Journal of Molecular Liquids 330 (2021) 115717

Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq

Choline chloride-based ternary deep band gap systems

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ARTICLE INFO

Article history:
Received 8 December 2020
Received in revised form 14 January 2021
Accepted 17 February 2021
Available online 20 February 2021

Keywords:
Deep eutectic systems
Band gap
Tauc plot
Urbach energy

ABSTRACT

The structural features derived by the UV-VIS spectroscopical analysis of five ternary eutectic systems containing choline chloride as Hydrogen Bond Acceptor (HBA), and ethylene glycol and a second Hydrogen Bond Donor (HBD, water, glycerol, methanol, ethanol, and 2-propanol), have been determined and described in terms of band gap and Urbach energies (BGE and UE). In particular, the relationship between the molar composition of the ternary eutectic systems and the BG and U energies was assessed through the Tauc plot methodology, which revealed a peculiar behaviour of the system containing 2-propanol. Thus, a Design of Experiments (DoE) approach, specifically a two-levels full factorial screening experiment, followed by a Surface Responding Analysis (SRA), allowed to determine the influence of the composition and other operational parameters (temperature and resting time) on the structural disorder of the final ternary eutectic system. An unprecedented effect of the DES-aging was observed on the UE, indicating that the structure of these systems evolves during time by increasing its order.

1. Introduction

Selected combinations of Hydrogen Bond Donors (HBDs) and Hydrogen Bond Acceptors (HBAs) have shown the capability to form eutectic mixtures characterized by a low melting point, which results to be lower than the ones of the single constituents [1,2]. When this drop exceeds certain values, the mixture is labelled as Deep Eutectic System (DES) [3–5]. Since the first report of Abbot and co-workers on the description of a DES between choline chloride and Urea [6], many eutectic systems have been developed. DESs have found application, only to cite some examples, as solvents for biomass or metals [7,8], media for the trapping of Volatile Organic Compounds [9] catalysts or non-innocent solvents in organic synthesis [10–13], and also as templates for zeolite synthesis [14].

The peculiar and in many cases improved properties of DESs (with respect to classic organic solvents) are strictly related to their unique structure, characterized by an intense network of hydrogen bonds between the formal constituents which act as Hydrogen Bond Acceptors (HBAs) and Hydrogen Bond Donors (HBDs). From a chemical physical point of view, the behaviour of masses and charges in a DES can be described by the *hole theory* [15], which considers a molten salt as a system where empty spaces are continuously generated by local fluctuations in density [16]. In a DES, the average size of such spaces is big enough for anion to move within the holes. This model well explains for many eutectic systems their high viscosity, as well as increased densities and other physical properties [17,18].

Although it is largely accepted that physical properties of DESs are in some way related to the structure of their network along with the nature of HBAs and HBDs, for the best of our knowledge, specific information about the relationship between composition and behaviour of the systems are not yet available. Given a eutectic system, the understanding of how each component and each operational factor (temperature, concentration, time) affects a given physical parameter of the DES, represents a key step to engineering eutectics systems envisioning exclusive applications.

Recently, some of us employed a Design of Experiments (DoE) approach, combined with multivariate statistical analysis, to elucidate the effects of HBA, HBDs, and their combination on the band gap energy of some binary eutectic systems containing choline chloride [19] or triphenylmethylphosphonium bromide [20]. In particular, some of these systems, named as Deep Band Gap Systems (DEBAGs) showed a relevant drop of the direct band gap energy in correspondence to the eutectic composition, suggesting a relationship between band gap energy and the eutectic molar ratio. Moreover, when hydrophilic HBAs as choline chloride or choline acetate are used, the water role became relevant. Formally, when a DES is added with water or it picks up water from air [21] an additional HBD is introduced. Although a physiological amount of water in a DES is usually tolerated and its structural effect neglected [22] some studies revealed that a detailed engineering of the properties of a eutectic mixture can be achieved by tuning the water presence in the system [23]. The presence of water has relevant effects not only on common physical parameters as density and

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https://doi.org/10.1016/j.molliq.2021.115717
0167-7322/

viscosity [24–27], but also on the speed of sound [28], NMR relaxation [29] and the band gap energy [19].

