Scientific Journal of Gdynia Maritime University

No. 137/2026, 72–83
ISSN 2657-6988 (online)
ISSN 2657-5841 (printed)
DOI: 10.26408/137.05

Submitted: 10.04.2025
Accept 06.11.2025
Published: 30.03.2026

COMPUTATION OF THE HYDROPHOBICITY PARAMETER (EACN) OF OIL PHASE INGREDIENTS ESSENTIAL FOR PREDICTING PROPERTIES OF MICROEMULSIONS USING THE HLD-NAC THEORY

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Abstract: In this work we describe in detail our novel computation method for predicting the EACN hydrophobicity parameter of hydrophobic compounds, which is used in the HLD equation developed by Salager. The values of EACN obtained in such a way can be applied to predict the phase equilibria in emulsions or microemulsions as well as various physicochemical properties describing these systems by means of the HLD-NAC theory. Application of the HLD-NAC theory as a predictive tool for excipients used in cosmetic or pharmaceutical products may enable the rational design of new products based on emulsions or microemulsions, eliminating in practice the need for many problem-solving efforts and long-term product development. The computed values of the EACN were compared with the experimental data published in the literature for 141 hydrophobic compounds. We have found that the computation method described in this work applied to various hydrophobic compounds leads to the numerical values of the EACN being in a very good agreement with the corresponding experimental values published in the literature. It should be noted, however, that the proposed method is not suitable for predicting EACN values of triglycerides and edible oils.

Keywords: hydrophobicity parameter, fragrance compounds, perfumery raw materials, microemulsions, HLD-NAC theory.

1. INTRODUCTION

Several concepts can be found in the literature that have been used to quantify the hydrophobic nature of oils applied in emulsion or fragrance products. For surfactants used as emulsifiers [Zieliński 2021; Abbott 2026] one can find the idea of the hydrophilic-lipophilic balance (HLB) originally introduced by Griffin (1949) with some modifications [Kawakami 1953; Davies 1957; Heusch 1970; McGowan 1990; Guo,

Rong and Ying 2006; Dong, Hui and Rong 2017], the phase inversion temperature (PIT) method [Shinoda and Sagitani 1978], the critical packing parameter (CPP) concept [Israelachvili, Mitchell and Niham 1976; Israelachvili 2011], the inorganic-organic balance (IOB) method [Fujita 1954], the hydrophilic-lipophilic difference (HLD) concept [Salager et al. 2000] and the HLD-NAC theory [Acosta 2008; 2020; Acosta and Bhakta 2009; AlKhatteb and AlSofi 2021; Bu et al. 2022; Acosta et al. 2025].

In recent years, considerable attention has been paid to the development of more stable surfactant-based emulsions and microemulsions as well as the solubilization of perfumery raw materials in various water-based formulations [Acree Jr et al. 2022]. In each case, the important role in predicting the stability of the formulation is played by the hydrophobicity of hydrophobic components (i.e. oils, fragrances) used in the product. In order to quantify the hydrophobicity of oils, the concept of equivalent alkane carbon numbers (EACN) was introduced by Wade and coworkers [Castellino et at. 1976; Wade et al. 1977]. It is important to characterize oils in surfactant-oil-water (SOW) systems in order to predict the stability of microemulsion based products. The knowledge of the oil's EACN value allows one to predict whether a particular oil should at equilibrium form Winsor microemulsions of type I, II or III, or a direct or inverse emulsion after stirring [Lukowicz et al. 2018]. There is a very recent book chapter by Acosta et al. (2025) that deals with the concept of HLD and the practical application of HLD-NAC theory. The application of HLD-NAC theory as a predictive tool using cosmetic or pharmaceutical excipients can allow the rational design of emulsion-based products, eliminating in practice the need for many problem-solving procedures and long-term product development [Bilchak 2024].

