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Recent Trends in Application of Amphiphilic Block Copolymer Based Hydrogels

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Abstract: ***Amphiphilic block copolymer based hydrogels (ABCHs) with hydrophobic and hydrophilic blocks can be constructed by employing synthetic as well as natural polymers. One of the crosslinking methods is covalent crosslinking, in which both hydrophilic and hydrophobic parts are chemically crosslinked together. In the present chapter, we have explored the ABCHs synthesized through free radical solution polymerization technique, in which, instead of using conventional crosslinking agents, the monomers contribute towards a hydrophilic polymer segment as well as crosslinking. The properties of these hydrogels, such as biocompatibility, low toxicity, maintained biodegradability, self-assembly, and response to environmental stimuli, have also been detailed. Since the hydrogels have appropriate mechanical properties and can regulate the release of hydrophobic drugs, they have found several uses in the biomedical field. A hydrophobic drug can be uniformly loaded into ABCHs because of a suitable connection between the hydrophobic drug and the hydrophobic polymer chain. They are also capable of delivering both hydrophilic and hydrophobic drugs because of the presence of both segments. We have further detailed the ABCHs widely used in drug delivery, wound dressing, tissue engineering, and various biomedical applications.***
Keywords: Amphiphilic block copolymer, Hydrogels, Crosslinking, Biocompatibility, Biomedical application.

I. INTRODUCTION

Amphiphilic block copolymers (ABCs) are copolymers that have chains that are both hydrophilic and hydrophobic. An additional kind of widely used amphiphilic molecules with a large molecular weight are ABCs. The unique chemical structure of ABCs, which contain both hydrophilic and hydrophobic blocks, makes them extremely captivating [1]. While hydrophilic blocks offer swelling, transport, and biocompatibility, hydrophobic domains provide nanoscopic cavities and physical crosslinks for insoluble cargos. Due to these blocks, ABCs can self-assemble into a variety of nanostructures in bulk and in solution, which are comparable to the aggregates of small-molecule amphiphiles [2]. The morphologies of these resulting nanostructures can be significantly influenced by the hydrophilic/hydrophobic ratio, copolymer content, solvent characteristics, and other factors [3]. Diblock copolymers, as well as triblock copolymers, have been the most widely considered copolymers. In addition to some polyacids and polybases, other hydrophilic blocks including polyethylene oxide (PEO), polyethylene glycol (PEG), polyvinyl caprolactam (PVCL), polyvinyl alcohol (PVA), poly(N-isopropylacrylamide) (PNIPAM), and polyethylenimine (PEI) are identified. On the other hand, polybutadiene (PB), polycaprolactone (PCL), polypropylene oxide (PPO), polylactic acid (PAA), polystyrene (PS), polymethylacrylate (PMA), etc. can be the hydrophobic blocks. Some common examples of the hydrophilic and hydrophobic blocks, considering their charges, are illustrated in Figure 1 [4]. During the synthesis of the amphiphilic block copolymer, a crosslinking agent and an initiator were added, which led to the creation of a hydrogel through in situ free radical crosslinking polymerisation [5]. A sol–gel phase transition can be used to construct physically crosslinked hydrogels in situ in response to external stimuli such as temperature, pH, redox, light, and enzymes. Amphiphilic block copolypeptide hydrogels are one type of injectable hydrogel that is based on chemically synthesized polypeptides. The structure phase behaviour results from self-assembly, which is the creation of domains of hydrophilic groups (in contact with a polar solvent) and domains of hydrophobic groups (in contact with a nonpolar solvent) [6]. Although typically are combined hydrophilic and hydrophobic domains, amphiphilic block copolymer-based hydrogels (ABCHs) are a multifunctional substance. Their regulated release and multifunctional qualities make them widely used in biomedical applications (drug administration, tissue engineering, wound healing) [7]. This article covers the synthesis of ABCHs with hydrophilic and hydrophobic segments. Because of their mechanical strength, biocompatibility, and controlled release features, these ABCHs have a lot of potential for use in biomedical applications.

