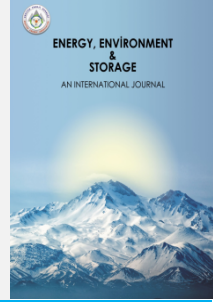




# Energy, Environment and Storage

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## Investigation of Deep Eutectic Solvent Based Super Dielectric Electrolytes for Supercapacitors

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**ABSTRACT.** This study investigates a new type of electrolyte based on deep eutectic solvents. Choline chloride based deep eutectic solvents were prepared and they were used as base ionic solvents for super dielectric theory. Deep eutectic solvent was mixed with a non-conducting material such as fumed silica, alumina. The mixture shows a super dielectric behavior which is used as electrolyte for electrochemical double layer capacitors also known as supercapacitor. The supercapacitor cells were composed of an electrode, a paper-based separator and this super dielectric electrolyte. The electrode of commercial standard supercapacitor is used first as an electrode. Second an electrode slurry was prepared in order to make custom electrode. Then the performance of both cells was investigated. The specific capacitances of cells were measured and the amount of increase at the capacitances was evaluated. The results showed that up to 14-fold increase of the specific capacitances of the commercial supercapacitor have been achieved. Also, up to 12-fold increase of the specific capacitances of our custom-made cells have been achieved. The charge discharge characteristics and ESR values of the cells confirms that the cells show outperforming properties. Deep eutectic solvents based super dielectric electrolytes are very promising electrolytes for high energy density supercapacitors.

**Keywords:** Super Dielectric, Supercapacitor, Equivalent Series Resistance (ESR)

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### 1. INTRODUCTION

The needs of energy storage systems are increasing rapidly nowadays because of developing mobile systems, electrical vehicles and renewable energy system such as solar and wind power generation facilities. The major energy storage systems still use lead acid and lithium-ion batteries. The problem of existing battery systems such as highly cost, short life, safety and difficult production processes increase the needs of promising energy storage systems. One of the possible alternatives of current battery system is electrochemical double layer capacitors [1,2].

Electrochemical double layer capacitors (EDLCs), also known as supercapacitors or ultracapacitors are the most promising energy storage technology [3]. In EDLCs the charge is electrostatically stored at the electrode and electrolyte interface. These devices can be charged and discharged within seconds. They can give high power (up to 10 kW) and a high cycle life (>500,000). Now, the commercial EDLCs contain activated carbon as active

material and electrolytes based on quaternary ammonium salt dissolved in organic solvents (acetonitrile (ACN) or propylene carbonate (PC)). The cell voltage of these EDLCs are 2.3 V and 2.7 V. In order to use EDLCs in today applications, their energy storage capacities need to be improved. The specific energy of EDLCs is defined by the equation  $E=1/2CU^2$ , where C and U are the capacitance and operative voltage of the EDLC, respectively [4-6].

Traditional electrolytes for EDLCs can be generally divided into aqueous, organic, IL and solid- or quasi-solid types [7,8]. The working voltage of a EDLCs is determined by the decomposition of an electrolyte on the electrode surface at high potential. The working voltage of a EDLCs can be raised from 2,3 V to 2.7 V using organic electrolytes instead of aqueous electrolytes. The common working voltage of commercial EDLCs with organic electrolytes is 2.7 V. Because the energy density of a EDLCs is directly proportional to the squared working voltage, the research is focused on developing

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electrolytes with high conductivity, good chemical and thermal stability, and wider electrochemical window. An organic electrolyte is composed of an organic solvent and a supporting electrolyte [9]. The organic solvent must have low volatility, good electrochemical stability and large dielectric constant.

In recent years, as a new type of green electrolyte, Ionic Liquids (IL) has been widely used in EDLCs. IL is featured with wide electrochemical window, relatively high conductivity and ion mobility. Deep Eutectic Solvents (DES) are also a new type of green electrolyte which has wide electrochemical windows as alternative of ILs. DES can be produced cost effectively and easily. Super dielectric materials are also newly proposed subject and getting more interest recently. Their dielectric constant can be between 106-1012 [10].

