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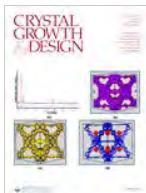
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Virtual Issues



Mikhail Antipin Memorial Issue

Vol. vi, Iss. 14

May 29, 2015

[Table of Contents](#)

[Cover Details](#)

This virtual special issue, which includes 30 research articles, honors the life and work of Dr. Mikhail Antipin, who led the X-ray Structural Center at the Institute of Organoelement Compounds, Russian Academy of Science, in Moscow, Russia, since 1995. Mikhail Antipin also co-supervised a research team in New Mexico for fifteen years, where he participated in projects funded by NASA, AFOSR, NIH, and NSF. He published more than 1000 scientific papers and supervised 38 Ph.D. and Dr. Sci. projects during his distinguished career in science. His broad research interests included air-sensitive organometallics, structural studies of liquids, phosphororganics, mesogene crystals, pharmaceutical co-crystals, high-precision studies of electron density distributions, and thermal motion of atoms in crystals.



Anion-Controlled New Inorganic Materials

Vol. vi, Iss. 13

December 31, 2014

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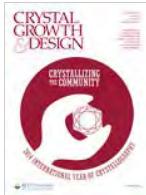
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This virtual special issue contains selected papers with focus on recent developments covering synthesis and various potential applications of anion-controlled new inorganic materials. The virtual special issue covers selected contributions made at the 1st International Symposium on "Advances and enhanced functionalities of Anion-controlled New Inorganic Materials (ANIM1)", E-MRS Spring Meeting 2013 Symposium T, which was held in Strasbourg, France, May 27-30, 2013.

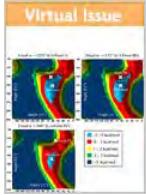


International Year of Crystallography 2014 (IYCr2014)

Vol. vi, Iss. 12
 December 31, 2014

[Table of Contents](#)
[Cover Details](#)

The United Nations declared 2014 to be the International Year of Crystallography (IYCr2014) to commemorate the centennial of X-ray crystallography since the first X-ray diffraction experiment was performed on a crystal of copper sulfate pentahydrate. This *Crystal Growth & Design* virtual special issue is dedicated to celebrate IYCr2014. It is a compilation of selected papers that provides a glimpse of various research activities in the fast growing field of crystal engineering.



10th International Workshop on the Crystal Growth of Organic Materials (CGOM10)

Vol. vi, Iss. 11
 February 14, 2014

[Table of Contents](#)
[Cover Details](#)

This virtual issue features selected papers from the 10th International Workshop on the Crystal Growth of Organic Materials (CGOM10) which was held at the University of Limerick in the Republic of Ireland from June 11–14, 2012.



14th International Conference on the Crystallization of Biological Macromolecules (ICCBM14)

Vol. vi, Iss. 10
 January 31, 2014

[Table of Contents](#)
[Cover Details](#)

This virtual special issue includes 12 peer-reviewed papers from the 14th International Conference on the Crystallization of Biological Macromolecules (ICCBM14) meeting which was held in Huntsville, Alabama, United States, from September 23–28, 2012.



In Honor of Prof. Gautam R. Desiraju

Vol. vi, Iss. 9
 October 4, 2013

[Table of Contents](#)
[Cover Details](#)

This virtual special issue honors the work of Gautam R. Desiraju on the occasion of his sixtieth birthday.

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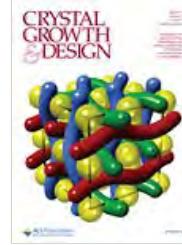
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VIRTUAL SPECIAL ISSUE
Anion-Controlled New Inorganic Materials

Guest Editors: Ivoil P. Koutsaroff and Amparo Fuertes

1 2 3 4 5 6 7 8 9 10


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Design of Pyroelectric Mixed Crystals Having a Varying Degree of Polarity: The L-Asparagine-H₂O/L-Aspartic Acid System