In practise, the EACN value is used as a dimensionless parameter, being a quantitative measure of the relative hydrophobicity (lipophilicity) of the oil phase of the emulsions or microemulsions. This parameter is usually expressed as the equivalent alkane carbon numbers in the molecule of the components forming the oil phase of the emulsion or microemulsion based product. It was assumed that for saturated and non-branched aliphatic hydrocarbons the EACN value corresponds to the total number of carbon atoms present in the hydrocarbon chain of the alkane [Wade et al. 1977]. It was also assumed that the EACN value for a benzene molecule is equal to zero. For small size oils even negative EACN values are possible, especially for oils with some electron-rich heteroatoms, such as oxygen or nitrogen. For other molecules, such as fragrance compounds or perfumery raw materials, the EACN values have to be determined experimentally.

The EACN is the quantity applied in the HLD equation in the late 1970s [Salager 1979; Bourrel and Schechter 1988] to describe the hydrophilic-lipophilic nature of the oil molecules used in the quantitative description of the phase equilibria as well as the physical properties of emulsions and microemulsions. The HLD equation essentially describes the balance between the salinity and temperature of

the system, the hydrophobicity of the oil phase (EACN), the characteristics of the surfactants (Cc) and cosurfactants (i.e. presence of aliphatic alcohols). It has been found experimentally that positive values of the EACN correspond to hydrophobic compounds while negative ones are asigned to hydrophilic compounds. This type of information is necessary in the HLD-NAC theory to predict physicochemical properties affecting a quality of emulsions-based products, such as the huge number of fragrance or cosmetic products [Acosta 2020] available in the market.

According to Salager et al. (2000) the stability (or instability) as well as type of the formed microemulsion based systems (such as fragrance or cosmetic emulsions) can be simply described by the value of HLD. In general, for the systems described by HLD < 0, microemulsions of Winsor type I are formed, while for the systems with HLD > 0 microemulsions of Winsor type II are formed. In the case of oil and water mixtures formed at low surfactant concentrations described by the HLD value close to 0, the unstable triphase systems are observed (Winsor type III).

Table 1 shows some experimental EACN values of the oil phases of various emulsions and microemulsions published in the literature. The data given in this table are divided into two groups: hydrophobic compounds of petrochemical origin and silicone drivatives (left hand side of Table 1) and those of natural origin including fragrance compounds, natural oils, fatty acid esters and glicerol trialkanoates (right hand side of Tab. 1).

Table 1. Experimental EACN values of oil components of various emulsions

Petrochemical oils	EACN	Natural oils and fatty esters	EACN
n-Alkanes, C_nH_{2n+2}	n	Ambrettolid	1.0
Isododecane	10.0	Cyneole	-1.6
Isohexadecane	12.2	Damascone	-1.2
Paraffin	18-25	Delta-3-carene	2.9
1-Octene	3.9	Eucalyptol	-1.6
1-Decene	5.5	ω-6-Hexadecenolactone	1.0
1-Dodecene	8.1	β-Ionone	-1.9
1-Octadecene	14.2	D-Carvone	-3.1
1-Octyne	-1.8	Longifolene	7.0
1-Decyne	0.1	Linalool	-11.4
1-Dodecyne	2.0	Limonene	2.0
1-Tetradecyne	3.5	Mentane	6.2
Asphaltene	1	Menthene	3.3
Purified kerosene	8.6	Menthone	-1.5
Benzene	0	Mircane	10
Toluene	1	Nerol	-21.9
Ethylbenzene	-1.8*	Cytronelyl acetate	-0.2

