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# POLYMERIC IONIC LIQUIDS AS AN OBJECT OF MODERN CHEMISTRY AND CHEMICAL TECHNOLOGY: A MINI-REVIEW OF ARCHITECTURE, STRUCTURE–PROPERTY RELATIONSHIPS, AND APPLICATION AREAS

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Polymeric ionic liquids have emerged as a distinct class of functional macromolecular materials at the interface of polymer chemistry, physical chemistry, and materials science. Unlike low-molecular-weight ionic liquids and conventional polyelectrolytes, these systems combine a high density of ionic centers with a tunable macromolecular architecture, which results in specific supramolecular organization, relaxation dynamics, and segmentally controlled mechanisms of ionic transport. This paper summarizes an architecture-oriented approach to describing polymeric ionic liquids and proposes a classification logic based on how charged fragments are integrated into the polymer chain: side-chain ionic polymers, ionene-type polymers, and crosslinked/network structures. Key structure–property dependences are analyzed, including the effects of ion-pair chemistry, glass transition temperature, molecular weight, and domain organization on ionic conductivity and the stability of operational characteristics. It is shown that insufficient standardization of macromolecular parameters and limited experimental verification of domain structures reduce reproducibility and comparability of results and hinder the development of generalized «structure–property» models. The prospects for a transition to rational design are substantiated through controlled synthesis of ionene architectures, systematic correlation of «structure–glass transition temperature/rheology–conductivity» parameters, and the creation of hybrid materials of the polymeric ionic liquid–ionic liquid and polymeric ionic liquid–nanofiller types. Application areas in electrochemical energy technologies, membrane processes, electrocatalysis, and sensing are summarized, and the need to integrate sustainability criteria, particularly life-cycle assessment, long-term stability, and recyclability potential, is emphasized.

**Keywords***:* polymeric ionic liquids, ionene-type polymers, macromolecular architecture, structure–property relationships, ionic transport, domain organization, membrane materials, electrochemical energy technologies.

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## *Introduction*

Ionic liquids (ILs) and IL-based materials occupy an important place in modern chemistry and chemical technology due to their combination of low volatility, high thermal and electrochemical stability, and broad opportunities for structural design [1,2]. Initially, ILs were positioned as promising «green» solvents in line with the principles of green chemistry formulated by Anastas and Warner [3]. However, subsequent studies have demonstrated that the environmental viability of ionic systems cannot be assessed solely on the basis of vapor pressure or thermal stability; a comprehensive analysis of toxicity, synthesis energy intensity, and material behavior throughout the life cycle is required [4–6].

A logical stage in the development of ionic liquid chemistry has been the formation of the field of polymeric ionic materials, in particular polymeric ionic liquids (PILs). The transition from low-molecular-weight systems to a macromolecular state improves the stability of functional media, reduces migration of ionic components, and expands the range of application-relevant properties [7,8]. At the same time, polymerization of ionic fragments causes qualitative changes in interionic interactions, relaxation processes, and phase behavior, making it necessary to consider PILs from the standpoint of polymer science rather than merely as modified ionic media. Therefore, recent review papers indicate a shift from treating PILs as “solid analogues” of ionic liquids toward analyzing them as full-fledged macromolecular systems where chain architecture, ionic group density, and crosslinking type are decisive [5,7–9].

### *Polymeric ionic liquids as a class of macromolecular compounds*

Historically, the first polymeric ionic systems were developed within polyelectrolyte chemistry, where charged groups are predominantly localized in the side chains of macromolecules and are compensated by mobile counterions [10–13]. Such systems played an important role in shaping concepts of electrostatic interactions in polymer media; however, their physicochemical nature differs substantially from that of PILs.

PILs are characterized by a high density of ionic centers combined with macromolecular architecture and a reduced role of an external solvent. As a result, the ionic component ceases to be auxiliary and becomes a structure-forming factor that governs the glass transition temperature, segmental mobility, rheological behavior, and transport properties [11,14–17]. The generalization of current data allows PILs to be viewed as an evolutionary outcome of two fields–ionic liquid chemistry and functional polymer chemistry. In early works, ILs were considered primarily as media alternative to molecular solvents with unique electrochemical characteristics [16–18]; however, the further development of the «designer solvents» concept has led to the understanding that combining ionic nature with macromolecular architecture provides fundamentally new possibilities for property control in materials [9,19].