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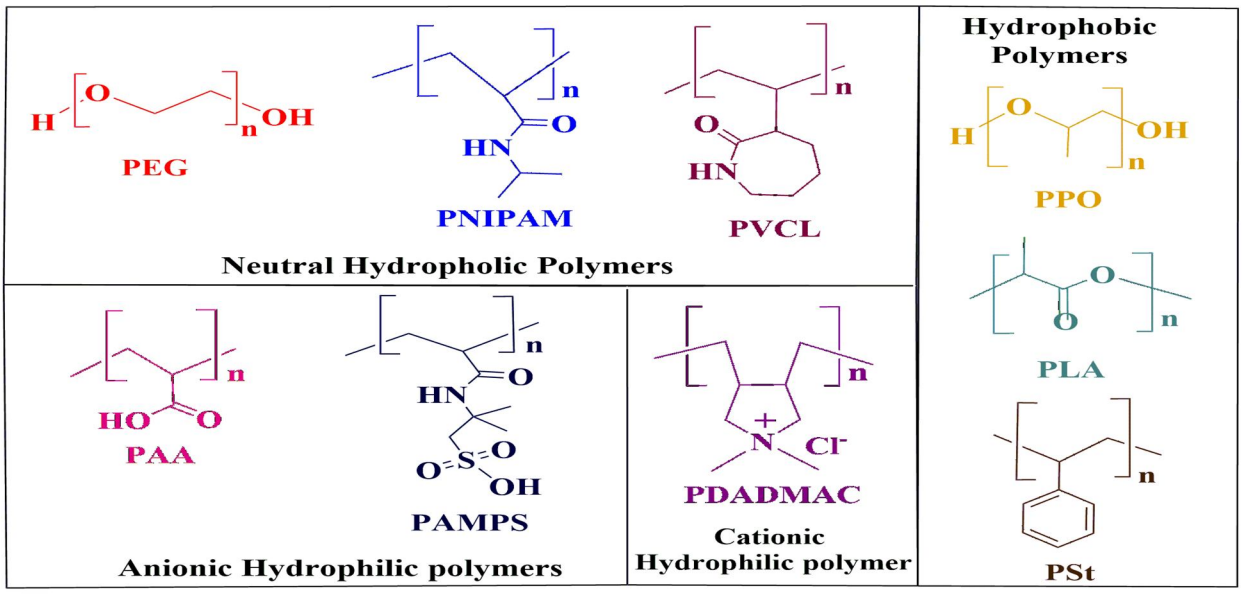

Figure 1. Some examples of Hydrophilic and Hydrophobic Polymers

II. AMPHIPHILIC BLOCK COPOLYMER BASED HYDROGELS

A common technique to generate amphiphilic block copolymer based hydrogels (ABCHs) is to use hydrophilic and hydrophobic segments, respectively, for constructing three-dimensional (3-D) networks that form block copolymers. The resulting three 3-D ABCHs have significant water absorption capacity while preserving their semi-solid structure. They exhibit remarkable biostability and prospective drug release [8]. The hydrophilic and hydrophobic metrics, the concentrations of the initiator or polymer, and the reaction factors (time, temperature, etc.) can all be changed to modify the characteristics of hydrogels, such as swelling–deswelling rate, stiffness, degradability, and mech size [9]. The kind of monomers, charge density, and degree of cross-linking affect the responsiveness hydrogels to external stimuli. By absorbing large amounts of water, ABCHs appropriately expand, and contract to provide controlled medication release. These characteristics enabled them, appealing for a number of uses, including several in the biomedical domain, such as cell encapsulation, medication administration, tissue engineering, and gene therapy carriers [10]. Hydrogels are capable of holding different amounts of water due to their nature being influenced by chemical and physical stimuli. For example, polystyrene-b-poly(methyl methacrylate) is fabricated by polymerizing styrene first, and then methyl methacrylate (MMA) from the reactive end of polystyrene chains. Researchers in clinical research consistently discover, however, that the use of drug carriers may result in low loading efficacy, possible adverse effects, and poor biocompatibility and biodegradability. Effective clinical application is further hampered by the comparatively high cost and intricate synthesis needed for these biological materials that serve as drug carriers [11]. Both polyethylene glycol and poly (ethylene oxide) are water-soluble, crystalline, thermoplastic polymers. By combining with various hydrophobic segments, PEG and PEO, which are hydrophilic, can be used to create amphiphiles and subsequently create hydrogels [12]. Conventional hydrogels, on the other hand, have weak and flexible mechanical qualities and are quickly broken, even under moderate tension. Due to their particularly high mechanical strength and toughness, double-network (DN) hydrogels have recently been proposed and attained an enormous amount of attention [13]. The design of the dual properties of amphiphilic hydrogels based on peptides has received a lot of attention, not only because of its advantageous properties including high compatibility, biodegradability, and ease of synthesis, but also because of their potential uses in the biological and pharmacological arenas. The ability of peptides to self-assemble into various nanostructures, such as nanofibers, which can then go on to form amphiphilic hydrogels [14]. Copolymeric hydrogel networks usually do not dissolve in water. Two kinds of copolymeric hydrogels are poly(vinylpyrrolidone-co-acrylic acid) and poly(lactic acid)-poly(ethylene glycol) (PLA-PEG). The poly(ethylene glycol)-block-poly(-lactic acid) nanoparticles (PEG-b-PLA NPs) used in the design of ABCHs allow the dual loading of a hydrophilic molecule into the aqueous bulk of the gel and a hydrophobic molecule into the PEG-b-PLA NPs [15]. In particular, using peptide amphiphiles (PAs) with both hydrophobic and hydrophilic chains is an alternative technique for creating amphiphilic peptides. On activating macroradicals, the development of functional monomers generates branching, which in response produces cross-linking [16].