In the present study, we synthesized a novel deep eutectic solvent-based super dielectric electrolytes and used on both standard commercial and custom-made supercapacitor electrodes to demonstrate its effect. As an outstanding result, the synthesized super dielectric electrolyte exhibited the specific capacitance of up to 14-fold increase for standard commercial supercapacitor electrodes and 12-fold increase for custom made supercapacitor electrodes.

## 2. EXPERIMENTAL

The chemicals choline chloride (ChCl), urea, monoethylene glycol, glycerin, malonic acid, boron nitride and alumina were purchased from Sigma Aldrich, Germany. Fumed silica, xylitol and carboxymethyl cellulose (CMC) were purchased from Introgen, Türkiye and the activated carbon and Styrene-butadiene rubber (SBR) were obtained from Nanografi, Türkiye. The commercial supercapacitors were purchased from Greencap, South Korea. All chemicals were used without further purification.

### 2.1 Preparation of Deep Eutectic Solvents (DES)

Deep eutectic solvents (DESs) are new class of ionic liquid (IL) because they share a lot of characteristics and properties with ILs [11-15]. DESs contain large, nonsymmetrical ions. They have low lattice energy and low melting points. They are usually obtained by the complexation of a quaternary ammonium salt with a metal salt or hydrogen bond donor. Different type of DESs can be made by using suitable chemicals [16].

The following DESs were prepared by mixing at magnetic stirrer at 80 C about 1 hour. Then they were slowly cooled to room temperature. The molar ratios were used in order to make deep eutectic solvents as **ChCl + Urea** (1:2); **ChCl + Glycerin** (1:2); **ChCl + MEG** (1:2); **ChCl + Malonic acid** (1:2); **ChCl + Xylitol** (1:2). These are well known DESs and their characterization is available at scientific literature [12-14]. Our interest is to see the super dielectric properties of DESs when they mixed with non-conducting powders.

The electro chemical voltage window of these DESs is given Table 1. The supercapacitor cells are charged

according to this table. This EPWs were accepted as a cell voltage but, the real stable cell voltages were below these voltages because of water content in the DESs. DESs contain water since the chemicals that we use are not anhydrous. If the chemicals are anhydrous and the electrolyte is injected in controlled atmosphere, EPWs will be at Table 1. We did not use a controlled atmosphere to see the performance of DESs at open atmosphere.

**Table 1:** The EAL, ECL and EPWs of ChCl Based DESs [17-19].

DESs	E <sub>AL</sub> (V)	E <sub>CL</sub> (V)	EPWs (V)
ChCl + glycerol	-2.21	1.38	3.59
ChCl + urea	-2.75	1.54	4.29
ChCl + MEG	-2.35	1.26	3.61
ChCl+malonic acid	-2.55	1.70	4.25
ChCl+xylitol	-2.67	1.66	4.33

### 2.2 Preparation of Super Dielectric Electrolyte

Super Dielectric Materials subject is a new theory that can be verified by experimental studies [20-22]. The super dielectric theory was first discovered by Prof. Jonathan Phillips and his friends.

According to theory, any porous, non-conducting material with a liquid containing a high concentration of ionic species will potentially be a Super Dielectric Material (SDM). The dielectric constant of SDM can be measured by making a parallel plate capacitor. The dielectric constant can be obtained from the time constant and this standard equation:

$$C = \epsilon_0 \epsilon_R A/d$$

where  $\epsilon_0$  is the permittivity of free space ( $8.85 \times 10^{-12}$  F/m) and  $\epsilon_R$  is the dielectric constant. The area of the plate surface is A and the distance between the two electrode surfaces is d. The measured dielectric constant is between  $5 \times 10^9 - 1.2 \times 10^{11}$  [23].

Different type electrostatic capacitors (EC) were made by super dielectric materials that have energy density greater than 200 J. cm<sup>3</sup> [24,25].

According to Super Dielectric Materials theory, the electrolyte is needed to mix with any non-conducting nano powder. Our deep eutectic solvents were mixed with fumed silica for one sample, mixed with boron nitride for second sample. The produced electrolyte has gel like properties. This electrolyte is our new electrolyte for EDLCs. Five different types of DESs and three different types of non-conducting powders were used in order to make 15 different types of super dielectric electrolyte. Table 2. shows these super dielectric electrolytes (Fig. 1).

