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Biopolymer Degradation and Structure–Property Relationships in Ageing: A Polymer Science Perspective

Mona Chaurasiya[1,*], Gajendra Prasad[2]

Abstract

Ageing, as viewed from the polymer sciences perspective, is perceived as the gradual modification of the structure-property-function interrelationship of biopolymers such as proteins, polysaccharides, nucleic acids and their molecular aggregates. Such changes include structural organization, mechanical behavior, physicochemical stability and functional effectiveness. Ageing is an inevitable multifactorial polymeric process. Due to the growing aging population globally, more age-associated complications emerge, which are correlated with the molecular degeneration of biopolymeric complexes. An increasing elderly population leads towards higher occurrences of age-associated alterations in biopolymers' structure, property and function. Ageing is influenced by many factors that includes alterations in biopolymers and their properties. Biopolymers, presently, are the focus of research due to their biodegradability and their effectiveness to replace the non-biopolymeric compounds. Aging involves processes like genomic instability, telomere shortening, epigenetic alterations, deregulation of protein homeostasis, malfunction of macroautophagy, mitochondrial dysfunctions, cellular senescence, exhaustion of stem cells, abnormal intercellular signaling, chronic inflammatory response, and dysbiosis. All these factors can be perceived as instances of macromolecular deterioration similar to those occurring during polymer and composite aging processes. This article provides an overview of biological concepts of aging through the lens of the polymer sciences approach to aging and incorporates the basic principles of polymers and materials science.

Keywords: Ageing; Biopolymers; Structure–Property Relationships; Microstructural Properties; Polymer Degradation

INTRODUCTION

The phenomenon of aging is quite complex and multi-faceted process characterised by a gradual loss of functional ability at the biological level. Ageing leads towards increased vulnerability towards incurring diseases. In terms of molecular perspective, the process of aging can be explained by a series of modifications within biopolymers, including proteins, polysaccharides, nucleic acid, and related macromolecules that give shape to cells and determine their functional ability [1]. From the view of polymer science, this can be seen as changes within the macro-molecular architecture, microstructure, and physio-chemical properties that are associated with environmental, genetic and metabolic factors [2-5].

An increasing ageing population trend on a global level has drawn attention towards elucidation of the principles involved in the process of ageing that includes processes associated with of loss of function and degeneration. This is similar to the what is found in artificial polymers, biological polymers; as these also experience structural

***Author for Correspondence**
Mona Chaurasiya

[1]Research Scholar, Department of Biotechnology, L.N. Mithila University, Darbhanga, Bihar, India
[2]Associate Professor, Department of Botany, L.N. Mithila University, Darbhanga, Bihar, India

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deterioration characterized by cross-linking reactions, changes in structure and surface alteration, resulting in instability of the molecules and functional loss [10]. Such modifications are often associated with increase the occurrences of various diseases like cancer, heart diseases, diabetes, and neurodegeneration etc. [6, 32]. Hence, interpreting ageing from the perspective of polymer science and biomacromolecules would enhance the understanding of microstructural properties and molecular degradation in biological systems [7].

Recent advancements in the field of high-throughput "omics" studies, such as genomics, transcriptomics, and proteomics, have allowed for better understanding of macromolecules and their interactions [8, 9]. The high-throughput "omics" studies techniques mentioned above offer information about macromolecular structure of the biopolymers and their behavior in different physiological conditions. In other words, similar advanced techniques will help in understanding the behavior of biopolymers on parameters related to thermal stability and mechanical strength [65, 66] of biological macromolecules can be studied using "omics" studies. Thus, the phenomenon of aging may be reflected in terms of bio-macromolecular instability and degradation.

THEORIES OF AGEING FROM A POLYMER SCIENCE PERSPECTIVE

Ageing theories have been discussed within biological frameworks in the past may be understood using the concepts of degradation of biopolymers and materials science. Genomic instability [6], telomere shortening [31], response towards nutrient [11], epigenetic change [12], free radicle and oxidative stress [14,15] and the build-up of defective macromolecules [16–18] can be seen as degradation reactions at the molecular level similar to chain excision, oxidative degradation and chemical modification seen in polymers. All these processes indicate poor molecular structure, loss of biological function and diminished structural stability.

