

ARTICLE

Synthesis and properties of novel acetamidinium salts

Zdeněk Jalový,^a Robert Matyáš,^a Jan Ottis,^a Aleš Růžička,^b Petr Šimůnek,^c and Miroslav Polášek^d

Acetamidines are starting materials for the synthesis of synthesizing many chemicals, some of which go on to be substances, such as imidazoles, pyrimidines and triazines, which are further used for synthesis of biochemically active compounds and as well as energetic materials. Acetamidinium chloride, which is hygroscopic, is currently one of the only commercially available acetamidinium salts. The aim of this study was to synthesize and characterize a range of acetamidinium salts that will in order to allow overcome the inconvenience connected associated with acetamidinium chloride to be avoided acetamidinium chloride, which is the only commercially available acetamidinium salt. Acetamidinium salts were synthesized. The acetamidinium salts were characterized and characterized by with elemental analysis, mass spectrometry, NMR and, in the case of energetic salts, differential thermal analysis (DTA). The structures of several previously unknown acetamidinium salts were determined established by X-Ray diffraction analysis. Hygroscopicities of eight of the acetamidinium salts were monitored over time at a 90% humidity of eight acetamidinium salts were evaluated. The different hygroscopicity values obtained of hygroscopicity were corroborated by the results of crystal structure analysis structures determined by X-ray analysis. We found that the acetamidinium salts with two-dimensional (2D) layered structures were (acetamidinium nitrate, formate, oxalate and dinitromethanide) show a lack of not highly hygroscopic. These were the nitrate, formate, oxalate, and dinitromethanide acetamidinium salts, and However, the compounds with a 3D-type of structure containing rather large cavities were highly hygroscopic. These were the (acetamidinium chloride, acetate, sulphate, and perchlorate acetamidinium salts) and possessing rather large cavities are quite hygroscopic.

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Introduction

Acetamidines are used as starting reagents starting materials in the synthesis of a number of many chemicals substances, such as imidazoles, pyrimidines, and triazines, which are further then used for synthesis of biochemically active or energetic compounds.^{1–5} In the field of energetic materials, acetamidine is employed in a starting material for the synthesis of 2-methoxy-2-methylimidazolidine-4,5-dione⁶ and 2-methylpyrimidine-4,6-diol.^{7–9} Both are further transformed to 2,2-dinitroethene-1,1-ethenediamine, also known as FOX-7 or DADNE⁷, which is an energetic material explosive with low sensitivity to external stimuli.¹⁰

The free base form of acetamidine is hygroscopic and decomposes into ammonia and acetonitrile at higher temperatures,¹¹ and produces a acetamidinium carbonate is formed within during 24 h one day when acetamidine is exposed to air at room temperature, when stored in contact with air.¹² Therefore, it is therefore unsuitable as a starting material, so for synthesis and the use of an acetamidinium salt is necessary for synthetic reactions.

Acetamidinium chloride (1) is one of the only commercially available salts of acetamidine and is the most commonly used and commercially available salt of acetamidine is acetamidinium chloride (1). It is prepared by the Pinner method from acetonitrile and alcohol in the presence of hydrogen chloride, followed by addition of ammonia is added to the iminoether intermediate to yield an iminoether.¹³ Many synthetic routes for acetamidines have been reviewed.^{20,21} Reaction of acetonitrile with cobalt or nickel nitrates and oximes gives yields acetamidinium nitrate (2).^{14,15} Another easily accessible acetamidinium salt, acetamidinium acetate (3), is readily prepared by the reaction from triethyl orthoacetate, ammonia, and ammonium acetate.¹⁶ This method is convenient for both laboratory and industrial scale synthesis, and use of the acetate may be further transformed into yield other salts, such as the formate (4)¹⁷, sulfate (5)¹⁸, and dinitromethanide salts (6).¹⁹ Many synthetic routes for acetamidines have been reviewed [20,21].

The main disadvantage of acetamidinium chloride is that it is its relatively hygroscopic high hygroscopicity. The formation release of the free base in methanol by using the use of sodium methoxide produces will produce sodium chloride, which is partially soluble in this solvent (~1 g/100 mL).²² The presence of any chloride source is unfavourable in certain syntheses, such as e.g. nitrations, and the complete removal of chloride is tedious.⁶

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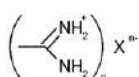
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Here, we describe the synthesis, crystal X-ray structure, hygroscopicity, and thermal stability of several of the acetamidinium salts shown in [Figure 1](#).

Results and discussion

Synthesis



- | | | |
|------------------------------|---|------------------------------|
| 1 X=Cl, n=1 | 4 X=HCOO, n=1 | 7 X=(COO) ₂ , n=2 |
| 2 X=NO ₃ , n=1 | 5 X=SO ₃ , n=2 | 8 X=ClO ₄ , n=1 |
| 3 X=CH ₃ COO, n=1 | 6 X=CH(NO ₂) ₂ , n=1 | 9 X=HSO ₄ , n=1 |

Fig. 1 List of acetamidinium salts studied/analyzed in this study.