**Table 2.** Our new super dielectric electrolytes

DESS	Non conducting powder
ChCl+glycerol	Fumed Silica
ChCl+urea	Fumed Silica
ChCl+MEG	Fumed Silica
ChCl+malonic acid	Fumed Silica
ChCl+xylitol	Fumed Silica
ChCl+glycerol	Boron Nitride
ChCl+urea	Boron Nitride
ChCl+MEG	Boron Nitride
ChCl+malonic acid	Boron Nitride
ChCl+xylitol	Boron Nitride
ChCl+glycerol	Alumina
ChCl+urea	Alumina
ChCl+MEG	Alumina
ChCl+malonic acid	Alumina
ChCl+xylitol	Alumina



**Fig. 1** - Some samples of prepared DESs

**2.3. Fabrication of EDLC cells**

EDLC cells were prepared 2 different methods. First a commercial supercapacitor cell used. Second an electrode was made by electrode slurry.

**2.3.1. Using Commercial Cell Electrode**

A commercial 2,7V, 500 F supercapacitor as shown in Fig. 2 was disassembled, and its electrode and paper type separator in Fig. 3 were used to fabricate of supercapacitor

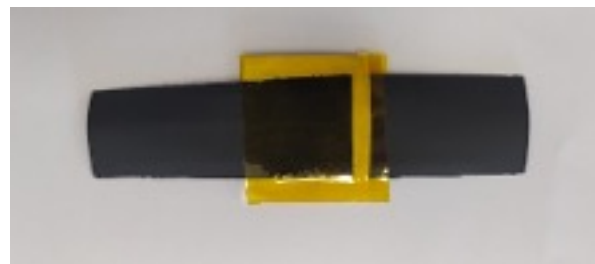


**Fig. 2** - Disassembled standard commercial supercapacitor



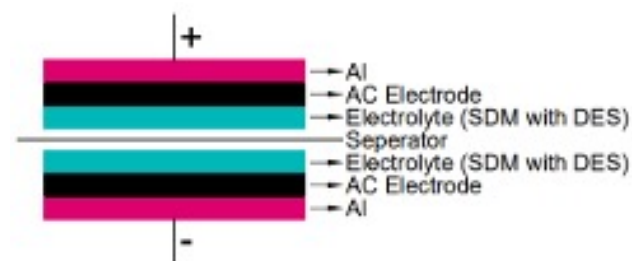
**Fig. 3** - The electrode and separator taken from disassembled standard commercial supercapacitor.

First, before disassembling process, its real capacity was measured by ZTE electronic load via PC. Its real capacity was between 380-430 F. The size of the electrode of the cell were measured. It is 4.5 cm x 77 cm. Then the specific capacity was calculated, and it was found 0,32 F/cm<sup>2</sup> as expected. In order to compare our results with the commercial cell easily, the electrode was cut 1 cm x 2 cm sizes and a new cell was constructed by using this electrode by using 1 cm x 1 cm of the electrode as shown in Fig. 4.



**Fig.4** - The finished cell

Then, our deep eutectic solvent based super dielectric electrolyte was injected on two electrode and the paper separator were used and the cell were constructed. Then the cell insulated by Kapton tape. Fig. 5 shows our cell structure used in our measurements.



**Fig. 5** - Cell structure of the supercapacitor

**3. MAKING CUSTOM ELECTRODE:**

**3.1. Preparation Electrode Slurry**

Three items are necessary for making a conductive ink. These are a solvent (water, ethanol, DMF etc.), a binder (water-based adhesive, etc.) and an active ingredient (activated carbon, graphene, a conductive material (carbon black, Super-P etc.) We used the same and common formula that is used at commercial supercapacitors. It is 80% Activated Carbon +10% Conducting Material + %10 Binder. We used activated carbon as active ingredient, graphite powders as conductive material and CMC+SBR mixture as binder. First, 0.25g of CMC added to 50 ml DI water and stirred at magnetic stirrer at 70 C about 1 hour then 0.25g of SBR added slowly into this solution and stirred about 1 hour at 70 C. 8,5g of activated carbon was mixed with 1 g of graphite powder. Then this mixture added to binder (CMC-SBR). The mixture was stirred at

vacuum mixer about 4 hours. Then the electrode slurry was obtained (Fig. 6). The viscosity of the slurry was measured with a digital viscosity meter. The viscosity was adjusted between 4000-5000 cpt by adding more DI water and vacuum mixing.