In particular, preliminary studies revealed that adding 10 wt% of water to systems containing choline chloride, a systematic decreasing of the band gap energy is produced [19].

In this context, with the aim to explore some structural properties of ternary eutectic systems, it is herein reported a study on the effect of a second HBD on the band gap energy and on the structure disorder in DESs containing choline chloride as HBA and ethylene glycol as HBD. Five families of eutectics containing choline chloride, ethylene glycol, and a third HBD (water, glycerol, methanol, ethanol, 2-propanol) were prepared, each one with a different molar ratio between HBA and HBDs and analysed by UV-VIS spectroscopy. Then, the corresponding band gap (BG) and Urbach (UE) energies were determined through the graphic Tauc plot method. For the model system composed by choline chloride / ethylene glycol / *2*-propanol, a Design of Experiments (DoE) approach followed by a statistical multivariate analysis was employed to evaluating the effects of the two HBDs, temperature, stirring time, and their combinations.

2. Experimental section

2.1. General synthetic procedure for DESs preparation

Chemicals were purchased from commercial sources and used as received. In particular, choline chloride (>98%) and 2-propanol (99.8%) were purchased from Sigma Aldrich, methanol, ethanol (96%) and glycerol (99.6%) from VWR, and ethylene glycol (99%) from Carlo Erba. Finally, H_2O was purified with a Millipore RiO_s 3 Water System.

The ternary mixtures (I-V) were prepared as follows: a common stock solution of DES choline chloride/ethylene glycol (1/2 M ratio) was prepared. The desired molar fractions of DES and HBDs reported in Table 1 were inserted in 5 mL capped vials for all the analysed ternary mixtures and stirred at 30 °C for 4 h. All the samples were analysed at r.t. within 15 min after the samples' preparation. The additional samples, necessary for the optimization of system V (Table 4), were prepared in accordance to the above reported equivalent ratios in 5 mL capped vials and stirred respectively at 25 °C and 80 °C for 1 h and measured at r.t respectively after 2 h or 24 h of resting time at r.t..

2.2. Spectroscopic UV-VIS analysis

The samples were analysed in a pure form and the UV–Vis spectra of all samples were recorded in a quartz cell (path length: 1.00 mm) with an Agilent Cary 60 UV–Vis Spectrophotometer.

2.3. Statistical analysis

Full factorial design and Surface Responding Analysis, as well as the statistical multivariate analysis, were performed using Statgraphics Centurion 18 software. The experiments were conducted in a single block and in randomized order.

Table 1
Systems prepared and corresponding nomenclature.

$\chi_{CC/EG}$-χ_{H2O}	$\chi_{CC/EG}$-$\chi_{glycerol}$	$\chi_{CC/EG}$-$\chi_{methanol}$	$\chi_{CC/EG}$-$\chi_{ethanol}$	$\chi_{CC/EG}$-$\chi_{2\text{-propanol}}$
1/0 (Ia)	1/0 (IIa)	1/0 (**IIIa**)	1/0 (**IVa**)	1/0 (**Va**)
0.66/0.34 (**Ib**)	0.66/0.34 (**IIb**)	0.66/0.34 (**IIIb**)	0.66/0.34 (**IVb**)	0.66/0.34 (**Vb**)
0.5/0.5 (**Ic**)	0.5/0.5 (**IIc**)	0.5/0.5 (**IIIc**)	0.5/0.5 (**IVc**)	0.5/0.5 (**Vc**)
0.34/0.66 (**Id**)	0.34/0.66 (**IId**)	0.34/0.66 (**IIId**)	0.34/0.66 (**IVd**)	0.34/0.66 (**Vd**)
0/1 (**Ie**)	0/1 (**IIe**)	0/1 (**IIIe**)	0/1 (**IVe**)	0/1 (**Ve**)

2.4. Design of Experiment (DoE)

A two levels full factorial design n^k ($n = +1, -1$) ($k = 4$) model was used to assess the response variables band gap energy (BGE) and Urbach energy (UE). The Statgraphics Centurion v 18 software was used for the Design of Experiment (DoE) and for the Surface Responding Analysis (SRA).