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For biomedical applications like drug delivery, ABCHs combine hydrophilic and hydrophobic segments to create robust, water-absorbing 3D networks. However, complicated synthesis, high cost, and low drug loading are limitations; more recent designs, such as double networks and peptide amphiphiles, enhance strength, compatibility, and drug loading.

III. SYNTHESIS OF AMPHIPHILIC BLOCK COPOLYMER BASED HYDROGELS

Amphiphilic co-networks (APCNs) are a form of hydrogel which can be defined for the continual existence of both hydrophilic and hydrophobic components, in the network structure, that are chemically cross-linked together. Three primary reaction techniques viz. prepolymer agent chemical coupling reactions, ionic crosslinking, and free radical crosslinking are used in their synthesis [17]. Free radical polymerisation, which has the advantages of high reactivity, high conversion, and minimal conditions for reaction, is the most widely used cross-linking method for generating hydrogels. Recently, ABCHs like poly(acrylamide)-co-poly(hydroxyethyl)methacrylate-co-poly(cyclohexylmethacrylate) hydrogel and P(Aam)-P(HEMA)-P(CHMA) hydrogels have been fabricated by free-radical solution polymerization technique. The free radical crosslinking method is used widely than other chemical crosslinking reactions for Aam/HEMA/CHMA based amphiphilic hydrogels. For the typical synthesis of a homogeneous mixture of hydrophilic monomers acrylamide (Aam), HEMA, and CHMA of double distilled water was prepared at room temperature (RT). Subsequently, azobisisobutyronitrile (AIBN) initiator was added and stirred under nitrogen purging and then placed in an oven for at RT. Rather than using the traditional crosslinkers, such as N, N-Methylene-bis-acrylamide (MBA), ABCHs have been synthesized by employing the cross linker HEMA in the free radical solution polymerization process [18]. Phase separation during crosslinking can be prevented by using amphiphilic functional multiblocks, as recently shown. As a consequence of the amphiphilic segments in the initial amphiphilic multi-blocks being covalently connected, amphiphilic multiblock hydrogels have been developed [19].