**Fig 6-** Electrode slurry and its visual testing on paper.

### 3.2. Coating Slurry

Copper or aluminium strips were cut to the desired scale. The surface of strip was cleaned with ethanol. The prepared electrode slurry was coated by Dr. Blade on a special Al foil (Fig 7). The coating thickness is 100  $\mu\text{m}$ . Then allowed to dry in an oven at 50 °C about 5 hours.



**Fig. 7 -** Film coating machine with drying oven (TOB-VFC-150) and coated electrode.

### 3.3. Calendaring Electrode

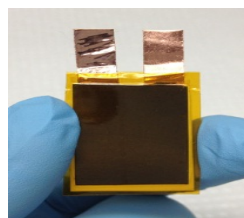
The coated electrode was processed at hot roll press (Fig. 8). The thickness of active material was reduced to 65  $\mu\text{m}$ . After this process the surface of the electrode became smooth and perfect. Then the electrode was vacuum dried at 60 C about 2 hours in order to remove the water content. Then the electrode was cut by a mechanical blade under hydraulic press.



**Fig. 8 -** Hot roll press machine (TOB-JS100L) and prepared electrode.

### 3.4. Making Cells

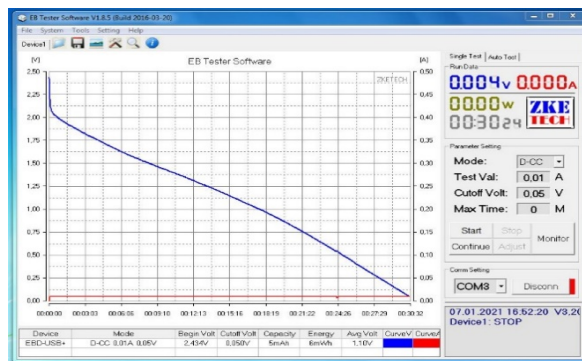
To make a supercapacitor, the cut electrodes were stacked with a NKK 4030 type separator then our electrolyte was added. The cell was insulated by a Captone tape. One of made supercapacitors is shown in Fig. 9. In order to understand the standard cell capacity of our electrode, 3M KOH solution was used as electrolyte first. Then, its specific capacitance was measured. It is 0.4 F/cm<sup>2</sup>



**Fig. 9 -** Finished Cell

### 3.5. Instrumentation

The ESR of the cell were measured by MESR-100 model digital ESR meter, 100 kHz in circuit test. The capacity of the cell was measured by charging and discharging the cell. The cell was charged by a DC power supply up to cell voltage of our electrolyte and the cell were discharged by ZTE electronic load via PC under constant current. According to  $C = \Delta t * I / \Delta V$  equation, the cell capacity was calculated. This process was repeated several times. It showed that, the cells are EDLCs but its discharge voltage characteristic is a little bit different. According to SDM theory, the discharge voltage is effective below 1V. Fig. 10 shows a discharge graph of ChCl + MEG + Boron Nitride Sample.



**Fig. 10 -** ChCl + MEG + Fumed Silica Cell.

The calculated Cell capacity is about 8,77 F for 2 cm<sup>2</sup> cell. The specific capacitance is 4,39 F/cm<sup>2</sup>



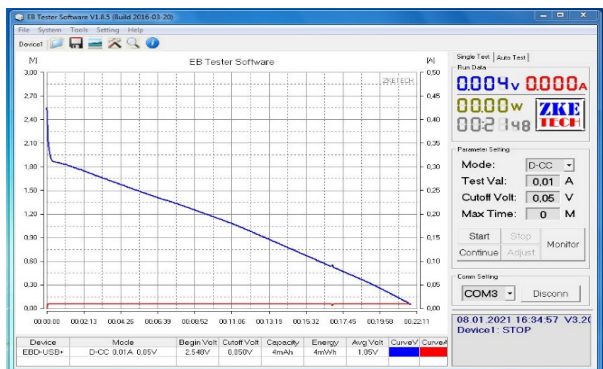


Fig. 11 - ChCl + MEG + Boron Nitride Cell.