Antagonistic pleiotropy [13], metabolic imbalances [18], malfunction [19] and changes in cellular communications [20] can be associated with disruptions of the interaction between macromolecules and energy transfer processes occurring in biological polymers. Likewise, proteostasis disruption [10] and autophagy defects [21, 25] are associated with incorrectly folded proteins and aggregates, which is similar to phase separation, heterogeneities in the microstructure, and defects in polymer composites. All these will eventually result in disruption of the structure-property relationship and functionality.

The extended hallmarks of ageing [10, 34] such as chronic inflammation [20], dysbiosis [21], and stem cell exhaustion [22], may be seen as consequences of progressive instability of biological polymer systems and their interfaces. In that sense, these phenomena are comparable with the interface degradation, decreased efficiency and microstructure damage occurring in polymer. Hence, ageing is perceived as a process of gradual loss of biological polymer systems and macromolecular degeneration.

BIOPOLYMERS AND MOLECULAR MECHANISMS

Biopolymers are naturally occurring polymers that are produced inside a living being such as a plant or an animal. Biopolymers are organic compounds obtained from natural sources, i.e. plants, animals, microorganisms, or other living beings and are biodegradable [55] as well as sustainable [56, 69]. Biopolymers are usually flexible, elastic or fibrous materials. Polysaccharides and proteins are some of the natural polymers produced by living organisms [57] whereas synthetic polymers are chemically produced [58] and are usually non-biodegradable or slow biodegradable [59,60] having high physiochemical resistance [61], toxicity [62], thermal stability and low sustainability [61].

The molecular weight or the molar mass of biopolymers affects its physical polymeric properties and is directly linked with the chain length. The higher the molecular weight of a biopolymer the higher is its tensile strength [67], melting temperature, resistance, toughness and viscosity. Thus, biopolymers which have higher molecular weight are difficult to process due to its high viscosity which relates to its properties (e.g. thermal, degradation). The molecular architecture of biopolymers affects its overall

design and thus affects its melting temperature and degradation rates [51]. Further, the linking in biopolymers such as cellulose, pectins, collagen, chitosan etc. are affected by water vapor annealing process, cellulose nanomaterials, pectin materials, cross-linking effects and others based on their mechanical properties [52,68,69]. As such in larger biopolymers e.g. protein that have high molecular weight have less diffusion coefficient suggesting inverse correlation [53] and are less likely to distort easily [54].

Oxidative degradation implicates disintegration of biopolymers wherein action of oxygen is involved and can be induced by factors such as thermal-oxidative reactions, heat, mechanical stress and weathering in the presence of free radicles. Free radicals react with oxygen-producing oxy- and peroxy-radicals. Free radicals may react with each other or remove hydrogen from the biopolymer chains [63] thus causing degradation of biopolymers. The oxidative degradation of biopolymers is associated with several factors, such as the chemical organisation, molecular weight, structure, oxygen diffusion, etc. Mostly, the oxidative degradation pathways for the main polymer and biopolymer classes have been established and the appropriate stabilizing strategies have been assessed. The oxidative degradation pathways for biopolymers irrespective of the degradation pathway can be summed up as (i) formation of free hydrogen radicals, resulting in shortening of bio-polymeric chain and/ or formation of unsaturated bonds; (ii) formation of free entities that are formed by diffusion of oxygen in chemicals leading to formation of either metastable or stable species e.g. carbonyl, hydroxyl groups, peroxides, hydroperoxides, etc. (iii) disintegration of some oxygen containing chemical entities resulting in autocatalytic effect such as hyroperoxides.[64].

Biopolymers are vital and are important as they control the structural and functional characteristics of all biological systems. The proteins, polysaccharides, nucleic acids, and other macromolecular structures, involved with living organism, provide mechanical stability, biochemical functionality and cell signaling capabilities essential for proper functioning of all biological systems. Aging results from the gradual loss of the ability of these molecules to maintain their mechanical characteristics, structure and self-assembly ability [23-24]. This ultimately leads to changes in the physical properties, structural stability, and biological functionality of the molecules involved.