A. Belitzky, I. Weissbuch, Y. Posner-Diskin, M. Lahav, and I. Lubomirsky

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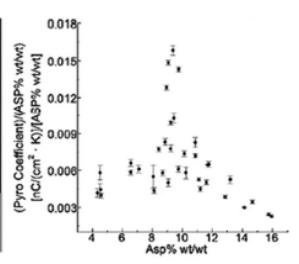
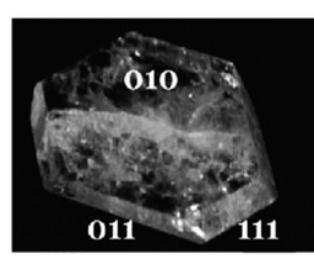
Publication Date (Web): April 13, 2015 ([Article](#))DOI: [10.1021/acs.cgd.5b00230](https://doi.org/10.1021/acs.cgd.5b00230)[» Abstract | Supporting Info](#) [ACS ActiveView PDF](#)
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Anion-Controlled New Inorganic Materials

Volume vi, Issue 13

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This virtual special issue on Anion-Controlled New Inorganic Materials (ANIM) includes selected papers from the Symposium T of E-MRS Spring 2013 entitled [Advances and Enhanced Functionalities of Anion-Controlled New Inorganic Materials](#) (ANIM1), which was held in Strasbourg, France, from May 27–30, 2013 and was organized by Ivoil P. Koutsaroff (Murata Manufacturing Co., Ltd., Japan), Stefan G. Ebbinghaus (Martin-Luther-Universität Halle-Wittenberg, Germany), Laurent Le Gendre (IETR – IUT de Saint Brieuc – Université de Rennes 1, France), and Amparo Fuertes (Institute of Materials Science of Barcelona (CSIC), Spain).

[Go to List of Articles](#)

In the widely investigated metal oxides, the modification of properties by changing the crystal structure and the cation composition is the usual way to explore materials. However, the possibility to tune the physical or chemical properties by the substitution of anions, i.e., the formation of a variety of mixed anion materials and compounds, expands the opportunities to find or improve their applications. In the last decade, oxynitrides have been considerably investigated leading to important discoveries, notably, new dielectric (paraelectric and potentially ferroelectric) and colossal magnetoresistive perovskites; superconducting, semiconducting, and ferromagnetic-like behaviors found in mixed anion layered chalcogenides; and new visible active photocatalysts or red emitting phosphor materials. Oxyfluorides have been developed as non-centrosymmetric optical materials, and oxyhydrides and oxychalcogenides with new electronic properties have been reported. The present symposium aimed at creating an international forum for the different types of researchers in the field of anion-modified inorganic materials (physicists – experimentalists and theoreticians, materials scientists, and solid state chemists).

The symposium facilitated interdisciplinary discussions between these differing "core groups", allowing researchers from across all relevant areas to consolidate their scope of research, thereby stimulating collaboration and thus strengthening the European and world-wide activities in this expanding field. It merged researchers from 15 different countries in the fields of mixed anion materials who presented 12 invited talks, 25 oral contributions, and 24 posters.

The present ANIM issue contains 17 articles, a few of which were published outside of the ANIM virtual special issue in *Crystal Growth & Design* or in *Chemistry of Materials* within the ACS. A related article¹ was published by Royal Society of Chemistry and two others^{2,3} were published by Elsevier.

A few papers cover in depth the structural properties of alkaline earth tantalum and niobium oxynitride perovskite-type systems as well as rare earth tantalum and niobium oxynitrides. Partial order of oxide and nitride anions consistent with the formation of planes of disordered cis-anion chains is observed in oxynitride perovskites SrTaO₂N and LaTaON₂ materials. No loss of anion order is observed up to 1100 °C, demonstrating the thermal stability of anion order, and it is evident that the anions are segregated when the materials are synthesized.⁴

The crystal structures of RTa₂O where R = Ce, Pr are reported for the first time. CeTa₂O and PrTa₂O have a⁻b⁺a⁻ octahedral tilting and exhibit no long-range ordering of anions, a combination that leads to *Pnma* symmetry. Density functional theory (DFT) calculations show that ordered structures where the [TaN_{4/2}O_{2/2}]³⁻ octahedra adopt a cis configuration are considerably more stable than those where the octahedra adopt a trans configuration. However, alternate patterns of long-range ordering that retain the local cis configuration have very similar enthalpies. The fact that there are multiple cis-ordered configurations with similar enthalpies helps to explain why the strong preference for a local cis-configuration does not lead to long-range anion ordering.

