**Brij30/DEB**	−3.66	25	ST	[51]	pyrene	5.03	25	UV–Vis	[51]
NC04	**Brij30/DTAB**	−3.53	25	ST	[51]	naphthalene	2.34	25	UV–Vis	[51]
NC05	**Brij30/DTAB**	−3.53	25	ST	[51]	anthracene	4.11	25	UV–Vis	[51]
NC06	**Brij30/DTAB**	−3.53	25	ST	[51]	pyrene	4.58	25	UV–Vis	[51]
NC07	**Brij35/CG4**	−0.38	25	ST	[47]	phenanthrene	1.8	25	UV–Vis	[49]
NC08	**Brij35/CG4**	−1.27	25	ST	[48]	naphthalene	–	25	UV–Vis	[48]
NC09	**Brij35/CG4**	−1.27	25	ST	[48]	pyrene	–	25	UV–Vis	[48]
NC10	**Brij35/CG5**	−1.42	25	ST	[48]	naphthalene	–	25	UV–Vis	[48]
NC11	**Brij35/CG5**	−1.42	25	ST	[48]	pyrene	–	25	UV–Vis	[48]
NC12	**Brij35/CTAB**	−1.03	25	ST	[53]	naproxen	18.9	25	UV–Vis	[53]
NC13	**Brij35/CTAB**	−1.03	25	ST	[53]	lidocaine	–	25	MLC	[54]
NC14	**Brij35/CTAB**	−1.03	25	ST	[53]	coumarin	–	25	MLC	[54]
NC15	**Brij35/CTAB**	−1.03	25	ST	[53]	isovanillin	–	25	MLC	[54]
NC16	**Brij35/CTAB**	−1.03	25	ST	[53]	phenol	–	25	MLC	[54]
NC17	**Brij35/CTAB**	−1.03	25	ST	[53]	syringic acid	–	25	MLC	[54]
NC18	**Brij35/CTAB**	−1.03	25	ST	[53]	ferulic acid	–	25	MLC	[54]
NC19	**Brij35/CTAB**	−1.03	25	ST	[53]	vanillic acid	–	25	MLC	[54]
NC20	**Brij35/DTAB**	−3.98	25	ST	[53]	naproxen	21.4	25	UV–Vis	[53]
NC21	**Brij35/DTAB**	−3.98	25	ST	[53]	triphenylamine	11	25	UV–Vis	[23]
NC22	**Brij35/DTAB**	−3.98	25	ST	[53]	triphenylphosphine	22.9	25	UV–Vis	[23]
NC23	**Brij35/DTAB**	−3.98	25	ST	[53]	triphenylphosphine oxide	1.87	25	UV–Vis	[23]
NC24	**Brij35/TTAB**	−3.8	25	FL	[22]	reposal	−0.3	–	UV–Vis	[15]
NC25	**Brij35/TTAB**	−3.8	25	FL	[22]	pentobarbital	−0.4	–	UV–Vis	[15]
NC26	**Brij35/TTAB**	−3.8	25	FL	[22]	butobarbital	−0.4	–	UV–Vis	[15]
NC27	**Brij35/TTAB**	−3.8	25	FL	[22]	barbital	−0.3	–	UV–Vis	[15]
NC28	**Brij35/TTAB**	−2.17	25	ST	[53]	naproxen	25.6	25	UV–Vis	[53]
NC29	**Brij56/CG1**	−2.55	25	ST	[47]	anthracene	3.33	25	UV–Vis	[47]
NC30	**Brij56/CG1**	−2.55	25	ST	[47]	pyrene	4.06	25	UV–Vis	[47]
NC31	**Brij58/CG6**	−3.24	30	ST	[50]	naphthalene	2.97	30	UV–Vis	[50]