cont. Table 1

Propylbenzene	-0.8*	Geranyl acetate	-0.6
Butylbenzene	0.4	Linalyl acetate	-0.9
Hexylbenzene	2.2	Menthyl acetate	-0.1
Octylbenzene	3.8	Pinane	4.3
Decylbenzene	6.0	α-Pinene	2.9
Dodecylbenzene	7.9	β-Pinene	2.2
p-Cymene	-0.3	α-Terpinene	0.9
p-Xylene	-2.4	γ-Terpinene	1.7
Styrene	3	Terpinolene	0.7
Naphthalene	1	Rose oxide	-1.7
o-Dichlorobenzene, o-$C_6H_4Cl_2$	-5	Hexadecyl acetate	5.0
Ethyl benzoate	-1.0	Tetradecyl propionate	6.8
Cyclohexane	2.2	Decyl butyrate	5.0
Cyclooctane	4.1	Hexyl caprylate	6.2
Cyclodecane	5.5	Octyl caprylate	8.1
Methylcyclohexane	3.2	Ethyl capronate	2.2
Ethylcyclohexane	4.2	Ethyl laurate	3.8
Propylcyclohexane	5.7	Butyl laurate	7.2
Butylcyclohexane	7.0	Hexyl laurate	9.4
Decylcyclohexane	14.4	Ethyl myristate	5.3
Dodecylcyclohexane	17.3	Isopropyl myristate, (IPM)	7.5
Cyclohexene	-1.2	Ethyl palmitate	6.8
cis-Decaline	6.3	Ethyl oleate	7.0
2-Octanone	-3.4	Di(2-ethylhexyl) adipate	9.7
2-Decanone	-2.1	Glycerol tricaprylate	12.2
2-Undecanone	-1.3	Glycerol tricaprynate	13.8
2-Dodecanone	-0.6	Glycerol trilaurate	15.7
Diisopropyl ether, $(iC_3H_7)_2O$	2.3	Glycerol trimyristate	18.5
Dibutyl ether, $(C_4H_9)_2O$	3.4	Glycerol trimyristoleate	18.0
Dipentyl ether, $(C_5H_{11})_2O$	4.2	Glycerol tripalmitate	21.2
Dihexyl ether, $(C_6H_{13})_2O$	6.2	Glycerol tripalmoleinate	19.0
Hexyl metacrylate	1.1	Glycerol tristearate	23.9
Octanenitrile, $C_7H_{15}CN$	-1.7	Glycerol trioleate	21.2
Decanenitrile, $C_9H_{19}CN$	-0.5	Glycerol trilinolenate	20.0
Dodecanonitrile, $C_{11}H_{23}CN$	0.4	Pine oil	-12.1
Chlorooctane, $C_8H_{17}Cl$	1.0	Jojoba oil	11
Chlorodecane, $C_{10}H_{21}Cl$	3.5	Coconut oil	12
Chlorododecane, $C_{12}H_{25}Cl$	5.6	Flax oil	13

cont. Table 1

Chlorotetradecane, $C_{14}H_{29}Cl$	8.0	Sunflower oil	14
Chlorohexadecane, $C_{16}H_{33}Cl$	9.8	Palm oil	15
1,1,1-Trichloroethane, CH_3CCl_3	-2.5	Corn oil	16
Trichloroethylene, CCl_2=CHCl	-3.8	Canola oil	17
Tetrachloromethane, CCl_4	0	Olive oil	18
Tetrachloroethylene, CCl_2=CCl_2	2.9	Peanut oil	19
Cetiol-S	17	Sqalane	24.4
Hexamethylodisiloxane	12.2	Sqalene	16
Silicone oil	14.6	Vitamin E	5.2

* – values estimated based on group contributions listed in Table 2 (own research).

Source: [Castellino et al. 1976; Salager 1999; Salager et al. 2001; Poprawski et al. 2003; Wittahayapanyanon et al. 2006; Queste et al. 2007; Bouton et al. 2009; 2010; Phan, Harwell and Sabatini 2010; Hammond and Acosta 2012; Do et al. 2015; Ghosh 2015; Lukowicz 2015; Wan et al. 2016; Marquez et al. 2019; Chen et al. 2022].