EuWON₂ and NdVO₂N are among other oxynitride perovskite systems that have been studied recently. An important aspect of the crystal structures of AMX₃ perovskites is their tendency to form superstructures of long-range ordered tilts or rotations of the MX₆

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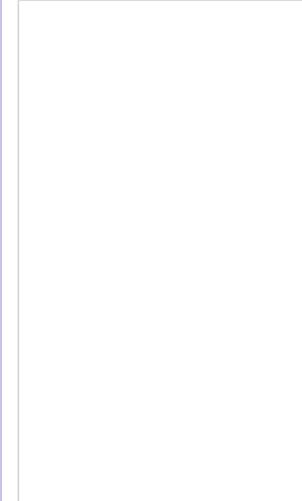
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octahedra. The anion order model based on the degree of O/N order in solid oxynitrides based on both ionic and covalent approaches also accounts for observations that ATaO_2N ($\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$) oxynitrides have high dielectric constants but do not show ferroelectric ordering transitions, unlike pure oxide analogues such as BaTiO_3 .

However, the most recent experimental studies demonstrated a room-temperature ferroelectricity in compressively strained SrTaO_2N thin films epitaxially grown on SrTiO_3 substrates. Local piezoresponse measurements revealed the existence of small classical ferroelectric domains with dimensions of 10–100 nm, likely stabilized by compressive epitaxial strain, surrounded by a matrix with a relaxor ferroelectric-like behavior, characterized by the absence of spontaneous polarization. The ferroelectric domains and the relaxor-like matrix were identified as trans- and cis-type phases, respectively, based on results of first-principles DFT calculations that mapped the total energy of SrTaO_2N for various lattice parameters. The effective dielectric constant of epitaxial SrTaO_2N thin films with a thickness of 290 nm was in the range of 2200 at room temperature with a dielectric loss of 0.04 (1–10 KHz).⁵ In contrast, the dielectric constant of epitaxial CaTaO_2N thin film was approximately 12 with $\tan \delta < 0.01$. This value is comparable to that of bulk CaTaO_2N ($\epsilon' \approx 30$), suggesting that the small ϵ' is intrinsic to CaTaO_2N . On other hand, polycrystalline $(\text{Ba}, \text{Sr})\text{TiO}_{3-x}\text{N}_y$ thin films could have dielectric constants in the range of 1100 with a dielectric loss less than 0.01 (1 KHz) for 100–180 nm thick films. In contrast, the dielectric constant of LaTiO_2N thin films were somewhere between SrTiO_3 and $(\text{Ba}, \text{Sr})\text{TiO}_{3-x}\text{N}_y$ but with higher dielectric losses.

It had been observed by high angle angular dark field-scanning transmission electron microscopy (HAADF-STEM) electron energy-loss spectroscopy (EELS) methods that $(\text{Ba}, \text{Sr})\text{TiO}_{3-x}\text{N}_y$ oxynitride grown films exhibit the presence of close to anion stoichiometric (ABO_2N_1) 10 nm-sized clusters within the oxide perovskite matrix, and such N-containing clusters were dispersed somehow homogeneously across the grain boundaries and are present within some of the grains of the oxynitride perovskite films which remain in a superparaelectric (nonpolar) state. Both LaTiO_2N and $(\text{Ba}, \text{Sr})\text{TiO}_{3-x}\text{N}_y$ thin films have very significant dielectric polarizabilities (nonlinearities) or DC bias voltage tunabilities.

