SUPPLEMENTARY MATERIAL:

The presence of water in the sample will have an impact on the physical properties of Choline[$FeCl_4$]. It is then a crucial matter to clarify whether or not any water was present. To this end several tests were performed to prove that no water was present in the compound and that its stability is sustained over time.

1. Elemental analysis for Choline[$FeCl_4$] indicates a good agreement between calculated and expected molecular formula; $C_5NH_{14}OCl_4Fe$
2. The decomposition curves of TGA/DTG of Choline$FeCl_4$ do not reveal any initial weight loss below 100 ºC, associated with water absorbed on the surface from the atmosphere.
3. All further heating and cooling cycles of DSC were reproducible and were similar to the first cooling and second heating, as is mentioned in the manuscript
4. Powder X-ray thermodiffractograms of Choline$FeCl_4$ were recorded in air heating up from 225 to 400 K and cooling down to 300 K. During the temperature decrease the reversibility of the second process compared with the heating one was preserved. In addition, the presence of ice peaks is not detected.
5. Raman spectroscopy was also used to investigate the absence of water in Choline[$FeCl_4$]. In all samples tested, we observed the non-presence of both strong peaks near to 3230, 3390, 3510 cm^{-1} and a weak peak at about 1581 1640 cm^{-1}, which confirmed the absence of hydration water [1], as can be observed in Fig.S.1 of the supplementary material.
6. We have not seen any changes in the electrochemical response if the heating and cooling cycles are compared. The material presents a high stability in air in the measured temperature range. We have carried out the impedance measurements in a heating and cooling cycle (maintaining the sample 2 h at 450 K) and the obtained similar behaviour during this cycle.

[1] D. M. Carey,G. M. Korenowski, , J. Chem. Phys. 1998, 108(7), 2669-26675.

Figure Captions

Figure S.1. Raman spectra of Choline[$FeCl_4$] between 100 and 3500 cm^{-1}.

Figure S.2. D2B neutron data of Choline[$FeCl_4$] (λ□=1.5938 Å) at 300 K.

Figure S.3. Low temperature ZFC magnetic susceptibility at difference fields from 0.5 to 50 kOe. The inset shows the evolution of Neel temperature (T_N) with the applied magnetic field.

Figure S.4. Temperature dependence of the real (χ') and imaginary (χ'') components of the ac magnetic susceptibility for Choline[$FeCl_4$] measured under an ac field h_{ac} = 3 Oe and frequencies from 50 to 10000 Hz.

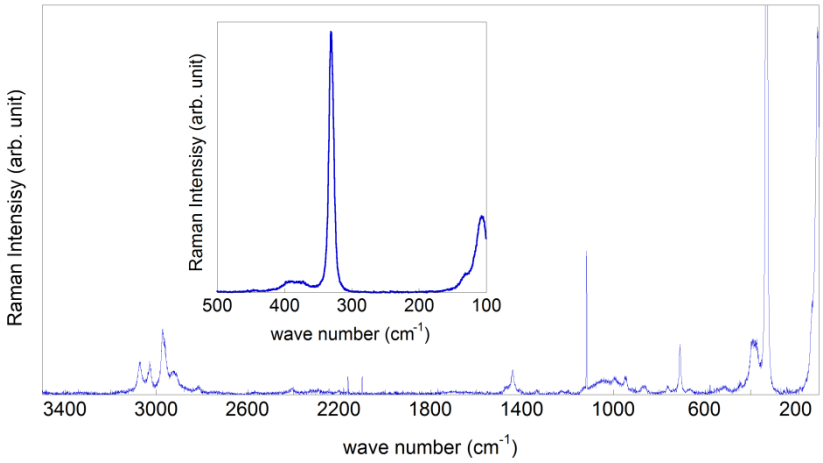
Raman Intensisy (arb. unit)
Raman Intensisy (arb. unit)
500
400
300
200
100
wave number (cm-1)
3400
3000
2600
2200
1800
1400
1000
600
200
wave number (cm-1)

Fig S.1

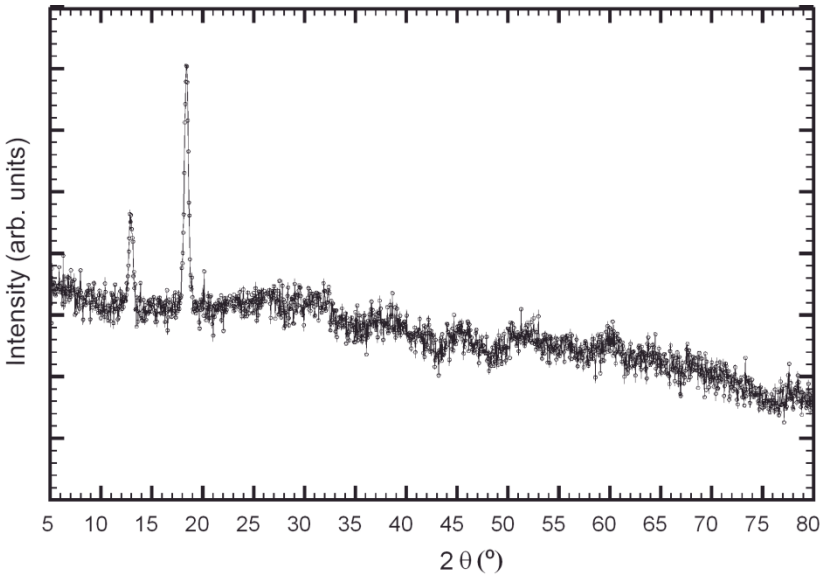
Intensity (arb. units)
5
10
15
20
25
30
35
40
45
50
55
60
65
70
75
80
2 θ (°)