Zhu and Yang [9] showed that polymerization of ionic fragments is not simply a way to immobilize an ionic liquid in the solid state; it also induces a reorganization of interionic interactions and relaxation dynamics. Unlike low-molecular-weight ILs, where transport is governed by the collective dynamics of ionic clusters, in PILs segmental motion of the polymer backbone plays a key role; this brings PILs closer to conventional polymer electrolytes but distinguishes them by a higher density of ionic centers and a weaker dependence on an external solvent [9,20].

For many PILs, nanoscale phase segregation into polar and nonpolar domains has been established [19,21–23]. The spatial characteristics of these domains depend on macromolecular structure, the nature of ionic groups, and synthesis conditions, and they significantly affect thermodynamic and kinetic properties of the materials.

### *Classification of polymeric ionic liquids*

There is no unified approach to PIL classification in the modern literature, which is largely due to the interdisciplinary nature of these materials. They are grouped by the type of ionic groups, the nature of counterions, the synthesis route, or application area [7,8]. However, from the standpoint of polymer chemistry, the decisive criterion is macromolecular architecture and the way ionic fragments are incorporated into the polymer chain. Even for identical ion pairs, materials may exhibit markedly different electrochemical characteristics depending on chain structure [24,25]. Therefore, an architecture-based approach is appropriate: side-chain ionic polymers, ionene-type polymers, and crosslinked/network structures [5,9].

#### *Side-chain polymeric ionic liquids*

The most common class comprises PILs with ionic groups in side chains, obtained by polymerization of ionic monomers or by post-synthetic modification of neutral polymers [14,15,23]. Such systems feature a high local concentration of charged fragments, which promotes formation of stable ionic associates and increases Tg. This often creates a trade-off between ionic conductivity and mechanical stability [24–26].

Analysis of application-driven studies indicates that in many works the polymer matrix is treated mainly as a mechanical host for an ionic phase, whereas fundamental macromolecular parameters (molecular weight, dispersity, spacer length and flexibility) are frequently not normalized or not analyzed. This complicates comparison between studies and limits the establishment of universal structure–property relationships. Reviews emphasize the need to shift from empirical formulation screening to controlled synthesis with reproducible macromolecular characteristics [5,19].

#### *Crosslinked and network polymeric ionic systems*

Crosslinked and three-dimensional PILs exhibit high mechanical strength and thermal stability, making them promising for membrane technologies and catalysis [27–30]. However, a rigid network architecture restricts segmental mobility, so ionic transport is governed predominantly by counterion diffusion within local domains.

*Ionene-type polymeric ionic liquids*

A fundamentally different strategy is implemented in ionene-type PILs, in which charged fragments are incorporated into the main chain of the macromolecule [16,17,31]. This architecture provides a more uniform charge distribution, reduces the likelihood of forming rigid ionic domains, and creates prerequisites for maintaining segmental mobility even at high ionic group densities. Ionene systems are therefore regarded as a promising platform for reconciling glass transition temperature and ionic conductivity [24,25].

Current studies indicate that incorporating charged fragments into the backbone reduces the degree of microphase segregation and weakens the role of rigid ionic domains. As a result, ionic transport in such systems is more tightly coupled to segmental mobility than to local counterion diffusion, distinguishing them from conventional polyelectrolytes. At the same time, synthetic complexity and the limited number of scalable routes for preparing ionene-type PILs remain important barriers to broad implementation [5,9].

***Supramolecular organization and domain structure of PILs***

One of the key features of PILs is self-organization into nanoscale polar and nonpolar domains. For low-molecular-weight ILs, such nanostructuring is well known and is related to the amphiphilic nature of ions [20,32]. In PILs, domain organization is complicated by the presence of a macromolecular chain, which acts simultaneously as a structure-forming and constraining factor.

According to Silva et al. [20], domain organization is determined not only by the length of alkyl substituents but also by backbone rigidity and crosslinking density. In crosslinked PILs, polar domains tend to be localized and form isolated regions, restricting transport. In contrast, in linear ionene-type systems, domains may form percolation pathways that enable effective charge transport even at high Tg [5,9].

Importantly, in a considerable portion of application-focused studies, domain structures are not experimentally analyzed, and their role in property formation remains hypothetical. This constitutes a substantial methodological limitation, particularly for studies in electrochemical energy technologies and membrane processes [24,30]. Therefore, when interpreting transport properties of PILs, it is advisable to consider Tg, segmental dynamics, and domain organization simultaneously as interrelated parameters.

***Structure–property relationships***

Establishing structure–property relationships is a central task in polymer science. For PILs, this task is complicated by the combination of polymer factors (molecular weight, dispersity, chain architecture) with strong Coulombic interactions [33]. A generalization of current data shows that PILs do not exhibit a universal correlation between ionic conductivity and viscosity or ionic group concentration, typical of low-molecular-weight ILs. Instead, PIL conductivity is determined by a set of factors in which Tg, segmental mobility, and supramolecular domain organization play key roles [20,32,33].