3. Results and discussion

Five ternary mixtures containing choline chloride, ethylene glycol and water (I), glycerol (II), methanol (III), ethanol (IV), and *2*-propanol (V) were prepared by mixing the three components and stirring them at 30 °C during 4 h. For each system, 5 different molar ratios were considered and a total of 25 mixtures obtained as reported in Table 1.

Choline chloride and ethylene glycol are known forming a DES in 1/3 M ratio. For the purpose of the present study, in order to monitor the behaviour of the system upon addition of a second HBD, CC/EG has been considered as a single species, with a molecular weight resulting from the sum of the formers' molecular weights. At this point, five mixtures have been prepared for each additional HBD, characterized by the molar ratio reported in Table 1.

Each system reported in Table 1 was subjected to UV-VIS investigations and analysing the absorbance data, the energy corresponding to the direct transition from the valence to the conduction bands was determined by employing the Tauc plot method. A detailed description of the procedure is reported in references 19 and 20, and the complete list of the Tauc plots can be found in the Supporting Information. In Fig. 1 a representative Tauc curve is reported for each system I-V.

In figure it is possible to observe how, the change in the band gap energy produced by the presence of a second donor is minimal mixtures Id-IVd in respect to the known DES choline chloride / ethylene glycol, and all the systems show a BGE which ranges between 5.9 and 6.1 eV. The only exception it is represented by sample Vd corresponding to choline chloride/ethylene glycol/2-propanol, which shows a BGE value of 5.35 eV, resulting about 0.5 eV lower than the reference (CC/EG).

Nevertheless, looking only at the BGE values, few differences can be highlighted between the systems considered. Much more information can be acquired by the analysis of the structural disorder through the variation of the Urbach Energy (UE). Considering the exponential part of the α *vs* energy curve, which it is known as Urbach tail [30] it is possible to determine the Urbach Energies (UEs) for systems I-V. The UE is indicative to the degree of structural disorder of the material and it is a useful parameter in doping or annealing studies: it easily allows to

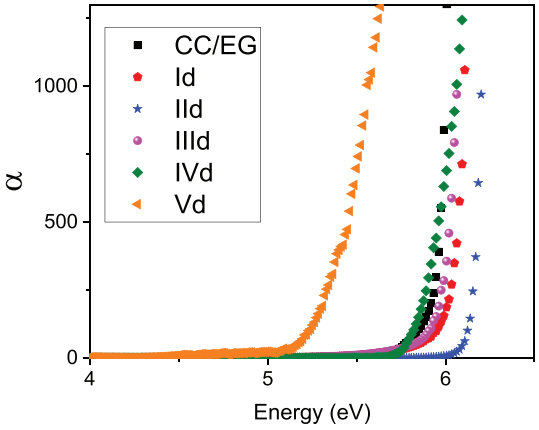

Fig. 1. Tauc plot for the direct band gap of systems I-V (choline chloride-ethylene glycol/HBD 0.34/0.66). Choline chloride / ethylene glycol (CC/EG 1/3) curve is reported for comparison.

understand if the presence of an additive increase (by lowering the UE) or decrease (by increasing the UE) the crystallinity of the system [31].

In Table 2, the complete set of BG and Urbach energies for the systems I-V at different molar ratio are reported.

Critically observing the data reported in Table 2, it is possible to notice that the effect of the introduction of a second HBD into the eutectic system composed by choline chloride and ethylene glycol (1/3) has a relevant impact on the structural disorder. By plotting the UEs as function of the molar composition of the system it is possible to evaluate the increase or decrease related to the disorder degree upon the addition of the second HBD with respect to the starting materials (Fig. 2).

It is evident from the plots reported in Fig. 2 that only for the system V (choline chloride / ethylene glycol / 2-propanol) it is possible to observe a UE behaviour where the mixture between HBA and the HBDs has an increased crystalline degree with respect to the starting materials (Fig. 2, green dots). This observation confirms the hypothesis formulated from the analysis of the BG energies, relative to a peculiar effect of the 2-propanol in the ternary system V. It is worth to mention that even in the case of system III (methanol, blue dots) a minimum of UE exists, corresponding to the ternary system (choline chloride/ethylene glycol – methanol 0.66–0.34), but the entity of such effect is slightly relevant if compared with system V.