The calculated cell capacity is 7,07 F for 2 cm<sup>2</sup> cell. The specific capacitance is 3,54 F/cm<sup>2</sup> for commercial supercapacitor electrode cell (Table 3). The best one is ChCl-MEG- Fumed Silica cell, it has about 14-fold increased capacity over KOH electrolyte. Table 4 shows specific capacity of custom-made supercapacitor cells. The best one is ChCl-MEG cell, it has about 12-fold increased capacity over KOH electrolyte. ChCl-MEG shows the best performance because of low viscosity and high conductivity of MEG.

Table 3. Specific capacity of commercial supercapacitor electrode made cells.

Electrolyte Type	Specific Capacitance (F/cm <sup>2</sup> )
ChCl+glycerol - Fumed Silica	2,90
ChCl+urea - Fumed Silica	3,11
ChCl+MEG - Fumed Silica	3,54
ChCl+malonic acid - Fumed Silica	2,59
ChCl+xylitol - Fumed Silica	2.33
ChCl+glycerol - Boron Nitride	2.03
ChCl+urea - Boron Nitride	2,09
ChCl+MEG - Boron Nitride	3,25
ChCl+malonic acid - Boron Nitride	2.17
ChCl+xylitol - Boron Nitride	1,78
ChCl+glycerol - Alumina	2.07
ChCl+urea - Alumina	1.98
ChCl+MEG - Alumina	2,60
ChCl+malonic acid - Alumina	1.55
ChCl+xylitol - Alumina	1,67

Table 4. Specific capacity of Custom-made supercapacitor cells

Electrolyte Type	Specific Capacitance (F/cm <sup>2</sup> )
ChCl+glycerol - Fumed Silica	3,64
ChCl+urea -Fumed Silica	3,21
ChCl+MEG- Fumed Silica	4,27
ChCl+glycerol -Boron Nitride	3.73
ChCl+urea -Boron Nitride	3,69
ChCl+MEG -Boron Nitride	4,77

#### 4. RESULTS AND DISCUSSION

ESR values of the cell were measured with a digital ESR meter. CV measurements were taken with a cyclic voltmeter device.

##### 4.1. Electrochemical properties of EDLCs cells

ESR Values of Some Capacitors

470 uF 16 V standard capacitor 11 mΩ

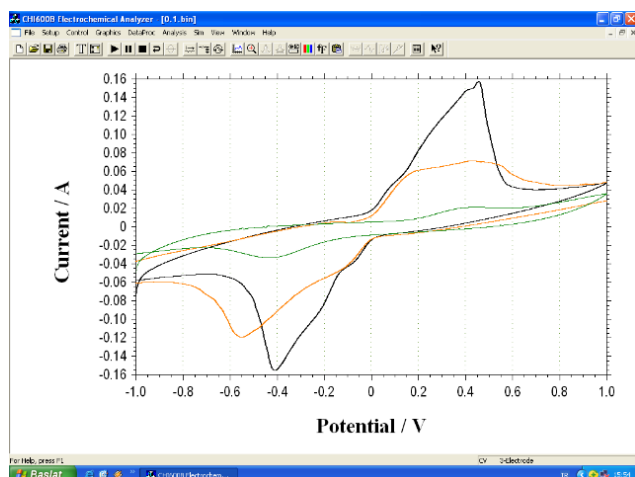
10F 2.7 V supercapacitor 30 mΩ

ESR value of commercial supercapacitor cell is 30 mΩ

Table 5. ESR values of same samples (commercial cell electrode)