Tg is among the most sensitive characteristics of PILs. With increasing ionic group concentration, Tg typically increases due to formation of ionic domains that act as physical crosslinks [14–17]. Introducing flexible aliphatic fragments or using less coordinating counterions can reduce Tg, but this is often accompanied by decreased mechanical or thermal stability [25].

Molecular weight and its distribution are fundamental parameters for polymers; however, for PILs these characteristics are often omitted, hindering comparison across studies [26]. Available data suggest that increasing molecular weight may reduce segmental mobility and, consequently, ionic conductivity, although strong ionic interactions can partly compensate for this effect. An additional challenge is the lack of standardized methods for determining molecular weight of PILs. As noted by Salas et al. [5], many publications describe PILs without reporting Mw or Mn, which is atypical for polymer chemistry and significantly limits reproducibility.

In studies focused on electrochemical applications, high ionic conductivity is often claimed as the key advantage of PILs. However, proper interpretation requires considering y in close connection with Tg and segmental dynamics. According to Bodin et al. [25], increased conductivity in some PILs is achieved at the expense of reduced mechanical stability or thermal robustness. Matuszek et al. [26] showed that in hybrid PIL/redox ionic liquid systems, transport can be mixed, combining a segmentally controlled mechanism with local redox processes. This broadens design opportunities but complicates interpretation unless macromolecular parameters are carefully accounted for.

***Application areas***

PILs are actively investigated as electrolytes for lithium-ion batteries, supercapacitors, and other electrochemical systems, where they offer improved safety and an expanded operating temperature range [24–26,34,35]. A substantial body of work addresses membrane technologies, particularly proton-exchange membranes for fuel cells, where PIL components enhance conductivity and chemical stability [27–30,36]. Individual studies also demonstrate the promise of PILs in electrocatalysis, sensing, and sorption processes [20,32,37–40].

At the same time, in most application-focused papers, PILs are treated as functional matrices or additives, whereas the influence of macromolecular architecture on transport mechanisms, degradation pathways, and property reproducibility is analyzed only fragmentarily. This limits rational materials design and underscores the need for a systematic approach that integrates ionic liquid chemistry with polymer science concepts [24–26,30,34].

### *Methodological limitations in contemporary PIL research*

Despite the rapid growth in publications, the current state of PIL research is characterized by several methodological limitations. One of the key issues is the fragmented approach to property analysis: emphasis is placed mainly on application-relevant characteristics (ionic conductivity, electrochemical window, thermal stability), while macromolecular parameters of the polymer component often remain outside systematic analysis [5,9].

In a significant number of works, data on molecular weight, dispersity, and chain architecture are missing, which impairs reproducibility and comparability [5]. Even with identical ion pairs, differences in macromolecular architecture may lead to different ionic transport mechanisms and relaxation behavior [24,25]. Another limitation is the predominance of empirical design, where optimization proceeds by compositional screening rather than by developing generalized «structure–property» models [24,30,34].

A separate issue is insufficient attention to supramolecular organization and domain structure. In many studies, domain formation is postulated by analogy with low-molecular-weight ILs without experimental verification of spatial characteristics or transport relevance [20,32]. This hampers correct interpretation and the development of physically grounded models.

Thus, further progress requires a transition from descriptive and empirical studies to a systematic methodology rooted in polymer science, with controlled macromolecular parameters and verified supramolecular organization [5,7–9].

### *Scientific novelty and distinctions of this review*

The scientific novelty of this review lies in a systematic consideration of PILs not as derivatives of low-molecular-weight ionic systems, but as a full-fledged class of macromolecular compounds with specific structure–property patterns. Unlike reviews primarily focused on applications or functional performance [19,24,26], this work emphasizes macromolecular architecture as the governing factor for ionic transport, relaxation phenomena, and supramolecular organization.

Structure–property relationships are summarized from a polymer-science perspective, and the lack of standardization of macromolecular parameters in application-driven studies is critically analyzed [5,9]. The potential of ionene-type PILs as a platform for coordinated control over segmental mobility and ionic transport – often insufficiently covered in existing reviews – is highlighted as well.

### *Unresolved scientific problems and development prospects for polymeric ionic liquids*

Despite significant progress in synthesis and applications, PIL research still faces several fundamental challenges. One of the most important is the lack of consistent physicochemical models that adequately describe the interplay between macromolecular architecture, supramolecular organization, and ionic transport mechanisms. Many existing models are borrowed from theories of low-molecular-weight ILs or classical polymer electrolytes and do not capture the specific combination of high ionic center density with macromolecular segmental dynamics [5,9,20].