2. MATERIALS AND METHODS

The aim of this work was to determine a relationship between the chemical structure of various hydrophobic molecules and the experimentally available values of the EACN of corresponding oils reported in the literature in order to derive a novel set of numerical EACN values attributed to various structural elements of oily molecules. We have also assumed that it is possible to apply the EACN group contribution values obtained in such a way to predict the values of the EACN of various hydrophobic compounds which are used in the HLD equation for composition design of emulsions and microemulsions as well as for the prediction of their phase behavior and number of physicochemical properties described in HLD-NAC theory developed by Acosta (2004). Such information is necessary to design novel high quality fragrance or cosmetic products in a reasonably short time.

This work collects experimental EACN values published in the literature for various hydrophobic compounds (i.e. oils) with a linear or branched hydrocarbon moieties. We have assumed that the experimental value of the EACN in the HLD equation for a given oil can be represented as a simple sum of group contributions assigned to the structural elements forming a single hydrophobic molecule (i.e. oil).

In order to derive the numerical values of the EACN group increments describing the structural elements of the molecules, we have assumed a simple form of the multilinear regression equation, including contributions attributed to the structural elements present in the molecules of hydrophobic compounds and a few correction factors assigned to intramolecular interactions between some components of the molecule.

Finally, in this study we used 40 basic group types and 9 correction factors to classify 141 various structures of hydrophobic compounds (oils) used in surfactant,

cosmetic or fragrance products. A generic equation of the group contribution model used in this work can be given in the following form:

$$EACN = \sum_i N_i \cdot EACN_i + \sum_j M_j \cdot EACN_j$$

where:

N_i – number of structural "i" elements present in the hydrophobic molecule,
$EACN_i$ – contribution of the "i" group to EACN value of the hydrophobic molecule,
M_j – number of correction factors for specific configuration of structural groups of the "j" type in the hydrophobic molecule,
$EACN_j$ – correction factor of the "j" type.

3. RESULTS AND DISCUSSION

The data presented in Table 1 show that the presence of branching in the hydrophobic chain structure of the hydrophobic molecules tends to reduce their lipophilicity (e.g. decrease in the EACN value) which is consistent with the suggestion of Graciaa et al. (1982a, b).

In the literature one can find only a relatively small number of experimentally determined values of the EACN for hydrophobic compounds (Tab. 1). Therefore, in order to compute the numerical values of group contributions and correction factors based on the 141 experimental EACN data sets available in the literature, we have applied a simple multiple linear regression (MLR) equation using the least-squared method with the Levenberg and Marquadt algorithm implemented in the Solver tool in the Excel 2019 spreadsheet.

The results of our computational work are presented in Table 2 and shows the numerical values of 40 group increments and 9 structural corrections computed for several structural elements, which can be used to calculate the EACN values for various hydrophobic compounds.

It should be noted that for aliphatic halogen containing compounds, this computation method is limited only to the structures having a single chlorine or bromine atom. The application of the contributions of molecular structure elements listed in Table 2 to calculate the EACN value for hydrophobic components of the oil phase of microemulsions is illustrated by the calculation of the EACN value for naphthalene: EACN = 8 · (Car-H) + 2 (-Car=) + 2 · (correction for a 6-membered ring) = 8 · (0.1235) + 2 (0.7522) + 2 · (-0.7466) = 1.00. Experimental EACN = 1.0 [Abbott 2026].

Figure 1 shows the relationship between experimental and computed values of the characteristic curvature for 141 hydrophobic compounds.

Figure 1 shows the experimental EACN values, and those computed in this work produce almost perfect linear plot passing the origin with a unit slope, hence

allowing for effective prediction of the EACN values for various hydrophobic ingredients (except edible oils) frequently used in the preparation of emulsions or microemulsions, such as fragrance compounds or perfumery raw materials.