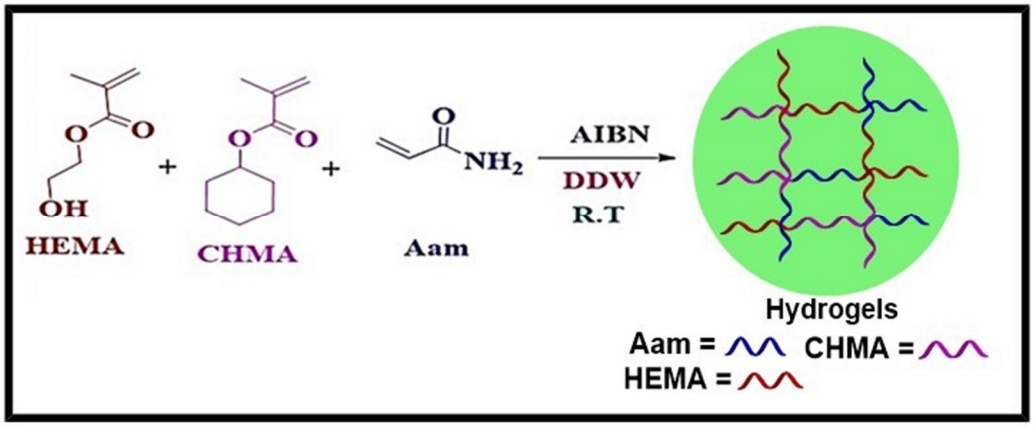

Figure 2. Synthetic route of amphiphilic block copolymer based hydrogels (ABCHs)

IV. PROPERTIES OF AMPHIPHILIC BLOCK COPOLYMER BASED HYDROGELS

Amphiphilic block copolymer based hydrogels, shown in **Figure 3**, have a variety of useful features, including high mechanical strength, biocompatibility, self-assembly, and response to environmental stimuli [12].

A. Mechanical Properties

In terms of their swelling behavior, the mechanical properties are linked with the external stimulating environment [8]. Hydrogels' mechanical properties can be modified according to the requirements for specific applications. The stiffness, elasticity, and strength of hydrogels are influenced by variables such molecular weight, crosslinking density, and polymer composition. For tissue engineering and controlled drug release, hydrogels containing a variety of functional groups may exhibit a variety of mechanical features [14]. The density of the network's bridges and nodes, not the sheer quantity of loops, is the primary factor influencing the mechanical characteristics of multiblock hydrogels. Drug delivery scaffolds based on amphiphilic block copolymer always deteriorate in their mechanical properties and stability under the function of the molecules that contain the drug. Hence, one of the most important goals in the research field of drug delivery systems is to increase the mechanical strength of the hydrogel after drug encapsulation [16].

B. Biocompatibility

ABCHs consist of amphiphilic polymers that include both hydrophilic and hydrophobic portions. These portions allow them to go through a sol–gel phase transition. ABCHs are more likely to exhibit inherent biocompatibility as injectable in the field hydrogels when crosslinking agents are absent and hydrophobic and hydrophilic chains are combined. In order to promote tissue regeneration and therapeutic efficacy, biocompatible hydrogels reduce adverse effects and make interactions with biological surroundings more feasible [20]. By utilizing significant design concepts, researchers designed polymer–nanoparticle (PNP) gels made of NPs that are both biocompatible and biodegradable. The PEG-b-PLA PNP hydrogels are a viable alternative to PNP gels since they can be produced with a diameter of 100 nm in a consistent and scalable form. They are also biocompatible and allow the differential release of various drugs in species following subcutaneous implantation [15].

C. Self-Assembly

ABCHs are formed when biomolecules, such as polysaccharides, proteins, peptides, or amphiphilic polymers, self-assemble under specific conditions. The natural molecular recognition and self-assembly capabilities of proteins and peptides enable the creation of hydrogel networks and hierarchical structures [11]. Block polyelectrolytes are designed to assemble directly to create a network of extensively connected nodes that reduces loop formation. Physical hydrogels are typically produced by the self-assembly of amphiphiles and block copolymers. In essentially suitable circumstances, hydrophilic and hydrophobic chains can self-assemble, encapsulate hydrophobic medications, and release them in response to environmental changes. These materials are widely used in drug delivery and cell culture [12]. For drug delivery and tissue engineering, hydrogels' mechanical strength, biocompatibility, and self-assembly can be adjusted. Designs emphasise stable drug loading, safe biological interaction, and controlled release via hydrophilic–hydrophobic chain assembly.

V. APPLICATIONS OF AMPHIPHILIC BLOCK COPOLYMER BASED HYDROGELS

Figure 3 portrays several advantageous applications of hydrogels based on ABCs, including drug delivery, tissue engineering, wound dressings, and other biomedical applications.