A major unresolved issue is the role of domain organization. Although polar/nonpolar domains are reported in a number of works, their spatial characteristics and influence on transport have not been studied systematically. The absence of correlations between structural data (SAXS, WAXS, AFM) and macroscopic properties (conductivity, Tg, rheology) complicates the establishment of universal relationships [20,32].

Controlling molecular weight and dispersity remains another challenge. In many publications, PILs are described without key macromolecular parameters, preventing meaningful comparison and predictive modeling [5]. The development of controlled polymerization methods for ionic monomers and scalable routes to ionene-type polymers remains a priority [9].

Considerable difficulties are also associated with long-term stability in practical systems. Most studies report short-term electrochemical or thermal stability, while degradation processes related to macromolecular restructuring, phase segregation, or counterion migration are addressed only fragmentarily [24–26,30]. This is particularly critical for energy and membrane applications requiring stability over thousands of cycles [34].

Future progress is expected to rely on rational design based on clear structure–property principles. Ionene-type PILs are of particular interest as an architecture-controlled platform capable of tuning segmental mobility and ionic center density without inducing overly rigid domains [5,9]. Hybrid systems such as PIL–ionic liquid and PIL–nanofiller are also promising, where synergy between polymer architecture and ionic dynamics may be achieved [24,26].

An important direction is the integration of green chemistry principles and life-cycle assessment into PIL design. Environmental viability should be evaluated considering synthesis energy intensity, recycling possibilities, and long-term stability, not merely operational performance [5,7,8].

Nevertheless, results reported in a number of recent studies indicate that the field can move from empirical formulation screening to controlled property formation via a polymer-chemistry-driven approach to synthesis and analysis. In particular, ionene-type PIL studies show that synthesis parameters (temperature, time, solvent composition and polarity, concentration regimes) can serve as tools for controlling molecular weight and structural state [41,42]. This logic is crucial for overcoming reproducibility issues and enabling correct interlaboratory comparisons.

In [41], it was shown that premature precipitation, collapsed chain conformations, and reduced end-group reactivity can arise predictably from an unfavorable choice of polymerization conditions. Optimizing temperature–concentration regimes and selecting solvents that stabilize a homogeneous reaction medium allow higher macromolecular parameters and more stable rheological and transport characteristics to be obtained.

The promise of this approach is particularly evident for ionene-type systems based on tetrahydro-1,4-oxazine. In [42,43], it was established that varying substituents at the quaternary nitrogen atom, spacer type, and the distance between charge centers enables coordinated tuning of Tg/melting behavior and ionic conductivity. Importantly, these studies rely not only on reporting «high values» but on correlation analysis between structural parameters, thermal characteristics, and s, bringing the field closer to predictive «structure–property» models for PILs.

Overall, the current development stage of PILs is defined by the need to integrate polymer-science methodology with ionic-system design concepts. The most promising trajectory is the development of ionene-type polymeric ionic liquids as an architecture-controlled platform for establishing reproducible structure–property relationships and enabling rational design of materials with targeted functional performance [5,9,41–43].

### ***Conclusions***

The current state of polymeric ionic liquid research indicates that the field is undergoing a methodological transition: from empirical accumulation of application-driven results toward systematic analysis of structure–property relationships from a polymer-science perspective. PILs should be regarded as an independent class of macromolecular materials in which chain architecture, ionic center density, and supramolecular organization are decisive.

It is shown that the absence of universal correlations between ionic conductivity and parameters typical for low-molecular-weight ILs is caused by the complex interplay of segmental mobility, glass transition temperature, and domain structure. Therefore, interpretation of transport properties requires simultaneous consideration of macromolecular parameters that are not systematically reported in many application-oriented studies.

Ionene-type polymeric ionic liquids represent the most promising architectural platform for coordinated regulation of segmental mobility and ionic transport. Their development enables a transition toward reproducible structure–property relationships and predictive materials design. Controlled synthesis of PILs with targeted molecular weight and predictable physicochemical characteristics is possible only by integrating ionic liquid chemistry with polymer-science methodology.

It is also shown that ionene-type dimeric ionic liquids can serve as effective model systems for interpreting relaxation and transport processes in PILs, and as functional components in hybrid «PIL–ionic liquid» materials. Further progress in the field is associated with standardization of macromolecular parameters, systematic consideration of supramolecular organization, and integration of green chemistry and life-cycle assessment approaches, providing a basis for implementing PILs in high-technology electrochemical, membrane, and sustainable chemical processes.