Table 2. Group contributions for calculating the EACN values of oil phase components of emulsions and microemulsions

Structure elements	EACN	Structure elements	EACN
Aromatic components		-CH= (in ring)	-1.2203
$CarCH_3$	1.1258	>C= (in ring)	1.0430
$CarCH_2$-	-2.7382	$-CH_2$- (R) (in vicinity of -O-)	-5.7560
Car-CH<	-0.5606	>CH- (R) (in vicinity of -O-)	-1.6334
Car-CH=	4.9802	>C< (R) (in vicinity of -O-)	-5.0098
Car-C#	508.156	>C=O (in ring)	-7.1315
Car-H	0.1235	-COO- (in ring)	-4.5969
-Car=	0.7522	-O- (in ring)	4.0885
-Car-COO-	-2.8596	**Functional groups**	
-Car-Cl	-2.3737	-OH	-17.8220
Aliphatic components		-CN	-9.5245
$-CH_3$	1.0016	-COO-	-5.4388
$-CH_2$-	0.9904	>C=O	-11.0334
>CH-	0.0591	-CH=CH-CO-	-5.5128
CH_2=	-1.8512	-O-	4.2914
-CH=	-0.2837	-Cl	-6.5348
>C=	-0.0273	-Br	-6.3727
>C<	0.1804	**Structural correction factors**	
H-C#	505.394	3-membered ring	-2.3923
#C-	-513.319	4-membered ring	-0.2554
$-CH_2$- (in vicinity of -O-)	-2.3264	5-membered ring	0.2985
-CH< (in vicinity of -O-)	-2.2098	6-membered ring	-0.7466
>C< (in vicinity of -O-)	0.9151	6+-membered ring	-0.5152
Alicyclic components		Symmetric ether	-3.2485
$-CH_2$- (in ring)	0.6231	Second chlorine atom	-1.9892
>CH- (in ring)	0.8921	Conjugated system -C=C-C=C-	-2.2433
>C< (in ring)	-1.0836	Beta branching	-0.6725

Symbols: Car – a carbon atom being a component of the aromatic system.
(R) – a component of an alicyclic ring. # – a triple bond

Source: own research.

Our analysis of the chemical structure of various fragrances and perfumery raw materials clearly indicates that their polarity (described by means of the EACN value) varies in the following way:

- EACN increases with the increase in the number of the alkyl chain length;

- EACN decreases with the increase in the number of polar groups;
- EACN decreases with the increase in the degree of unsaturation for substances having a similar structure;
- substances having a similar structure to the ketone group have higher values of EACN than those with ester groups;
- the presence of a conjugated system of double bonds in the structure of fragrance substances leads to a decrease in the EACN value (i.e. an increase in polarity);
- the numerical value of EACN depends also on the number and the size of aromatic and alicyclic rings present in the structure of the substances.

It should be noted that our group contributions to the EACN value of hydrophobic molecules listed in Table 2 have also some predictive power. For example, the numerical value of the EACN parameter calculated on the basis of the data given in Table 2 for the simplest three-carbon unbranched group [$CH_2CH_2CH_2$] is 2.97, while for the analogous branched group [$CH(CH_3)$-CH_2] it is 2.05. According to the physical interpretation of the EACN value, the substances having lower values of EACN are less hydrophobic (ie. more hydrophilic or more polar). This is in excelent agreement with the results of experiments in the available literature indicating lower lypophilicity for molecules having branched hydrophobic moieties.

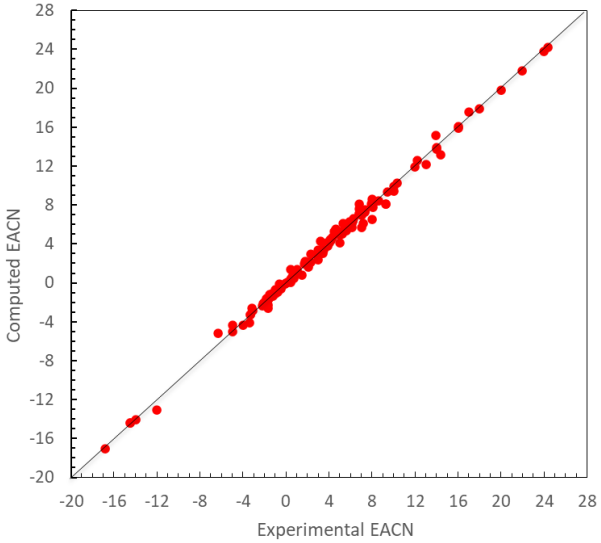

Fig. 1. Comparison of computed and experimental EACN values for 141 hydrophobic compounds

Source: own research.