To investigate the deep decreasing of UE observed in the case of system V, a Design of Experiments (DoE) approach was employed followed by a multivariate statistical analysis. The data reported in Table 2 and Fig. 2, clearly highlight the possibility to generating a ternary deep band gap and *deep Urbach energy system* by combining choline chloride, ethylene glycol, and 2-propanol in specific relative amounts. Nevertheless, the effect of the single experimental conditions (temperature, time, relative amounts of HBDs) and of their combination on the disorder degree it is unknown. For a deeper analysis on these important aspects, a two-levels full factorial 2^k (K represents the experimental factors considered) screening experiment was indeed conducted.

In particular, 4 experimental factors have been considered as indicated in Table 3.

Table 2
BG and Urbach energies for the considered systems.

Entry	$\chi_{CC/EG}$-χ_{H2O}	BG (eV)	Urbach (meV)
1	1/0 (**Ia**)	6.02	363.1
2	0.66/0.34 (**Ib**)	6.02	466.2
3	0.5/0.5 (**Ic**)	6.0	445.2
4	0.34/0.66 (**Id**)	6.02	390.2
5	0/1 (**Ie**)	6.42	773.4
	$\chi_{CC/EG}$-$\chi_{glycerol}$		
6	0.66/0.34 (**IIb**)	6.02	475.3
7	0.5/0.5 (**IIc**)	5.98	443.7
8	0.34/0.66 (**IId**)	5.98	513.4
9	0/1 (**IIe**)	5.23	321.7
	$\chi_{CC/EG}$-$\chi_{methanol}$		
10	0.34 (**IIIb**)	5.99	341.8
11	0.5 (**IIIc**)	6.00	364.4
12	0.66 (**IIId**)	5.9	389.9
13	1 (**IIIe**)	5.98	370.5
	$\chi_{CC/EG}$-$\chi_{ethanol}$		
14	0.34 (**IVb**)	6.0	492.8
15	0.5 (**IVc**)	6.0	390.3
16	0.66 (**IVd**)	6.0	547.6
17	1 (**IVe**)	6.0	314.7
	$\chi_{CC/EG}$-$\chi_{2\text{-propanol}}$		
18	0.34 (**Vb**)	6.02	193.2
19	0.5 (**Vc**)	5.96	146.4
20	0.66 (**Vd**)	5.35	210.5
21	1 (**Ve**)	5.85	540.8

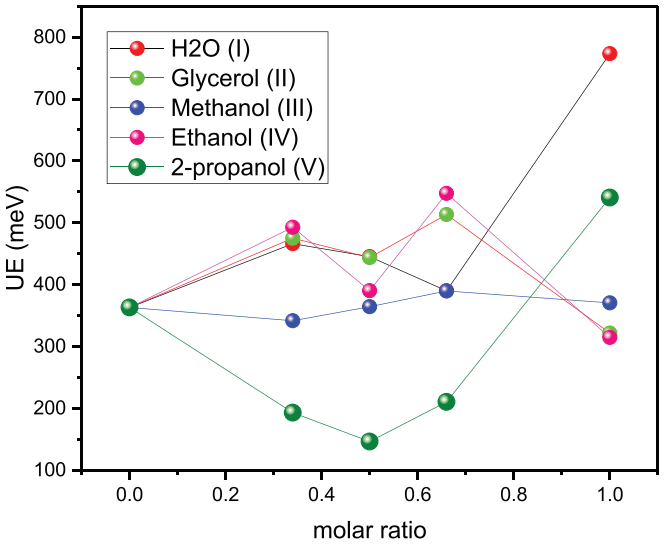

Fig. 2. Urbach energies as function of the molar composition for systems I-V.

Table 3
Factors considered and corresponding limits.

Factors	Low	High	Units	Continuous
glycerol	0.5	7	eq.	Yes
2-propanol	0.5	7	eq.	Yes
Temperature	25	80	°C	Yes
Resting time	2	24	h	Yes

Table 4
list of experiments conducted.