Table 5. *Cont.*

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
NC32	**Brij58/CG6**	−3.24	30	ST	[50]	anthracene	2.65	30	UV–Vis	[50]
NC33	**Brij58/CG6**	−3.24	30	ST	[50]	pyrene	2.57	30	UV–Vis	[50]
NC34	**Brij58/CG7**	−4.16	30	ST	[50]	naphthalene	1.05	30	UV–Vis	[50]
NC35	**Brij58/CG7**	−4.16	30	ST	[50]	anthracene	1.51	30	UV–Vis	[50]
NC36	**Brij58/CG7**	−4.16	30	ST	[50]	pyrene	0.2	30	UV–Vis	[50]
NC37	**Brij58/CG1**	−0.75	30	ST	[50]	naphthalene	1.9	30	UV–Vis	[50]
NC38	**Brij58/CG1**	−0.75	30	ST	[50]	anthracene	5.69	30	UV–Vis	[50]
NC39	**Brij58/CG1**	−0.75	30	ST	[50]	pyrene	5.31	30	UV–Vis	[50]
NC40	**Brij58/CTAB**	−2.99	25	ST	[53]	naproxen	22	25	UV–Vis	[53]
NC41	**Brij58/CTAB**	−3.05	25	ST	[51]	naphthalene	2.86	25	UV–Vis	[51]
NC42	**Brij58/CTAB**	−3.05	25	ST	[51]	anthracene	4.56	25	UV–Vis	[51]
NC43	**Brij58/CTAB**	−3.05	25	ST	[51]	pyrene	5.96	25	UV–Vis	[51]
NC44	**Brij58/CTAB**	−2.99	25	ST	[53]	triphenylamine	6.44	25	UV–Vis	[23]
NC45	**Brij58/CTAB**	−2.99	25	ST	[53]	triphenylphosphine	23.6	25	UV–Vis	[23]
NC46	**Brij58/CTAB**	−2.99	25	ST	[53]	triphenylphosphine oxide	9.62	25	UV–Vis	[23]
NC47	**Brij58/DTAB**	−5.25	25	ST	[53]	naproxen	34.3	25	UV–Vis	[53]
NC48	**Brij58/HBzC**	−2.98	25	ST	[51]	naphthalene	2.46	25	UV–Vis	[51]
NC49	**Brij58/HBzC**	−2.98	25	ST	[51]	anthracene	4.57	25	UV–Vis	[51]
NC50	**Brij58/HBzC**	−2.98	25	ST	[51]	pyrene	5.26	25	UV–Vis	[51]
NC51	**Brij58/TTAB**	−4.61	25	ST	[53]	naproxen	22.4	25	UV–Vis	[53]
NC52	**TX100/CG4**	−0.94	25	ST	[47]	phenanthrene	0.76	25	UV–Vis	[49]

Table 6. The interaction parameters of mixed micelles composed of anionic/cationic surfactants.

		Measurement of β				Measurement of B				
Sys. ID	**Surfactants**	β	T [°C]	**Method**	**Ref.**	**Solute**	B	T [°C]	**Method**	**Ref.**
AC01	**AOT/CG1**	−3.33	25	ST	[47]	anthracene	6.25	25	UV–Vis	[47]
AC02	**AOT/CG1**	−3.33	25	ST	[47]	pyrene	7.69	25	UV–Vis	[47]
AC03	**SC/DMIMB**	−9.03	25	FL	[62]	phenothiazine	–	–	UV–Vis	[62]
AC04	**SDC/DCP**	−3.69	25	ST	[63]	ethenzamide	–	37	HPLC	[63]
AC05	**SDC/DMIMB**	−9.95	25	FL	[62]	phenothiazine	–	–	UV–Vis	[62]
AC06	**SDeS/DeTAB**	−13.2	23	ST	[6]	butobarbital	−4.5	25	UV–Vis	[21]
AC07	**SDeS/DeTAB**	−13.2	23	ST	[6]	1-pentanol	–	25	GC	[20]
AC08	**SDS/DTAC**	−8.09	25	CD	[64]	heptabarbital	−8.8	25	UV–Vis	[21]
AC09	**TADPS/CPC**	−18.3	25	ST	[65]	toluene	–	25	UV–Vis	[66]
AC10	**TADPS/CTAB**	−15.7	25	ST	[65]	1-octanol	−0.61	25	UV–Vis	[66]
AC11	**TADPS/CTAB**	−15.7	25	ST	[65]	*n*-heptane	–	25	UV–Vis	[66]
AC12	**TADPS/CTAB**	−15.7	25	ST	[65]	toluene	–	25	UV–Vis	[66]

4. Discussion

The empirical equation that relates the interaction parameters β and B is reported by Treiner (Equation (10)). However, Treiner and coworkers claim that it is necessary to distinguish the nonpolar solutes from the polar solutes to apply the equation [21]. The nonpolar solutes tend to show positive B values even if β is negative, which often contradicts the prediction by Equation (10). They explain the discrepancy by the location of the interacting nonpolar solutes in the mixed micelle. In the mixed micelle, the nonpolar solute should be in the hydrocarbon core, and the enthalpy of the mixing should be close to zero following ideal mixing $\beta \sim 0$, which gives a positive value for Equation (10). The other reason for the different behavior of nonpolar solvents from polar solvents is the change in the aggregation number. Some nonpolar solutes increase the aggregation number and induce a structural change from spherical to cylindrical micelles. The analysis is sensitive to the change, resulting in an increase in micellar solubilization, because the Laplace pressure is smaller for a cylinder than a sphere.