We previously reported a procedure for the preparation of acetamidinium sulphate (5) from 1 via an ion exchange reaction from acetamidinium chloride (1) was earlier described by us.²² For our purposes, it may be considered a universal method for the preparation of acetamidinium salts from 1 (Fig. 2). We used this procedure to synthesize was now used for the synthesis of the nitrate (2, as well as) and acetamidinium the oxalate (7). Based on a previously reported method for the preparation of 5,¹⁸ we obtained acetamidinium perchlorate (8) from 3 and perchloric acid. Thus, it may be considered as a universal method for the preparation of acetamidinium salts starting from 1 (Figure 2).

The method used in the preparation of (5),¹⁸ starting from acetamidinium acetate and based on the reaction of the latter with an acid stronger than acetic acid, was now successfully used in the preparation of acetamidinium perchlorate (8) from 3 and perchloric acid. Acetamidinium perchlorate (We also prepared 8) was also prepared from 5 by an ion exchange reaction with barium perchlorate in water (Figure 3). Acetamidinium formate (4) was prepared from trimethyl orthoacetate and ammonium formate. A similar method has been published earlier by Taylor for preparation of 3.¹⁶

The method starting with acetamidinium acetate (3) based on the reaction with a stronger acid than the one we used (acetic acid) for acetamidinium sulphate (5)¹⁸ was now successfully used for preparation of acetamidinium perchlorate (8). This salt was also prepared from 5 by an ion exchange reaction with barium perchlorate in water (Figure 2).

Acetamidinium formate (4) was prepared from trimethyl orthoacetate and ammonium formate. A similar method has been published earlier by Taylor for preparation of 3.¹⁶

Hygroscopicities

The acetamidinium salts were weighed and stored under 90% humidity at 30 °C²³ for 1–21 days. Samples of ammonium acetate (10), guanidinium nitrate (11), and guanidinium chloride (12) were stored under identical conditions. The

hygroscopicities of the samples of acetamidinium salts were determined at 90% humidity and 30 °C²³ and to be the percent weight increase compared with the weight of the original sample. The results are summarized in Table 1, and the changes in hygroscopicity (%) over time are plotted in the comparison of these results with ammonium acetate (10), guanidinium nitrate (11) and guanidinium chloride (12) are represented as

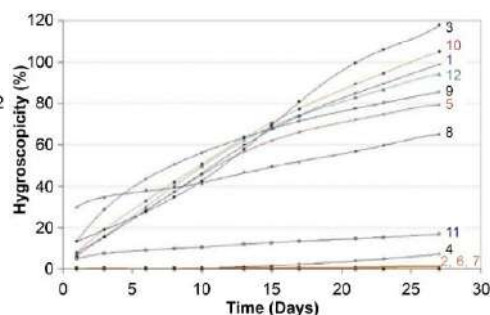


Fig. 24 Changes in the hygroscopicities of acetamidinium salts (1–8) over time and comparison with ammonium acetate (9), guanidinium nitrate (11), and guanidinium perchlorate (12).

the weight increase compared with the weight of the original sample, expressed in %. The results are given in Figure 4, and values for certain days are presented in Table 1. In the case of compounds with known structure determined by X-ray diffraction (XRD) techniques analysis, information about the spatial structure is also included. The influence of the structure on hygroscopicity is discussed later in this report.

X-Ray crystallography

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Sudhakar, C., et al. "Crystal growth, optoelectronic and biological properties of acetamidinium compounds: experimental and computational approaches." *Journal of Materials Science: Materials in Electronics* 34.3 (2023): 183.

Piper, Samantha L., et al. "Probing the secrets of hydrogen bonding in organic salt phase change materials: the origins of a high enthalpy of fusion." *Materials Advances* 2.23 (2021): 7650-7661.

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The acetamidinium cations may frequently serve as a counterions for a wide variety of anions, like simple halogenides, carboxylates, and complex metal anions and others. The parent acetamide is characterized by reveals

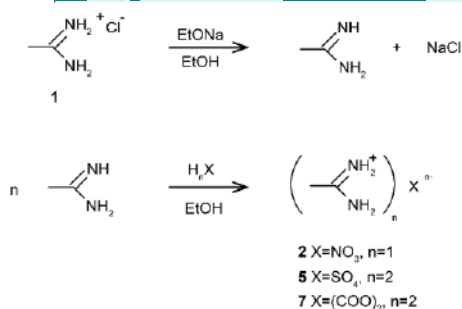


Fig. 42 Preparation of acetamidinium nitrate (2), sulfate (5), and oxalate (7) from 1.