Entry	Ethylene glycol (eq.)	*i*PrOH (eq.)	Temperature (°C)	Resting time (h)	Band Gap Energy	Urbach Energy
1	1.75	0.5	80	2	5.95	136.52
2	7	7	80	2	6	173.75
3	7	0.5	25	2	6.07	120.47
4	1.75	7	25	24	5.92	238.66
5	1.75	0.5	25	2	5.99	124.15
6	1.75	7	25	2	6.02	134.06
7	1.75	7	80	24	5.96	168.09
8	7	0.5	80	2	6.08	135.35
9	7	7	80	24	6.04	268.16
10	7	7	25	24	6.04	367.22
11	7	0.5	25	24	6.07	133.96
12	1.75	0.5	25	24	5.96	328.69
13	1.75	7	80	2	6.01	139.82
14	1.75	0.5	80	24	5.93	279.53
15	7	7	25	2	6.01	164.39
16	7	0.5	80	24	6.08	248.61

The considered factors and relative ranges are reported in Table 3. Glycerol and 2-propanol amounts have been expressed in equivalents in moles with respect to choline chloride (Table 3 and Table 4).

As indicated by the statistical model, 16 experiments have been conducted (Table 4) and the BG and Urbach energies have been calculated (responses) for each one.

On the basis of the data acquired, a statistical multivariate analysis for all the responses have been performed.

3.1. Band gap statistical analysis

The effect of the factors and their combination can be represented using the Pareto chart (Fig. 3).

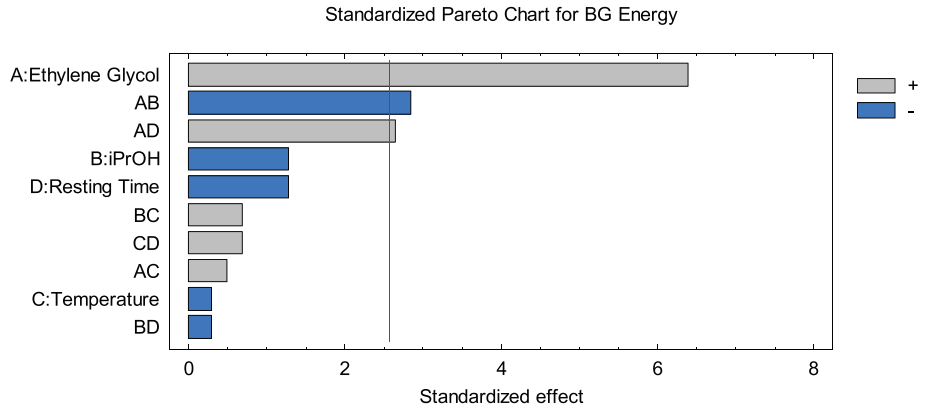

Fig. 3. Pareto chart for the response BG energy.

In Fig. 3 it is possible to notice that three factors showed a *P*-value <0.05, indicating that their influence on the response (band gap energy) can be assessed with a confidence interval > 95%. Focusing on the magnitude of such influence, the ethylene glycol amount represents the most important factor determining the band gap energy. This observation is not unexpected, as the ratio between choline chloride and ethylene glycol determines the formation of the eutectic with a minimum band gap energy, which is not significantly influenced by the presence of 2-propanol. On the other side, the second and the third factors are two combinations, respectively, the combination between ethylene glycol and 2-propanol (AB), and ethylene glycol and resting time (AD). The effect of AD clearly suggests a not-negligible effect of the second HBD, which acts in synergy with the ethylene glycol. As matter of fact, the effect of the 2-propanol on the BG energy, seems to be expressed not directly, but through an influence on the ethylene glycol, which, in turn, directly influence the response. Even more intriguing is the effect of AD on the BG energy. In fact, it indicates that the resting time has an influence on determining the band gap energy and this specific relationship seems to be related to the amount of 2-propanol. For the best of our knowledge, the observation of a variation in the optical properties of a DES depending on its aging, has never been reported so far.

This effect will be much more relevant in the further section, where the more sensible Urbach energy will be discussed.

It is worth to mention that these combined effects can be highlighted only through a DoE approach, followed by a specific multivariate statistical analysis.

3.1.1. Urbach energy statistical analysis

The same multivariate statistical analysis was performed on the response Urbach energy, and the effects of factors along with their combinations are reported in the Fig. 4.

The data represented in the Pareto chart confirm the importance of the resting time on the molecular structure of the system already observed during the band gap energy analysis.

