Is it Correct to Name DESs Deep *Eutectic* Solvents?

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Recent years of research and development have brought frequently used terms for new types of green solvents to the lexicon of scientists. This can lead to terminological inaccuracies. In particular, different names are being used for the same types of solvents: Deep Eutectic Solvents (DES); Natural Deep Eutectic Solvents; Low-Transition Temperature Mixtures; Low-Melting Mixtures. It would, therefore, be appropriate to eliminate certain inaccuracies and to use simplification, which means using the general term "Low-Temperature Transition Mixtures" or introducing the term "DES-like mixtures".

Keywords: Deep Eutectic Solvents; Natural Deep Eutectic Solvents; Low-Transition Temperature Mixtures; Low-Melting Mixtures; Terminological Inaccuracies

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Deep Eutectic Solvents and Terminological Inaccuracies

Biomass contains numerous valuable chemical substances, and it is logical that just burning biomass without previous extraction and isolation of at least some portion of such compounds is contrary to common sense. This is why many academic and industrial institutions are trying to find advantageous modes of biomass valorization, in other words, obtaining valuable compounds and substances through processing materials of biological origin.

Most such valorization procedures for isolation of chemical compounds from biomass involve extraction techniques. As for extractants, taking also environmental issues into account, a shift from typical organic solvents through ionic liquids to green solvents is clearly identifiable. Most organic solvents are flammable liquids with relatively high vapour pressure, low viscosity, infrequently considerable toxicity for living organisms, and a negative impact on the environment. Abbott is generally recognized as the father of a real breakthrough in extraction methods (Abbot *et al.* 2003). He described a new type of extraction reagents with attractive physicochemical properties, such as fire resistance, negligible vapor pressure, low electric conductivity, purposefully tunable viscosity, miscibility with water, easy recovery, and maintaining a liquid state in a wide temperature range. For such extractants, Abbott coined the term Deep Eutectic Solvents (DESs). Currently, the term Deep Eutectic Solvents (DESs) refers to mixtures of two or more components – hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) – which can bond with each other to form a eutectic mixture having a final solidification point that is lower than the melting point of the raw materials (HBD) and (HBA), becoming thus liquids at room temperature. When the compounds that constitute the DESs are exclusively primary metabolites, namely, amino acids, organic acids, sugars, or choline derivatives, the DESs are called Natural Deep Eutectic Solvents (NADESs). As the first example, a DES formed from urea (U) and choline chloride (ChCl) was described. Pure U and ChCl melt at 134 °C and 302 °C, respectively. The solidification temperature for their mixture with a 1:2 molar ratio is 12 °C, so this mixture is liquid at room temperature.

Over time, new terms have emerged to refer to low solidification point mixtures that are used, *inter alia*, in extraction processes. In practice, Low-Transition Temperature Mixtures (LTTMs) and Low-Melting Mixtures (LMMs) are most frequently used. As pointed out by several authors (de Oliveira Vigier and García-Álvarez 2017; Hansen *et al.* 2021; Yusuf *et al.* 2021), there is no significant difference between them. Thus, DESs, LTTMs and LMMS are neoteric solvents consisting of at least two components, which are capable of forming a liquid phase by a hydrogen-bond-promoted self-association. The solidification temperature is tacitly assumed to be below 100 °C, the lower, the better.

A problem lies in the fact that the term *eutectic* is included in the definition of all mentioned solvents (de Oliveira Vigier and García-Álvarez 2017; Hansen *et al.* 2021; Yusuf *et al.* 2021). Due to the requirements for the accuracy of scientific and technological terms, the use of the term eutectic in relation to these solvents is often incorrect. Exactly, the term *eutectic* was introduced by the British physicist and chemist Frederick Guthrie in 1884 (Guthrie 1884). A eutectic system is a mixture formed of two or more solids at such proportions that the melting/freezing point is as low as possible. The eutectic temperature is thus the lowest solidification temperature over all of the mixing ratios for the involved component species (Daněk 2006). It is precisely this fact that contradicts the usage of such terms in the literature, where in addition to a single invariant composition of a mixture, which has the lowest possible solidification temperature and is, therefore, a eutectic, mixtures with different HBA: HBD ratios are also referred to as eutectic mixtures. It can be mentioned that even with commonly used (for example based on choline chloride) DESs, generally no phase diagram has been constructed. In other words, generally there has been no effort to determine the ratio corresponding to a true eutectic mixture and the actual eutectic temperature.

As an example, mixtures of tetrabutylammonium bromide (HBA) and levulinic acid (HBD) with a molar ratio of 1:1, 1:2, 1:3 and 1:4, are all referred to as deep eutectic solvents (Bai *et al.* 2021). Five systems with different components (namely choline chloride: ethylene glycol and choline chloride: glycerol) ratios are referred to as DES (Tang and Row 2013). Further examples can be found (Jablonský and Šima 2019; Liu *et al.* 2021). Some open questions in this area have been indicated by Martins and his coworkers (Martins *et al.* 2019). In addition, several water-containing systems are also referred to as DESs. The cause of this state is understandable. If a system containing HBD and HBA has a sufficiently low solidification temperature (in other words, it is liquid at ambient temperature) and properties suitable for the required practical use, it is referred to as DES as if it were a system with a composition corresponding to the eutectic point. In addition, some DESs are considered designer solvents because their properties can be tailored by selecting different combinations of their components (Alhadid *et al.* 2019).

To eliminate the mentioned terminological inaccuracies, but to preserve traditional, generally accepted names, we recommend two options: the first is to prefer the general term "low-temperature transition mixtures," and the second is to use the term "DES-like mixtures" for systems where the existence of a eutectic point has not been proven.

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