Biopolymers are important in ageing studies. Biopolymers and their complexes are emerging as important resources for biomedical applications that support healthy ageing, including tissue scaffolds, drug delivery carriers, and also for regenerative medicine [38]. Biopolymer's exclusive structural, biochemical and other properties offer opportunities to address age-related wear and tear of biopolymeric functions for beneficial outcomes in ageing populations. Biopolymers, as such, play a crucial role in maintaining cellular structure, mechanical properties and functional stability; all of which undergo progressive deterioration during ageing [39].

In recent years, research in the field of polymer chemistry have resulted in understanding the behavior of biopolymers using nanoparticles, hybridization techniques, and methods of material characterization [26]. These methods allow for examination of the microstructural properties, thermal stability and intermolecular interactions in biological systems. The unique structural and functional characteristics of biopolymers are useful in the development of biomaterials for biomedical applications including tissue engineering, drug delivery, and regeneration medicine [27].

SPECIFIC BIOPOLYMER IN AGEING

Polysaccharides

The polysaccharide is an example of natural polymer that exhibits its functions structurally as well as functionally. The ageing of polysaccharide can be considered based on the structural functional relation, molecular stability, and exchange reactions in the biological system. Research has demonstrated the effect of the biological system on the regulation of oxidative stress and mechanical behavior [28-30]. Therefore, it can be stated that studying the polysaccharide is essential for the

functional efficiency of the biopolymer system.

In light of the polymer science viewpoint, these biopolymers, or more precisely, the biomolecules, are generally large sequences of monosaccharides bound together through glycosidic linkage. This long sequence of monosaccharides reveals a complicated structure such as linear, branched, and cross-linking structure of polymers. Such structures directly affect the functional performance of the biopolymers. There is an occurrence of modifications with advancing age, which are related to variations in the molecular weight of molecules, structural chains, and interaction among molecules.

Hyaluronic acid (HA) is one of the most widely researched polysaccharide components involved in the process of aging. HA is a type of linear glycosaminoglycan that is part of the extracellular matrix. The unique ability of HA to form viscoelastic substance is based on its high molecular weight and the ability to attract water due to hydrophilicity. However, under the influence of aging processes, HA experiences enzymatic degradation by enzymes known as hyaluronidases and oxidative stress, and thus its molecular weight lowers [35].

In a similar manner, other extracellular matrix polysaccharides like chondroitin sulfate and dermatan sulfate undergo a certain form of structure and composition changes over time. Changes that are linked to the charge, density, and protein binding capability that cause changes in the cellular matrix, and consequently alter its functionality. From a polymer science perspective, certain sulfation patterns, polymer length, and distribution can lead to polyelectrolyte properties which change over time and affect tissue elasticity and signaling [36].

Another significant connection between polysaccharides and aging can be found in advanced glycation end products (AGEs). Non-enzymatic reaction between reducing sugars and protein and polysaccharides leads to cross-linking and rigidity of biological tissues, similar to the polymerisation of a thermosetting polymer where cross-linking results in the decreased movement of chains and increased brittleness of the polymer. The same occurs in aging biological tissues with regard to the formation of cross-linking between collagen and polysaccharides in circulation and skin tissues [37].

Polysaccharides and their ageing from polymer science point of view, can be examined relative to the stability of biopolymers and environmental influences. Degradation due to factors like pH, temperature and oxidative stress involved in hydrolysis and oxidation, will result in deterioration of their molecular structure. The degrading processes are similar to the degradation process of biopolymers. Hence, polysaccharides act as model systems for examining the performance of biopolymers.

Transformation of bio-polysaccharides to combat the degradative effects caused by ageing is being actively pursued. The chemical structure, cross-link structure, and composites have all been explored as means of improving stability and restoring mechanical properties. For instance, hydrogel made from hyaluronic acid with a controllable level of cross-linking has been designed to emulate the extracellular matrix (ECM) [40].

