The polymorphic system of Butethal is composed of four different forms.

The system is part of a family of the Barbiturates, which comprise a class of drugs which exhibit widespread polymorphism. In an early survey it was found that of the barbiturates used medicinally about 70% were polymorphic. In the course of investigating a number of compounds in this family we studied on the molecule 5-ethyl-5-n-butylbarbituric acid.

We will present the solid state characterization of the different forms, utilizing a variety of techniques, including: single crystal X-ray structure analysis, X-ray powder diffraction, solid state NMR thermal analysis, graph set analysis, and lattice energy calculations, in an effort to understand and to control the structure and polymorphic form.

Two new acid selenates have been synthesized: \((\text{C}_6\text{H}_9\text{N}_2)(\text{HSeO}_4)\) (I) and \((\text{C}_6\text{H}_11\text{N}_2)(\text{HSeO}_4)\) (II), where \(\text{C}_6\text{H}_9\text{N}_2\) is 1,4-diazabicyclo[2.2.2]octane (DABCO) and \(\text{C}_6\text{H}_{11}\text{N}_2\) is N,N'-dimethylpipperazine (DMPPA).

Crystals of I and II were grown by slow evaporation of aqueous solutions with stoichiometric ratio of corresponding amine and selenic acid.

I and II were investigated by X-ray single crystal diffraction on an IPDS (Stoe) and a STADI-4 (Stoe), respectively. Unit cell parameters are:
- for I: \(a=7.447(2)\) Å, \(b=17.755(3)\) Å, \(c=10.269(3)\) Å, \(\beta=111.20(3)^\circ\), \(V=1266.0(6)\) Å³, \(Z=4\), space group \(P2_1/n\).
- for II: \(a=6.78(1)\) Å, \(b=12.20(3)\) Å, \(c=8.230(4)\) Å, \(\beta=108.1(3)^\circ\), \(V=646.9(4)\) Å³, \(Z=2\), space group \(P2_1/c\).

Asymmetric unit of I contains one cation \([\text{DABCO}]^+\) and two kinds of anions \([\text{HSeO}_4]^-\). One of anions was found to be disordered around Se-O bond. Both kinds of tetrahedra are connected by two hydrogen O...O bonds 2.57(1) Å and 2.60(1) Å forming infinite chains along \(a\). DABCO cations connect these chains along \(b\) by two hydrogen N...O bonds 2.652(8) Å and 2.705(8) Å to layers parallel to \(001\) plane. There are van-der-Waals interactions between layers.

In the structure II there are dimethylpipperazinium cations in chair conformation and tetrahedral HSeO₄ anions. These tetrahedra form infinite chains along \(c\) axis by means of hydrogen O...O bonds 2.57(1) Å and 2.60(1) Å forming infinite chains along \(a\). DABCO cations join these chains by hydrogen N...O bonds 2.78(1) Å. Thus packed layers are formed parallel to \(010\) plane. The van-der-Waals interactions are present between layers.

The infinite chains of HSeO₄ tetrahedra are the typical case of hydrogen bonding system for acid selenates. This case is compared to the cyclic hydrogen bonding systems, for instance closed dimers.

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