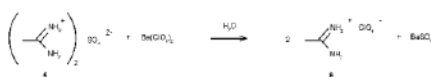


Fig. 3 Preparation of acetamidinium perchlorate (8) from 5 by ion exchange.

large cavities and an extensive network system of hydrogen bonding within its structure. The distances between the pivot carbon atom and the amino and imido nitrogen atoms are pivot carbon atom 1.344 Å and 1.298 Å, respectively and the amino and imido nitrogen atoms are rather distinct (1.344 Å for C-NH₂ and 1.298 Å for C=NH group).²⁴

The hydrogen bridging observed in acetamidinium chloride (1),²⁶ acetamidinium sulfate (5),¹⁸ and an one of the polymorphs of acetamidinium (2-hydroxyethoxy)acetate polymorph,²⁵ results in three-dimensional (acetamidinium chloride (1),²⁶ and acetamidinium sulphate (5)¹⁸ revealed 3D) structures with large cavities. On the other hand, acetamidinium tetrazolate²⁷ and acetamidinium dinitromethanide (6)¹⁹ show are two-dimensional (2D), the staircase-like 2D structures. Interesting examples are acetamidinium hexafluorosilicates, germanates, stannates, and titanates²⁸ are interesting examples of 2D structures. Other examples include the Re-Se cluster-acetamidinium adducts,²⁹ in which where multicentre Re-NH...F or NH...Se contacts were found.

For two of the compounds studied in this study, we used XRD analysis to determine the molecular crystal structures were determined by X-ray crystallography techniques of several acetamidinium salts. Acetamidinium oxalate (7), shown in Figure Figure 5, has a 2D structure with comprising interconnected layers interconnected with not too extensive/limited H-bonding. In contrast, acetamidinium perchlorate (8, Figure Fig. ure 6) has a 3D structure with layers that were interconnected with extensive H-bonding.

The

The perchlorate and oxalate structures were rather unique in the set of among the acetamidinium structures determined/examined. The molecular structure of the acetamidinium oxalate consists of is made up of two mutually similar acetamidinium units and one oxalate ion. All these ions participate in both compounds are interconnected by extensive H-hydrogen bonding systems. In the oxalate (7), eight- and fourteen-membered rings are formed by 7, as shown in Figure (7). Acetamidinium perchlorate (8) primarily forms rings with 22 members, as shown in are the main element of the perchlorate (8) structure (Figure 8). In our analysis, the acetamidinium C-NH₂ group formed an H-bond with a single oxygen atom in perchlorate, and the distance between the pivot carbon atom and the NH₂ moiety in this group was 1.323(3) Å. The other nitrogen-containing group formed two H-bonds with the perchlorate ion. The distance between the pivot carbon and nitrogen atoms in this group was the distances between the pivot carbon atom and the NH₂ moiety are rather different - 1.323(3) Å for the C-NH₂ group bonded by H-bonds only to one oxygen atom of the perchlorate ion, and 1.297(4) Å for the C-NH₂ group bonded by two H-bonds to the perchlorate ion. The molecular structure of the oxalate is made up of two mutually similar acetamidinium units and one oxalate ion. All these ions in both compounds are interconnected by extensive hydrogen bonding systems. In the oxalate (7), eight- and fourteen-membered rings are formed (Figure 7). The twenty-two membered rings are the main element of the perchlorate (8) structure (Figure 8).

In the oxalate structure, the distances between difference between the respective pivot carbon and nitrogen atoms were groups are even greater at 1.339(5) Å and 1.280(5) Å. Our observations were not consistent, which disagree with a delocalization, and they differed from values concept and the data found in the literature, which fall between (1.302 and 1.312 Å). In these groups/reports, the H-bonds to the oxalate moiety are equidistant.

In light of the results of the XRD analysis, the molecular structure of the oxalate is made up of two mutually similar acetamidinium units and one oxalate ion. All these ions in both compounds are interconnected by extensive hydrogen bonding systems. In the oxalate (7), eight- and fourteen-membered rings are formed (Figure 7). The twenty-two membered rings are the main element of the perchlorate (8) structure (Figure 8).

From a study of the above mentioned data and motifs, in combination with the hygroscopicity data presented in Table 1, on the hygroscopicities of the compounds, it is clearly indicate that the 2D layered structures compounds containing with layered 2D (counterions linked by H bridges) structures counterions linked by H-bridges - 2, 4, 6, 7, and 11 - were not hygroscopic (2, 4, 6, 7, and 11; for 7, see Fig. ure 9) are not hygroscopic. illustrates the of 7. On the other hand, the compounds that displayed 3D structures - (1, 3, 5, and 8 - were more hygroscopic. For 8, see Fig. ure 10 shows the [] for 8.) have rather high hygroscopicities. This was probably due to

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Please review each statement and the revised statements for accuracy.