Other studies have demonstrated that even the cationic co-substitutions can cause anion-driven properties control of the ferroelectric behaviors encountered in $(\text{Ba}, \text{Sr})_2\text{Ln}(\text{Fe}, \text{Nb}, \text{Ta})_5\text{O}_{15}$ tetragonal tungsten bronze (TTB). To this end, the traditional and empirical methods of solid-state chemistry are applied, especially to survey the effect of various substitution schemes on the properties of the $\text{Ba}_2\text{NdFeNb}_4\text{O}_{15}$ ferroelectric TTB. The anionic framework then emerges as the driving force behind these original behaviors, while consideration of additional experimental results suggests that incommensurate modulations are likely involved also. The other report covers the growth of a series of sub-centimetric, lead-free, relaxor $\text{Ba}_2\text{NdFeNb}_4\text{O}_{15}$ ($\text{Ln} = \text{Pr}, \text{Nd}, \text{Sm}, \text{Eu}$) single crystals with the TTB structure. Following thorough chemical and structural characterizations, their crystal chemistry is investigated. The square channel encompassing the lanthanide ions is found to play a central role in the crystal chemistry of these single crystals. This square channel is also strongly connected to the dielectric properties of these single crystals, and comparison with corresponding ceramics highlights the influence of Ln^{3+} vacancies in the emergence of relaxor or ferroelectric properties.

It had been observed through via combined Rietveld refinements of synchrotron and neutron powder diffraction analysis that the presence of long-range orientational ordering of the polar $\text{MoO}_3\text{F}_3^{3-}$ units destroys the a-glide plane of the nonpolar of $\alpha\text{-M}_3\text{MoO}_3\text{F}_3$ ($\text{M} = \text{K}, \text{Rb}$) structure lowering the symmetry to the polar space group I41. It had been proposed that in $\text{A}_2\text{BB}'\text{X}_6$ compounds a tolerance factor below unity, a relatively large difference in the ionic radii size of the B and B' cations, and a small difference in the ionic radii of the A and B cations favor the occurrence of noncooperative octahedral tilting even though their structure is related to the cubic double perovskite structure.

Single crystal growth and structural characterization of $(\text{NH}_4)_2\text{WO}_2\text{F}_4$ and $\text{Rb}_2\text{KMnO}_3\text{F}_3$ oxyfluoride materials are also covered in some of the reports. Crystal structure, superconducting, semiconducting, and ferromagnetic-like properties of $\text{Ce}(\text{O}, \text{F})\text{BiS}_2$ single crystals have been also reported.

New complex perovskites $\text{AM}_{0.2}\text{Ta}_{0.8}\text{O}_{2.8}\text{N}_{0.2}$ ($\text{A} = \text{Sr}, \text{Ba}; \text{M} = \text{Li}, \text{Na}$) have been reported being prepared by an intercalation-type synthetic approach using the layered perovskite $\text{A}_5\text{Ta}_4\text{O}_{15}$, as exemplified by the conversion from $\text{Sr}_5\text{Ta}_4\text{O}_{15}$, to $\text{SrLi}_{0.2}\text{Ta}_{0.8}\text{O}_{2.8}\text{N}_{0.2}$, while the charge compensation was maintained through the cooperative Li^+ insertion and aliovalent $\text{O}^{2-}/\text{N}^{3-}$ substitution, thereby offering a unique route to alkali-containing oxynitride compounds.

We thank the conference chairpersons of EMRS-2013, Alain Claverie, Arnulf Jaeger-Walda, Francesco Priolo, and Anke Weidenkaff, the remaining organizers of the symposium Stefan G. Ebbinghaus and Laurent Le Gendre, and the sponsors IETR, IUT Saint-Brieuc, and Murata Manufacturing Co., Ltd. who made this 1st ANIM Symposium possible. We acknowledge the invited speakers, the authors of the papers, the referees and editors of *Crystal Growth & Design*, and the publishers of this ANIM issue at the American Chemical Society. We also express our appreciation to Mihaela Rogers for the editorial preparation of this special issue.