Fig S.2

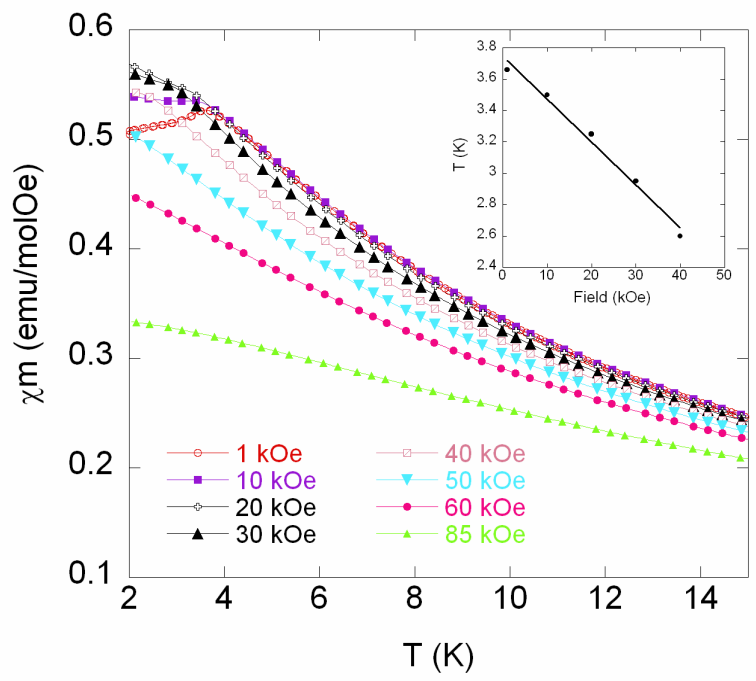
χm (emu/molOe)
T (K)
1 kOe
10 kOe
20 kOe
30 kOe
40 kOe
50 kOe
60 kOe
85 kOe
T (K)
Field (kOe)

Fig. S.3

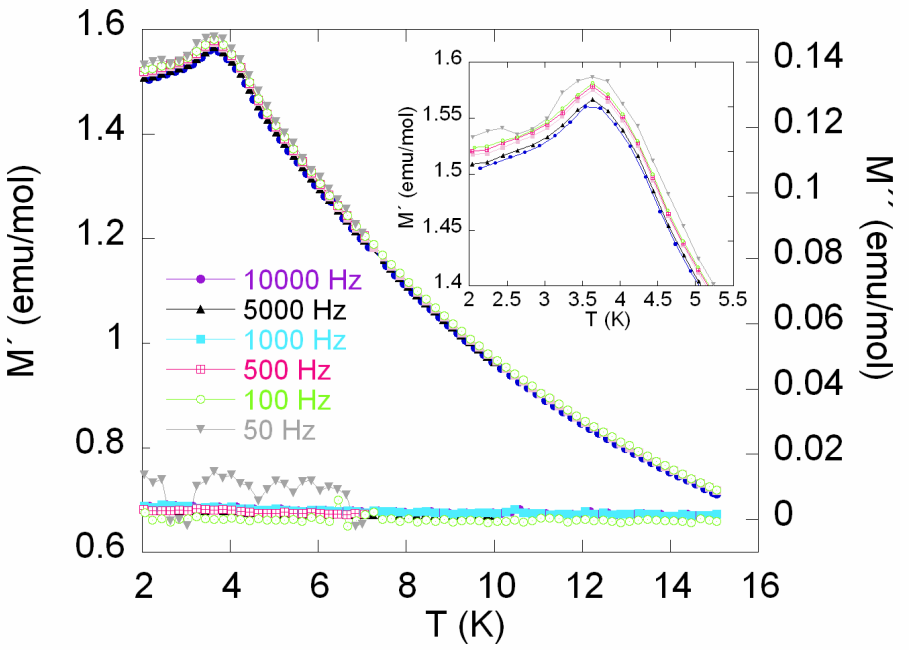
M´ (emu/mol)
M´´ (emu/mol)
T (K)
10000 Hz
5000 Hz
1000 Hz
500 Hz
100 Hz
50 Hz
M´ (emu/mol)
T (K)

Fig. S.4