This specific information, if analysed in the context of the degree of disorder of the system, suggests that the aging of a eutectic system has a relevant effect on the increasing of the structural order. It seems evident that the evolution of the structure rearrangement in a fresh-prepared eutectic system is not random but it evolves toward a decreasing of its intrinsic disorder, as a sort of time mediate self-rearrangement. This finding has potentially huge consequences when the structural feautures of a eutectic are pivotal, for example when it is used as non-innocent solvent for chemical synthesys or in catalysis. More research activity will be necessary to define the impact of such aging-driven changes on specific application of selected eutectic systems.

Considering the data acquired, it is possible to perform a further optimization by a statistical technique known as Surface Responding Analysis (SRA). When such approach is employed, the data relative to the selected response (UE in this case) obtained from the DoE dataset are used for determining a desirability function, which in the specific case herein discussed is related to the magnitude of the UE. Higher values of UE correspond to a low desirability and *vice versa*. This approach has been already employed for optimizing eutectic systems [19] as well for optimizing industrial processes [32] as the pectin production from sunflowers [33] the mercerization of cellulose [34] the recycling of waste vegetable oils [35] or for treating contaminated wastewaters [36].

From the experiments conducted on the model substrate, the variation of the UE was determined: it ranges from 120.47 meV to 367.22 meV. In Table 5, the values, respectively, of the predicted and experimentally observed desirability are reported.

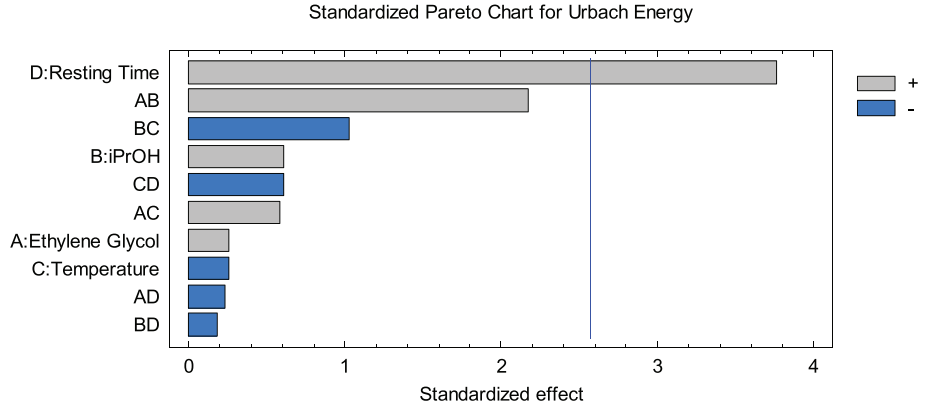

Fig. 4. Pareto chart for the response Urbach Energy.

The data reported in Table 5 allow to build a surface plot which represents the response of the system (in terms of desirability) to the changing of the considered factors (temperature, resting time, ethylene glycol, and 2-propanol amounts).

Looking at the plot reported in Fig. 5, it is possible to understand how for fixed values of temperature (52 °C) and resting time (13*h*), the lower values of Urbach energy are reached at high amounts of glycerol and low of high quantities of *2*-propanol (two red extreme parts of the surface). On the contrary, by reducing the amount of glycerol, the value of the Urbach energy considerely decreases.

Table 5
Desirability function parameters.

Entry	Urbach Energy (meV)	Desirability Predicted	Desirability Observed
1	136.52	0.187523	0.0668826
2	173.75	0.290792	0.217611
3	120.47	0.0	0.00190283
4	238.66	0.586065	0.480405
5	124.15	0.0910855	0.0168016
6	134.06	0.0480288	0.0569231
7	168.09	0.28396	0.194696
8	135.35	0.0543548	0.0621457
9	268.16	0.623424	0.599838
10	367.22	0.782381	1.0
11	133.96	0.340418	0.0565182
12	328.69	0.673849	0.844899
13	139.82	0.0	0.0802429
14	279.53	0.621996	0.64587
15	164.39	0.30146	0.179717
16	248.61	0.431713	0.520688

Fixing the ethylene glycol and 2-propanol amounts, varying the temperature and the resting time, it is possible to observe the huge effect of the resting time on the molecular disorder, expressed by the Urbach energy (Fig. 6), as already observed during the factors analysis.

