

Main-group-based rings and polymers

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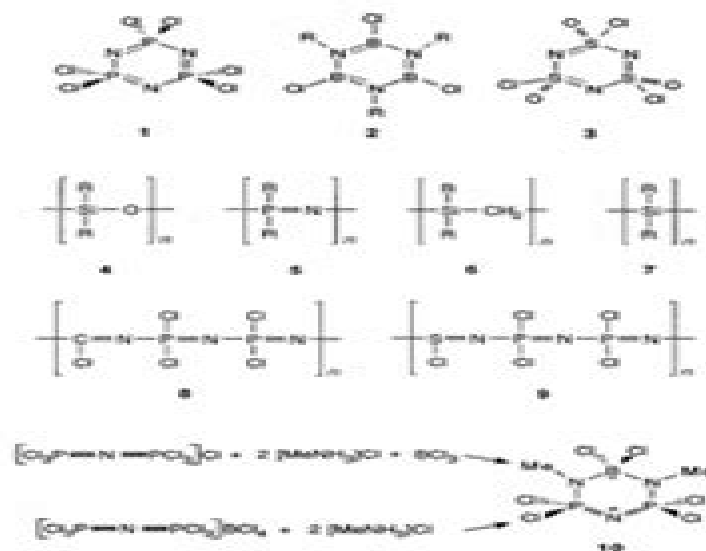
The synthesis and study of heterocycles containing main-group elements have played a pivotal role in the development of inorganic chemistry. Well known 'text-book' classes of inorganic rings include the cyclic phosphazene **1**, which was first prepared by Liebig and Rose¹ in 1834, and borazines such as **2**, the initial example of which was first synthesized by Stock and Pohland² in 1926. A further example is provided by sulfamides **3** which have been known since the 1950s.³ It is noteworthy that the ring skeletons in phosphazenes **1** and borazines **2** have been shown to be very robust and to permit facile halogen atom replacement reactions.⁴ This chemistry has been well studied and has provided much useful information on nucleophilic substitution reactions in inorganic chemistry. The bonding used to describe structures such as **1–3** and the possible application of the label 'inorganic benzene' has been the subject of much debate.

Current research continues to uncover fascinating new ring systems which pose intriguing questions with respect to their bonding or which exhibit unexpected reactivity. Some noteworthy examples include ten-electron gallium-based systems and pseudo aromatic cyclic silylenes,^{5,6} novel cyclic tellurium imides,⁷ and interesting aluminum–nitrogen heterocycles⁸ among others.⁹ In addition to these important fundamental questions, a further reason for studying inorganic rings is their potential use as precursors to polymers and solid-state materials.

The synthesis of long chains of atoms of inorganic elements, inorganic polymers, provides a substantial synthetic challenge but is motivated by the possibility of accessing new materials with interesting and useful properties.¹⁰ The most well known and widely used current routes to high molecular weight polysiloxanes **4**,¹¹ polyphosphazenes **5**,¹² and poly(carbosilanes) **6**,¹³ involve a ring-opening polymerization (ROP) reaction. With this in mind, interest in the polymerization behaviour of other inorganic rings has recently gathered momentum. Polysilanes **7**,¹⁴ poly(carbophosphazenes) **8**,¹⁵ and poly(thiophosphazenes) **9**¹⁶ represent examples of well characterized polymers which have been successfully prepared by ROP in the past decade.

The use of inorganic rings to construct solid-state materials with novel properties has also been successfully developed. For example, materials with interesting electronic, magnetic and conductive properties have been prepared from sulfur–nitrogen or selenium–nitrogen heterocycles.^{17,18} In addition, inorganic rings have attracted considerable attention as precursors to ceramics via thermolysis; examples include the use of aluminum–nitrogen or gallium–arsenic heterocycles to prepare AlN and GaAs, respectively.^{19,20}

In this perspective we describe two areas of research on main group heterocycles which have been pursued in our research group. These involve, first, boron–nitrogen–phosphorus heterocycles and secondly, rings of sulfur(vi), nitrogen and phosphorus atoms. Our focus has been on the discovery of new structures and reactivity and, in particular, on the potential use of these species as precursors to new polymers via ROP reactions.