Nucleic Acids and Telomeres Structures

The structure of nucleic acids are usually polymers that have unique properties which make them important for cellular activities. Nucleic acids, especially deoxyribonucleic acids (DNA), are examples of very natural biopolymers whose structures and functions are closely associated with the ageing phenomenon. In polymer science terms, DNA refers to a helix that has some flexibility, consisting of long chain of nucleic acids (A, T, G & C) that are bonded through phosphodiester bond. DNA has charges that are ordered in a unique arrangement that demands high organization/packing. Ageing involves alteration of packing that affects conformation.

The ends of the chromosomes are generally called telomeres, which are characterised by TTAGGG

repeat sequences, and these sequences are known to be involved in the process of aging. The telomere region consists of repeated units of noncoding DNA located at the ends of chromosomes and forms G-quadruplexes. It is a peculiar structure that normally consists of four strands and is abundant in guanines, maintaining its structure with help of cationic and hydrogen bonds. One can look at this particular nucleic acid structure through the prism of polymer science, where polymeric structures are folded depending upon the length of the chain and its strength along with end capping of the telomeres.

Progressive loss of the ends of DNA at the tips of the chromosome occurs during the division process of a cell causing a shortening of DNA. This shortening may be explained using the theory of polymer science through understanding the loss of the molecular structure of the DNA molecule during replication up to a point where the molecule may trigger ageing, which is therefore replicative, hence bringing about changes to the structure of the DNA molecule. Progressive shortening of the DNA molecule, especially in the telomere region, may be attributed to polymer degradation while keeping in mind a controlled cycle of division occurring at the end of the polymer instead of the middle to avoid extreme degradation [41].

Telomerase, an enzyme that protects the ends of the telomere, provides further polymer regulation of the DNA. The enzyme known as telomerase responsible for the protection of the ends of the telomeres gives an extra layer of regulation in terms of polymer protection. The nucleotide repeats found in the ends of chromosomes serve the purpose of protecting the chromosomes during the process of cell division in order to avoid shortening. The polymeric activity in relation to ageing occurs mostly in the somatic cells. However, indefinite cell division related to telomere activities is found in germ cells but not the somatic cells, except for cancerous cells. The polymeric cell division activities of somatic cells are usually well regulated due to the limited nature of polymer renewals, while on the other hand, abnormal telomere activity is observed in cancerous cells due to the indefinite replication which acts as a self-renewal polymer system.

Telomerase protects the ends by maintaining their polymer chain length even after some number of divisions. In doing so, it balances out the effect of telomere shortening [31], which is important in maintaining the replicating ability of a cell through cell division in the somatic cells. Telomere shortening may thus be considered to occur due to a reduction in the chain length of the biopolymers, just as in a synthetic system there is a reduction in the molecular weight of the polymer leading to its instability and vulnerability to breakdown.

The oxidative stress causes more damage to the telomere sequence as a result of alterations in the DNA molecule particularly in those areas where the concentration of guanine is high. These alterations cause changes in the stacking arrangement that is π–π as well as hydrogen bonds causing changes in the conformational properties of the DNA molecule. In the case of polymer science, this phenomenon can be seen as the degradation of the polymer due to accumulated defects in the polymer molecule [42].

Besides the modifications of the ends of the nucleic acids, the structural modifications within the chromatin structure result in changes in the protein that is formed from this DNA. Normally, the DNA wraps around the histone proteins, forming nucleosomes, to form a polymer complex structure. The aging process itself involves many genetic and epigenetic modifications; however, the latter are important for our discussion here. For example, DNA methylation and histone acetylation influence the DNA binding to the histones, affecting the pattern of gene expression. Such modifications are likely to cause the structural transition of the polymer network similar to glass transition or phase transition in artificial materials [34].

From the standpoint of polymer physics, telomerase may be considered a chain-extension/repair process, similar to the concepts of self-healing polymers or living polymers. However, its operation is strictly controlled and its malfunction causes cancer, where uncontrolled cell proliferation occurs due to unlimited replication capabilities [43]. Telomere DNA chains can adopt several secondary structures,

namely G-quadruplex and i-motif structure formations. This is important from the point of view of mechanical strength and enzymatic availability. In terms of polymer chemistry, these are chain transition processes. They happen depending on ion and pH concentrations.