The multivariate statistical analysis performed clearly shows that it is possible to engineering at the structural level these systems by changing the preparation method. This level of sensibility was never described in previous studies and represents a pivotal information especially when specific eutectic systems are prepared for dedicated applications where the structural features play a crucial role.

4. Conclusions

Five ternary eutectic systems containing one HBA (choline chloride) and two HBD (ethylene glycol and water, glycerol, methanol, ethanol or 2-propanol) have been prepared in different molar ratio and characterized in terms of band gap (BG) and Urbach (U) energies through the UV-VIS-based Tauc plot methodology. The analysis of the energy values for different composition and molar ratios revealed a peculiar behaviour of the system containing as second HBD 2-propanol. A Design of Experiments (DoE) approach combined with a multivariate statistical analysis allowed to going through the relationship between the preparation procedure and the consequent structural disorder of the eutectic system. The effect of the molar ratio between the component, the temperature, the resting time, as well as the effect of their combination on the BG and U energies were assessed through a two-levels full factorial screening design. A never described and not intuitive effect of the resting time was observed on the structural disorder of the eutectic, indicating a not negligible influence of the DES age on its structure. Finally, a Surface

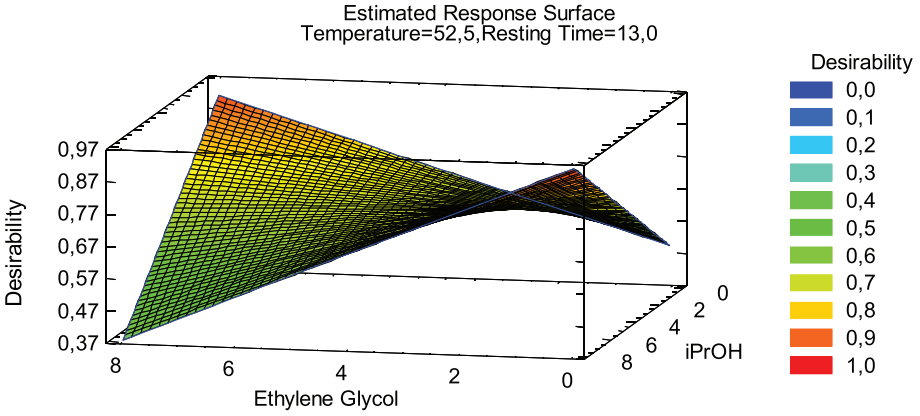

Fig. 5. Surface responding plot for Urbach energy (fixed temperature and resting time).

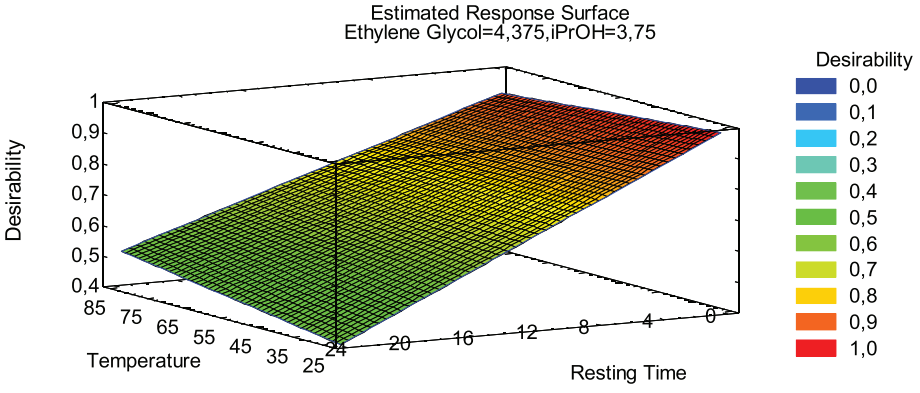

Fig. 6. Surface responding plot for Urbach energy (fixed ethylene glycol and 2-propanol amounts).

Responding Analysis (SRA) was employed for develop a statistical tool which allows to engineer the structural disorder of the system composed by choline chloride, ethylene glycol, and 2-propanol, by a fine tuning of the preparation conditions.

Declaration of Competing Interest

The authors declare that in this work have not any conflicts of interest.

Acknowledgements

All the Authors thank Mr. Alessandro Taras fort the helping in processing the statistical data.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.115717.

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