Figure 2 shows the relationship between the experimental and computed values of the characteristic curvature for 58 fragrances and perfumery raw materials. As can be seen in Figure 2 the experimental EACN values for components of fragrances and

those computed in this work produce a linear plot passing the origin, with a unit slope allowing for effective prediction of the EACN for fragrances and perfumery raw materials.

It should be noted, however, that although the presented simple computational method works quite well for the EACN values for a huge number of oily compounds used in both cosmetic and perfumery products [Bouton et al. 2010; Hammond and Acosta 2012], it cannot be used for triglycerides of higher fatty acids. Hence, in the case of designing microemulsion based products in which the oil phase components are the triglycerides and in the absence of appropriate experimental data for them, it is recommended to use the empirical equation developed by Witthayapanyanon et al. (2006), given in the following form:

$$EACN = 2.05 \cdot ASC + 0.071 \cdot UC - 24.0$$

where:

ASC – average number of carbon atoms of fatty acid moieties present in the structure of triglyceride molecule,

UC – percentage of unsaturated chains present in the structure of triglyceride molecule.

Furthermore, due to the current lack of sufficient reliable experimental data, this method cannot be applied to compounds having more than one chlorine atom located on the same carbon atom or a chlorine atom located on a carbon atom containing a double bond.

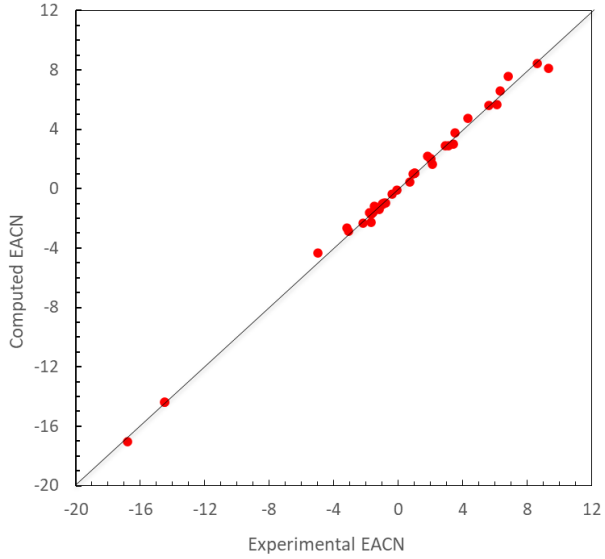

Fig. 2. Comparison of computed and experimental values of EACN for 58 fragrances and perfumery raw materials

Source: own research.

4. CONCLUSIONS

The results of the investigation into the EACN values of the hydrophobic compounds used in the HLD equation can be applied in the prediction of the phase equilibrium of emulsion and microemulsion systems. Here we propose a novel set of group contributions consisting of 40 group increments and 9 correction factors for fast and efficient calculation of the EACN values for various hydrophobic compounds. The group contributions derived for various oils of petrochemical origin and those used in preparation of fragrance compositions have been tested and the computed results compared with the corresponding experimental values of the EACN available in the literature.

It has been demonstrated that the newly proposed set of numerical values of both group contributions and correction factors correctly predicts ($r^2 = 0.9951$) the values of the EACN value appearing in the HLD equation, with a maximum error of 1.48 units and a mean square error of 0.33 for various types of 141 hydrophobic compounds.

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Zieliński, R., 2021, *Surfactants. Structure. Properties. Applications*, Wydawnictwo Uniwersytetu Ekonomicznego w Poznaniu, Poznań.

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