Due to recent developments in polymer-based nanotechnology, it is possible to create artificial mimics of telomeres as well as telomere probes. For instance, the synthesis of molecules and polymers that can stabilize G-quadruplexes has become a promising approach towards treatment of age-related conditions, illustrating the application of polymer chemistry concepts in the context of nucleic acids.

Oxidative stress is a key factor in the ageing of nucleic acids. Reactions between ROS and nucleic acid bases, as well as their phosphodiester backbone, cause strand breakage and alterations in nucleotide bases. Guanine being highly oxidisable makes it a target for ROS reactions, with telomeric sequences being especially prone to attacks. Hence, a faster than expected rate of telomere shortening occurs, independently of DNA replication [42]. When considering oxidative stress from the perspective of macromolecular chemistry, it may be treated as chain breaking, cross-linking, and generation of non-functional monomers.

The DNA is not present as a biopolymer in the cell, but exists as a complex composite material, where the genetic information is present. It is wound around the histone protein in the complex structure, consisting of both major and minor grooves to provide a structure in a hierarchical way. The DNA is present as a biopolymer that interacts with histone proteins to form a structural path for the cell machinery. Aging in the organism is known to affect the structure of DNA through nucleosome repositioning and epigenetic modifications, like DNA methylation. We know that methylated bases in the DNA protect themselves. This can be understood by considering the transition in polymers as in crystallization and phase separation.

The two biopolymers mentioned above are depicted to show similarity with the process involved in the structure and evolution, along with other processes like aging and degradation, which play an important role during aging. The inclusion of the concepts from the polymer science field, such as the length of biopolymers, stability and the kinetics associated with their degradation, will give a much better understanding of the aging process through which drug compounds can be developed.

Polyphenols

Polyphenols refer to organic compounds that are naturally occurring within plants and include a group of macromolecules or oligomers. These polyphenols have been found to participate in cell structure formation, defense, and regulation of various bio-processes in living beings. These polymers are organic compounds containing aromatic groups together with hydroxyl groups. There is a relationship between polyphenols and aging with regard to biological functions as well as material functions. With regard to biological functions, polyphenols participate in the oxidation processes and with regard to material functions, there is the formation of linkages.

The tannins, lignins, and flavonoids that form polyphenolic substances have particular polymer structure organization, for instance, the lignin formation process involves the polymer chain made of phenylpropanoid monomers having branched structure organization. The unstructured polyphenolic structure of these polymeric substances makes them thermally stable and strong enough to withstand enzyme degradation. Tannin has such a structure, which is linearly branched through the ester bonds, but unlike lignin, the structure of tannin substance is less complex. The structure formation of these biopolymers in natural conditions is similar to that of polymer that can resist stress without degrading.

Polyphenolic compounds, physically and chemically, denote macromolecules which possess a dense capacity of forming complexes or chelates with metals, hydrogen bonding and π–π interactions. This is because their physical and chemical features are associated with biopolymers, which include proteins,

polysaccharides, and nucleic acids. They share resemblance in that they form bonds like those formed by polyphenolic compounds.

The process of ageing is characterized by the reduction in biological activity and is viewed as oxidative and molecular damages build-up during one's life span. In this regard, polyphenols are known for having anti-ageing features which help minimize the oxidative damages as well as protection from biomacromolecules due to cross-linking reactions. This type of reaction helps stiffen up tissue structures. The Reactive Oxygen Species (ROS) are independent radicals that are neutralized through hydrogen donation by polyphenols, which halts their oxidative chain reaction. This is similar to the properties exhibited by phenolic stabalisers in synthetic polymers, which prevent oxidative degradation. Oxidative prevention depends on the molecular structure, degree of hydroxylation, and conjugation in the aromatic ring [44].

When polyphenols are oxidized, they lead to the production of quinones, which are very reactive electrophilic substances that have the capability of creating covalent bonds with proteins and polysaccharides. Cross-linking of covalent bonds resembles artificial resins. Therefore, ageing has been associated with stiffness, which is seen in polyphenolic raisins. In regard to aging cells, the relationship between polyphenols is attributed to the elasticity of collagen cells, whereby elasticity decreases with age [45].