✉ cm'D"? ci lgUfcZ Guest Editor
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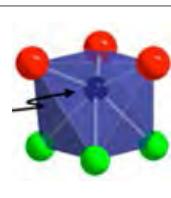


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Cryst. Growth Des., 13, 4623–4629

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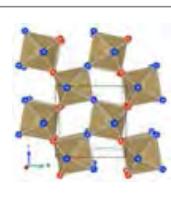


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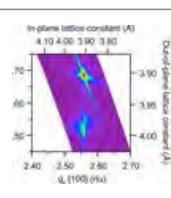


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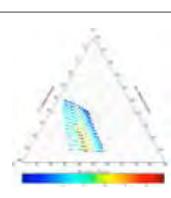


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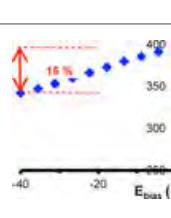


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Cryst. Growth Des., 14, 523–532

DOI: 10.1021/cg401259r



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Cryst. Growth Des., 13, 4852–4858

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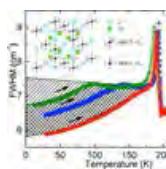
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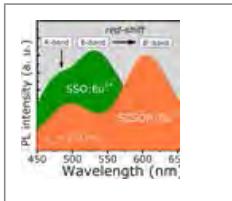
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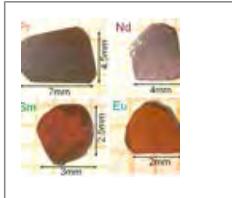
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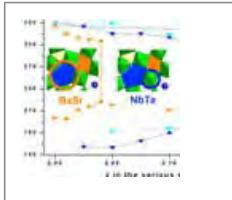
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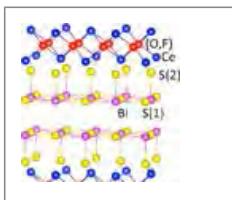
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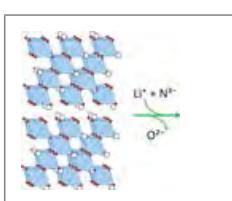
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Cryst. Growth Des., 14, 500–512
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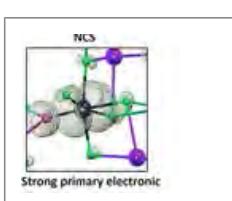
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Michaël Josse, Pierre Heijboer, Marjorie Albino, Flora Molinari, Florence Porcher, Rodolphe Decourt, Dominique Michau, Eric Lebraud, Philippe Veber, Matias Velazquez, and Mario Maglione
Cryst. Growth Des., 14, 5428–5435
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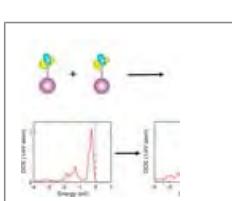
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Akira Miura, Masanori Nagao, Takahiro Takei, Satoshi Watauchi, Yoshikazu Mizuguchi, Yoshihiko Takano, Isao Tanaka, and Nobuhiro Kumada
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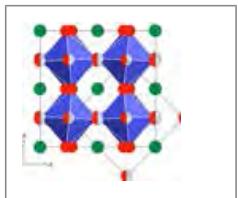
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Young-Il Kim, Younkee Paik, and Maxim Avdeev
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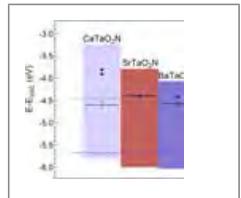
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5bUngjg'cZh YgYa TcWU'gHfYg]b'NbC!bB'Wta dci bXg
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H Yfa U`niFcVi gli5 b]cb!7\ Ujb`CfXYf`jb`Cl nb]f]XY'DYfcj g_]hrg
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9`YWfc]WGfi Wi fY`cZHUbHJi a `Cl nb]f]XY'DYfcj g_]hrg D\ c]cWUHng]g
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