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## ПОЛІМЕРНІ ІОННІ РІДИНИ ЯК ОБ’ЄКТ СУЧАСНОЇ ХІМІЇ ТА ХІМІЧНОЇ ТЕХНОЛОГІЇ: МІНІ-ОГЛЯД АРХІТЕКТУРИ, ЗАКОНОМІРНОСТЕЙ СТРУКТУРА–ВЛАСТИВОСТІ ТА ПРИКЛАДНИХ НАПРЯМІВ

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Полімерні іонні рідини сформувалися як окремий клас функціональних макромолекулярних матеріалів на стику хімії полімерів, фізичної хімії та матеріалознавства. На відміну від низькомолекулярних іонних рідин і класичних поліелектролітів, ці системи поєднують високу щільність іонних центрів із керованою макромолекулярною архітектурою, що зумовлює специфічну надмолекулярну організацію, релаксаційну динаміку та сегментально-контрольовані механізми іонного транспорту. У роботі узагальнено архітектурно орієнтований підхід до опису полімерних іонних рідин і запропоновано логіку класифікації за способом інтеграції заряджених фрагментів у полімерний ланцюг: системи з іонними групами у бічних ланцюгах, іоненові полімери та зшиті/сітчасті структури. Проаналізовано ключові структурно-властивісні залежності, зокрема вплив хімічної природи іонної пари, температури склування, молекулярної маси та доменної організації на іонну провідність і стабільність експлуатаційних характеристик. Показано, що відсутність стандартизації макромолекулярних параметрів і недостатня експериментальна верифікація доменної структури обмежують відтворюваність і порівнянність результатів та гальмують побудову узагальнених моделей «структура–властивості». Обгрунтовано перспективність переходу до раціонального дизайну через контрольований синтез іоненових архітектур, системне зіставлення параметрів «структура–температура склування/реологія–провідність», а також створення гібридних матеріалів полімерна іонна рідина–іонна рідина та полімерна іонна рідина–нанонаповнювач. Узагальнено прикладні напрями використання в електрохімічній енергетиці, мембранних технологіях, електрокаталізі та сенсориці й підкреслено необхідність інтеграції критеріїв сталості, зокрема оцінювання життєвого циклу, довготривалої стабільності та потенціалу рециклінгу.

**Ключові слова:** полімерні іонні рідини; іоненові полімери; макромолекулярна архітектура; структурно-властивостні залежності; іонний транспорт; доменна організація; мембранні матеріали; електрохімічна енергетика.

## POLYMERIC IONIC LIQUIDS AS AN OBJECT OF MODERN CHEMISTRY AND CHEMICAL TECHNOLOGY: A MINI-REVIEW OF ARCHITECTURE, STRUCTURE–PROPERTY RELATIONSHIPS, AND APPLICATION AREAS

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Polymeric ionic liquids have emerged as a distinct class of functional macromolecular materials at the interface of polymer chemistry, physical chemistry, and materials science. Unlike low-molecular-weight ionic liquids and conventional polyelectrolytes, these systems combine a high density of ionic centers with a tunable macromolecular architecture, which results in specific supramolecular organization, relaxation dynamics, and segmentally controlled mechanisms of ionic transport. This paper summarizes an architecture-oriented approach to describing polymeric ionic liquids and proposes a classification logic based on how charged fragments are integrated into the polymer chain: side-chain ionic polymers, ionene-type polymers, and crosslinked/network structures. Key structure–property dependences are analyzed, including the effects of ion-pair chemistry, glass transition temperature, molecular weight, and domain organization on ionic conductivity and the stability of operational characteristics. It is shown that insufficient standardization of macromolecular parameters and limited experimental verification of domain structures reduce reproducibility and comparability of results and hinder the development of generalized «structure–property» models. The prospects for a transition to rational design are substantiated through controlled synthesis of ionene architectures, systematic correlation of «structure–glass transition temperature/rheology–conductivity» parameters, and the creation of hybrid materials of the polymeric ionic liquid–ionic liquid and polymeric ionic liquid–nanofiller types. Application areas in electrochemical energy technologies, membrane processes, electrocatalysis, and sensing are summarized, and the need to integrate sustainability criteria, particularly life-cycle assessment, long-term stability, and recyclability potential, is emphasized.

**Keywords***:* polymeric ionic liquids; ionene-type polymers; macromolecular architecture; structure–property relationships; ionic transport; domain organization; membrane materials; electrochemical energy technologies.

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