In this work, the values of log$P$ are used just for the qualitative evaluation of the polarity of the solutes. More accurate and/or sophisticated parameters, such as experimentally determined log$P$ and solubility parameters, could help analyze the relationship between β and B, which may be performed in the future work. The purpose of introducing the log$P$ in this work is to distinguish polar and nonpolar solutes. Treiner and coworkers report that the borderline that may not follow Equation (10) is benzene (log$P$ = 2.03) and 1-octanol (log$P$ = 2.64). To safely apply Equation (10), log$P$ = 2.03 was chosen as the border at the start. However, it was found that the log$P$ value for anthracene was lower than benzene. Most of the B for anthracene is positive while β is negative (CC01, CC17, CC20, CC23, CC26, NC02, C05, C29, C32, C35, C38, C42, C49, and AC01), although there is an example with negative β and negative B (CC29). Therefore, the anthracene was regarded as at the borderline; thus, the borderline between polar and nonpolar was determined by the log$P$ of anthracene (1.96). If log$P \geq 1.96$, the solute is regarded as nonpolar, while if $1.96 > \log P$, the solute is regarded as polar. Many reports are complete for nonpolar solutes, and the determination of B for polar solutes is limited. Although the work and linear relation between β and B reported by Treiner and coworkers [16,21] is widely recognized [7,10,15,24,27,28,47,49,57,59,61,62,67], their model solutes 1-pentanol and barbital and their derivatives have seldom been reported by other researchers. The few reports on the polar solutes could be because the most popular measurement method is the UV–Vis measurement, and 1-pentanol without chromophore cannot be used. The barbital derivatives are the other solutes, but their structures are uncommon. The remaining polar systems were reported by Treiner and coworkers (CC31, NC24–27, NA06–11, AC06, and AC08) [15,21] and the author (AA06 and AA08) [27]. Figure 6 illustrates the distribution of β and B for nonpolar (red) and polar (blue) solutes. For the polar solutes, the β vs. B relation shows a linear relation, as pointed out by Treiner. The two examples AA06 and AA08 [27] were added to Treiner's systems. Although the methods of determining the parameter B are GC, UV–Vis, and MEKC, the linearity is maintained, confirming the equation's generality. However, it should be said that the amount of the data is insufficient, and the validity of Equation (10) remains questioned. Since the solute is polar, it is expected that there is no strong interaction between the surfactant and solute. However, even for the polar solute, $\beta = B$ does not hold, which could be because the effect of the polar solute is not negligible. Yet, the empirical relation Equation (10) implies that there is some relationship behind the two interaction parameters. More data are accumulated for the nonpolar system. The authors reported many nonpolar systems with various kinds of solutes, but most of them show a negative β and positive B, as pointed out by Treiner and coworkers.

Further study will be necessary to enrich the understanding of formation and partitioning in mixed micelle systems. The author proposes directions in this field to acquire an in-depth understanding of the formation and partitioning of the mixed micelle. First, for nonpolar solutes, more examples are necessary to test Equation (10). The reports on the formation and partitioning of polar solutes using mixed micelles are rare, and the validity of the equation remains questioned. A reanalysis of reported systems with polar solutes may increase the amount of data. However, the effort will not give information for nonpolar solutes, and the applicability of the equation remains limited. Therefore, uncovering parameters governing the relationship between β and B should be clarified. Second, the accumulation of analyzed examples of surfactant/surfactant/solute systems is necessary to experimentally verify Equation (11) reported by the author [28] based on "the quasi-simple ternary mixture" model. The equation gives a clear relation between β and B. Still, the determination of β_{123} requires the analysis of the ternary (two surfactants and a solute) system regarding the solute as the third component of the micellar phase. The example for the LDS (surfactant)/LFOS (surfactant)/*n*-hexanol (solute) ternary system was reported [28]. The resulting values of β and β_{123} were 1.7 and -0.11, and B was calculated as 1.8, giving a slightly better fit for the prediction of the partition coefficient, which is encouraging. Regarding the "solute" as a third surfactant is uncommon, but such treatment may give helpful information to uncover the relation between β and B. Third, finding a new approach to relate β and B other than Equations (10) and (11) could be possible, taking similar models developed for cosolvent solubility by Joubyan [29,30].

5. Conclusions

In this work, information on the formation and partitioning of solutes using various mixed micelle systems is tabulated. Researchers report different cationic, anionic, and nonionic surfactants, and their combinations. The log$P$ of anthracene was used to distinguish polar and nonpolar solutes. It was found that the information of parameter B is limited, and the Equation (10) proposed by Treiner cannot be verified. Three research directions for the formation and partitioning in mixed micelle systems are offered to acquire an in-depth understanding of the phenomenon. First, obtaining more data on the partitioning of polar solutes will be necessary. Second, an analysis of the ternary (two surfactants and a solute) system regarding the solute as the third component of the micellar phase will increase the information of β_{123}, which will give the information for surfactant–solute interaction. Third, finding a new theoretical model to relate β and B other than Equations (10) and (11) is important, where the cosolvent solubility by Joubyan could provide a hint.

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