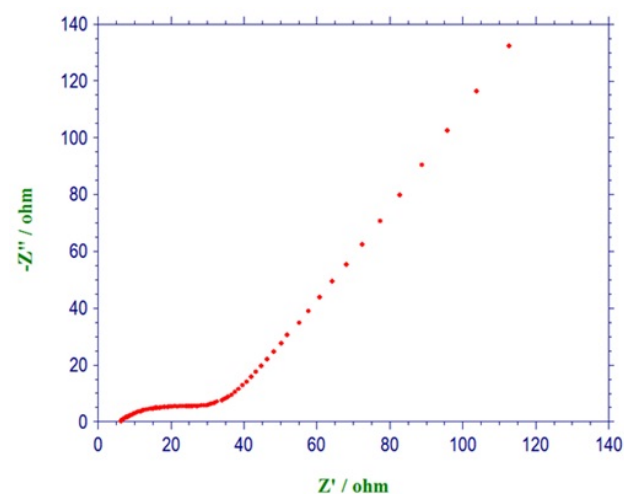
DESS - Non conducting powder	ESR(Ohm)
ChCl+glycerol - Fumed Silica	44,6
ChCl+urea - Fumed Silica	34,2
ChCl+MEG - Fumed Silica	18,6
ChCl+malonic acid - Fumed Silica	36,4
ChCl+xylitol - Fumed Silica	46,1
ChCl+glycerol - Boron Nitride	43,2
ChCl+urea - Boron Nitride	33,7
ChCl+MEG - Boron Nitride	16,4
ChCl+malonic acid - Boron Nitride	34,5
ChCl+xylitol - Boron Nitride	48,7
ChCl+glycerol - Alumina	45,0
ChCl+urea - Alumina	37,4
ChCl+MEG - Alumina	22,2
ChCl+malonic acid - Alumina	33,9
ChCl+xylitol - Alumina	45,1

As seen Table 5, ChCl + MEG samples have lower ESR because of the viscosity. Therefore, ChCl + MEG based electrolytes were studied. In order to understand our cell shows a real EDLC characteristics, a CV measurement was made. Fig. 12 shows that our cell shows a EDLC characteristics.



**Fig. 12** - A sample from CV measurement of our cells (on commercial supercapacitor electrode) (8 Black: ChCl + MEG with Fumed Silica, Red: ChCl + glycerol with Fumed Silica, Green: ChCl + urea with Fumed Silica).

Nyquist plots of ChCl-MEG type DES electrolyte - fumed silica cell is shown in Fig.13. It shows the behaviour of an ideal capacitor at straight line parallel to the imaginary axis.



**Fig. 13** - Nyquist plots of ChCl-MEG -Fumed silica electrolyte cell (on commercial supercapacitor electrode).

## 5. CONCLUSIONS

The commercial supercapacitor cells have 0,32 F/cm<sup>2</sup> specific capacitance and it gives 4,7 Wh/kg energy density.

Our **ChCl + MEG + Fumed Silica cell** (on commercial supercapacitor electrode) showed 4,54 F/cm<sup>2</sup> specific capacitance and it is about 14 fold increase and ChCl +MEG +Boron Nitride cell (on custom made supercapacitor electrode) showed 3,54 F/cm<sup>2</sup> and it is about 12 fold increase. The capacity increase comes from high dielectric constant of ChCl-MEG and non-conducting powder mixture.

The energy densities will be less than 12-14-fold since the electrolyte volume is bigger than the standard electrolytes. The electrolyte contains fumed silica and boron nitride. They increase the volume of the cells.

Deep eutectic solvent based super dielectric electrolyte works both commercial supercapacitor cells electrode and custom made cells electrode. But ESR values are very big because of high viscosity and content of nonconducting powders in electrolyte.

ChCl + Glycerol and ChCl + Urea showed worse ESR because of viscosity. By adding other chemicals to DESs, ESR can be lowered.

CV plot and Nyquist plot confirm the capacitive behaviour of the cells.

Specific capacitance increases with lower viscosity. ChCl-MEG gives the highest specific capacitance because of low viscosity and high ionic conductivity.

Deep eutectic solvent based super dielectric electrolytes are very promising for making high energy density supercapacitors at their usage in hydrogen energy systems.

## Acknowledgements

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