Extracellular matrix of a cell is created *via* a highly complicated interaction of biopolymers such as collagen, elastin, and glycosaminoglycans. Polyphenols are capable of binding to extracellular matrix – biopolymers, causing a transformation in the mechanical and physical characteristics of the biopolymers [48]. For instance, tannins cause collagen fibers to cross-link using hydrogen bonds and covalent bonds. This results in increased resistance to enzymatic degradation and thermal stability for tannins. This technique has been widely applied in the leather industry and is currently being researched for biomedical purposes.

Polyphenols are natural polymers where cross-linking occurs to raise viscosity and density, having regard for polymeric interaction. Therefore, polyphenols could enhance tissue strength although overlinking between the biopolymers causes hardness and decreased functionality particularly among collagens. Tissues like collagen which is a biopolymer, have a direct correlation with loss of functionality as we age and especially during movement due to increased hardness. For tissues to remain functional, there must be an optimum level that ensures efficiency and so study of oxidative stability and interaction among the biopolymers [33] would be necessary.

As pointed out earlier, lignin, one of the significant polyphenolic biopolymers, offers itself as a model of aging. It exhibits complex structure which is able to resist both condensation and depolymerization reactions. Lignin is the natural biopolymer that undergoes various effects like radiation, humidity, and chemical among others. All these effects occur in synthetic polymer too. Over time, there will be alterations in the cross-linkage pattern of the polymer, degradation of the polymeric chains, and change in the states of polymerization such as rigid, glassy, or rubbery states among others. The same happens in biological tissues where there is accumulation of toxic polyphenolic molecules leading to stress in cells. It becomes evident that in older plant tissues, the process of lignification results in aging as the cell wall becomes hard with low permeability [45].

The hybrid networks consist of protein and polysaccharide both being biopolymers. The polyphenol compounds have a high affinity for protein and polysaccharide biopolymers. Their bondings take place through the complex reaction of covalent bonds, non-covalent bonds, and surroundings. As the body ages, these bonding processes result in advanced glycation ends (AGEs) formation and other complex macromolecular compounds. Polymer science views these activities from a multidimensional point of view since these activities involve several characteristics influencing stability and evolution. The

polyphenols also show associations in both protection and catalysis in biopolymers [46]. The antioxidants halt the degradation process of the biopolymers by neutralizing the free radical ions. Therefore, it is imperative that protein structures and membranes are preserved during the ageing process caused by oxidative stress.

In subsequent reactions, intermediates such as those of oxidation of polyphenolic substances damage the polymer, hence hastening its breakdown. This is an interesting aspect of polymers based on the property of being a dynamic modifier, as observed during ageing. Polyphenolic substances have emerged as important biomaterials in the form of hydrogels, films and nanocomposites, showing mechanical strength, antioxidant nature and bio-degradability. For instance, catechol-based polymers made from mussel adhesive proteins have shown excellent adhesion and self-healing abilities through reversible covalent and non-covalent bonding [49-50]. These examples show how polyphenolic substances may be used in designing substances that influence ageing [47].

CHALLENGES & FUTURE DIRECTION

The use of biopolymers and advances in biopolymers remain an issue due to difficulties associated with measuring the correlation between modification of biopolymer properties and function deterioration caused by aging. In order to establish a better structure-function-property relationship, it is essential to integrate high-tech analytical techniques with multi-scale modeling approaches. The utilization of genomics, proteomics and metabolomics and other high throughput technologies could pave the way for creating large datasets that can be analyzed through computational methodologies.

Degradation kinetics, interfacial behavior and microstructure evolution in biopolymers need to be thoroughly investigated in future studies. Moreover, the design of biopolymer composites and biomaterials holds great potential in overcoming aging-induced issues.

CONCLUSION

Ageing can be easily analyzed through the lens of polymer science where it becomes clear that the phenomenon of ageing is characterized by the continuous degradation and change in the structure-function relationship of biopolymers. It is possible to see that structural organizational and physio-chemical changes at the level of macromolecules become the foundation for understanding the processes of ageing and health consequences. Combining the principles of polymer science and theories of biological ageing allows one to develop a holistic approach to the study of ageing phenomena at the molecular level.

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