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caused by an easier incorporation of water molecules into the larger cavities of the compounds with 3D structures, compared to the intercalation of water into the compounds with 2D structures.

NMR spectroscopy

The NMR data for acetamidinium salts **2**, **4**, **7**, and **8** obtained with deuterated water (D_2O) are summarized in Table 2. A closer inspection of the proton NMR spectra measured in D_2O revealed that there is an equilibrium between deuterated and non-deuterated molecules, which were identified by marked (strongly decrease in the signal intensities of acidic protons intensity of the signals of the acidic protons). The equilibrium is shifted almost entirely to the side of the deuterated forms, indicating that sample (approximately 98% of the molecules were deuterated). These observations contradicted findings published by Kopylovich,¹⁴ wherein no deuteration was described, and two signals per 2H were observed.

On the other hand, the δ direction of the equilibrium was reversed in mixtures containing deuterated dimethyl sulfoxide- d_6 ($DMSO-d_6$), in which approximately 90% of the compounds were in non-deuterated form can be found for all of the samples measured analyzed. With the exception of acetamidinium formate (**4**)

in all cases (excluding **4** in $DMSO-d_6$), two distinct separated broadened signals/peaks belonging to the 2 x NHaHb arrangement were observed, which were probably due attributable to the delocalization of the positive charge throughout the amidinium group. The only exception is acetamidinium formate **4** in $DMSO-d_6$ where produced one single broad signal peak, which represented comprising all four NH protons bound to nitrogen was detected. The interactions within several acetamidinium complexes were studied by Tominey and Krechl using NMR, XRD analysis, and quantum chemical treatment.^{17,27} Our NMR results were in accordance with the observations published by Krechl¹⁷ and similar to the results obtained by Tominey²⁷ for acetamidinium tetrazolate complexes. These observations may have been due to this may be caused by differences in the interactions between the formate anion and amidinium groups in different solvents. The interactions inside some acetamidinium complexes were studied by Tominey and Krechl by means of NMR, X-ray analysis and quantum chemical treatment.^{17,27}

Differential thermal analysis

Acetamidinium nitrate (**2**), acetamidinium dinitromethane (**6**), and acetamidinium perchlorate (**8**) are energetic materials. They have potential for use in pyrotechnic applications, where they may replace guanidinium nitrate or perchlorate salts (nitrate or perchlorate). The acetamidinium salts

difference is the have a higher carbon contents than acetamidinium salts compared to their analogous guanidinium analogs, as ones (replacement of the amino group in guanidines is replaced by a methyl group) in acetamidine. Nevertheless, acetamidinium salts still have a relatively high nitrogen content. Compounds **6** and **8** have exhibited acceptable decomposition temperatures as determined by differential thermal analysis (measured by differential thermal analysis (DTA)). The DTA thermograms from the compounds are shown in Fig. 11. Both the nitrate (**2**) and the perchlorate (**8**) decomposed upon melting. Thus, the decomposition temperatures of **2** and **8** were considered as being 183 °C and 248 °C, respectively. (Figure 11). The maxima of the decomposition ranges for **2** and **8** were 255 °C and 390 °C, respectively. For comparison, decomposition of guanidinium nitrate on the same thermal stability device started to decompose at 270 °C, and guanidinium perchlorate started to decompose at 350 °C using the same thermal stability device.

Conclusions

Acetamidinium salts were synthesized and characterized by elemental analysis, electrospray mass spectrometry, and NMR. The and, in the case of energetic salts were also examined by DTA. The structures of several previously unknown acetamidines have been identified proved by X-ray diffraction by XRD analysis. Hygroscopicities of eight acetamidinium salts were determined at 90% humidity in 90% humidity of eight acetamidinium salts have been evaluated. The results of the different values of hygroscopicity analysis were corroborated by the structural determinations performed by XRD-ray analysis. The acetamidinium salts with 2D layered structures were not hygroscopic, while the acetamidinium salts with 3D layered structures were highly quite hygroscopic.

Conflicts of interest

In accordance with our policy on Conflicts of interest please ensure that a conflicts of interest statement is included in your manuscript here. Please note that this statement is required for all submitted manuscripts. If no conflicts exist, please state that "There are no conflicts to declare".

Acknowledgements

The acknowledgements come at the end of an article after the conclusions and before the notes and references.

Notes and references

† Footnotes relating to the main text should appear here. These might include comments relevant to but not central to the matter under discussion, limited experimental and spectral data, and crystallographic data.

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"This is for interpretation of the key results and to highlight the novelty and significance of the work. The conclusions should not summarize information already present in the article or abstract. Plans for relevant future work can also be included."

I suggest replacing the summary of the analysis with a broader statement about the novelty of your findings and their importance to the field.

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