Boron–Nitrogen–Phosphorus Rings

Early work on B–N–P heterocycles

Cyclic species containing boron, nitrogen, and phosphorus atoms were first described about 30 years ago. The first report of a BP₂N₂ ring was published by Sherif and Schmalbach²¹ in 1966 and described the use of a [5 + 1] cyclocondensation reaction between [H₂NPh₂P=N–PPh₂NH₂]Cl and RBCl₂ (R = Ph or Cl) to yield boron–nitrogen–phosphorus heterocycles. However, by present day standards, the characterization was poor.²² Nevertheless, applications of these rings as pesticides were claimed in a patent.²³

The perchlorinated boron–nitrogen–phosphorus heterocycle **10** was subsequently synthesized by two very similar routes. In 1968 Becke–Goehring and Müller²⁴ reported the reaction of [Cl₂P=N–PCl₂]Cl with BCl₃ and [MeNH₂]Cl in 1,2-dichloroethane and gave a 77% yield of **10**. In 1971 a slight modification of the Becke–Goehring synthesis was reported by Binder.²⁵ This route involved the reaction of [Cl₂P=N–PCl₂]BCl₃ with [MeNH₂]Cl and gave improved yields of 84–90%. Convincing structural characterization of this white crystalline compound was provided by NMR spectroscopy. For example, ¹¹B NMR showed a

Rings Clusters And Polymers Of Main Group And Transition Elements

Sabine Zange



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Table of Contents Rings Clusters And Polymers Of Main Group And Transition Elements

1. Understanding the eBook Rings Clusters And Polymers Of Main Group And Transition Elements
 - The Rise of Digital Reading Rings Clusters And Polymers Of Main Group And Transition Elements
 - Advantages of eBooks Over Traditional Books
2. Identifying Rings Clusters And Polymers Of Main Group And Transition Elements
 - Exploring Different Genres
 - Considering Fiction vs. Non-Fiction
 - Determining Your Reading Goals
3. Choosing the Right eBook Platform
 - Popular eBook Platforms
 - Features to Look for in an Rings Clusters And Polymers Of Main Group And Transition Elements
 - User-Friendly Interface
4. Exploring eBook Recommendations from Rings Clusters And Polymers Of Main Group And Transition Elements
 - Personalized Recommendations

- Rings Clusters And Polymers Of Main Group And Transition Elements User Reviews and Ratings
- Rings Clusters And Polymers Of Main Group And Transition Elements and Bestseller Lists
- 5. Accessing Rings Clusters And Polymers Of Main Group And Transition Elements Free and Paid eBooks
 - Rings Clusters And Polymers Of Main Group And Transition Elements Public Domain eBooks
 - Rings Clusters And Polymers Of Main Group And Transition Elements eBook Subscription Services
 - Rings Clusters And Polymers Of Main Group And Transition Elements Budget-Friendly Options
- 6. Navigating Rings Clusters And Polymers Of Main Group And Transition Elements eBook Formats
 - ePub, PDF, MOBI, and More
 - Rings Clusters And Polymers Of Main Group And Transition Elements Compatibility with Devices
 - Rings Clusters And Polymers Of Main Group And Transition Elements Enhanced eBook Features
- 7. Enhancing Your Reading Experience
 - Adjustable Fonts and Text Sizes of Rings Clusters And Polymers Of Main Group And Transition Elements
 - Highlighting and Note-Taking Rings Clusters And Polymers Of Main Group And Transition Elements
 - Interactive Elements Rings Clusters And Polymers Of Main Group And Transition Elements
- 8. Staying Engaged with Rings Clusters And Polymers Of Main Group And Transition Elements
 - Joining Online Reading Communities
 - Participating in Virtual Book Clubs
 - Following Authors and Publishers Rings Clusters And Polymers Of Main Group And Transition Elements
- 9. Balancing eBooks and Physical Books Rings Clusters And Polymers Of Main Group And Transition Elements
 - Benefits of a Digital Library
 - Creating a Diverse Reading Collection Rings Clusters And Polymers Of Main Group And Transition Elements
- 10. Overcoming Reading Challenges
 - Dealing with Digital Eye Strain
 - Minimizing Distractions
 - Managing Screen Time
- 11. Cultivating a Reading Routine Rings Clusters And Polymers Of Main Group And Transition Elements
 - Setting Reading Goals Rings Clusters And Polymers Of Main Group And Transition Elements
 - Carving Out Dedicated Reading Time
- 12. Sourcing Reliable Information of Rings Clusters And Polymers Of Main Group And Transition Elements
 - Fact-Checking eBook Content of Rings Clusters And Polymers Of Main Group And Transition Elements

- Distinguishing Credible Sources
- 13. Promoting Lifelong Learning
 - Utilizing eBooks for Skill Development
 - Exploring Educational eBooks
- 14. Embracing eBook Trends
 - Integration of Multimedia Elements
 - Interactive and Gamified eBooks

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