A. Drug Delivery Systems

Hydrogels consisting of covalently cross-linked polymers, to enable targeted delivery of medicines, have been recently developed. Further, amphiphilic triblock co-polymers, like poly(*N*-isopropylacrylamide)-*b*-poly(4-acryloylmorpholine)-*b*-poly(2-(2-nitrobenzyl)oxy)carbonyl)amino)ethylmethacrylate) (PNIPAM-*b*-PNAM-*b*-PNBOC) have been used in the construction of thermoresponsive hydrogels that are effectively incorporated with hydrophobic drugs. Thus fabricated hydrogel has been loaded with both the hydrophobic and hydrophilic drugs like doxorubicin and gem, respectively. Below the lower critical solution temperature, the hydrophobic and temperature-responsive components of the triblock copolymers first assemble into micelles. The hydrogels of physically cross-linked micellar nanoparticles have been formed by using higher polymer concentrations and temperatures above the critical gelation temperature [10]. The chemical structure and composition of polymeric smart materials allow them to react to a variety of external stimuli, including ionic strength, temperature, pH, light, electrical, magnetic, chemical, and biological stimuli. Supramolecular hydrogels containing PCL-PEG have been created for drug delivery. The efficient noncovalent intermolecular binding interactions between two or more linear polymers and cyclodextrin CDs are the phenomena that cause supramolecular hydrogel systems [21]. Due to their ease of use, minimal negative impact on tissues when compared to certain other stimuli, and ability to distinguish between body temperature and room temperature, thermoresponsive hydrogels have been the subject of much research. Poly(ε-caprolactone)-Poly(ethylene glycol)-Poly(ε-caprolactone) PCL-PEG-PCL, amphiphilic triblock copolymers made of polyethylene glycol (PEG) and poly (ε-caprolactone) (PCL), form a thermoresponsive, in situ forming, and biodegradable hydrogel that has been examined as a drug delivery system [22].

B. Tissue Engineering

A special class of biocompatible three-dimensional polymers known as hydrogels can function as a scaffold and replicate the characteristics that distinguish various bodily tissues. Variable covalent bonds and non-covalent interactions, including hydrophobic, electrostatic, and hydrogen bonding, are exploited to create self-improvement hydrogels. Hydrogels have a broad range of medicinal uses, including medication administration, tissue engineering, surface coating, and regeneration [23]. Hydrogel scaffolds act as matrices or scaffolds that trap cells, enabling them to proliferate and develop, ultimately repairing the damaged tissue. Hydrogels with higher biocompatibility for use in biomedicine can be created using more biogenic amphiphilic materials [17].

Synthetic polymers used for developing hydrogel matrices for cardiac tissue engineering include polycaprolactone (PCL), poly(ethylene glycol) (PEG), polylactide (PLA), polylactide-co-glycolic acid copolymer (PLGA), polyacrylamide (PAAm), and polyurethane (PU). Synthetic polymers are preferred over biological ones, due to the ease with which their physicochemical properties, such as water affinity, modulus, and degradation rate, may be modified according to the demands of cardiac muscle tissue engineering [24]. PEG–PCL, PEG–PLA, and polypeptide-based hydrogels are examples of amphiphilic block copolymer hydrogels that are utilised as scaffolds in bone, cartilage, nerve, and soft tissue engineering because of their biocompatibility, controlled degradation, and capacity to stimulate cell growth [12]. Polypeptide-based amphiphilic hydrogels, such as poly(L-lysine)-b-poly(L-leucine), have also been designed to mimic natural protein structures, supporting muscle and cartilage tissue engineering by enhancing cell adhesion and proliferation [25].

C. Wound Dressing

The relatively easy manufacturing conditions and the incorporation of numerous bioactive ingredients make hydrogels a suitable option for wound dressings. To attain the most effective wound healing results, a thorough understanding of hydrogel production and the biochemical cues of the wound healing process is advantageous [26]. Because of its reversible phase transition properties, PNIPAM is frequently used to create hydrogel dressings that are sensitive to temperature changes. Furthermore, when the system changes from hydrophilic to hydrophobic and the PNIPAM solution changes from homogeneous to heterogeneous, microphase separation may take place; for this reason, the temperature is frequently referred to as the lower critical solution temperature (LCST). One of the most common synthetic polymers used as a cutaneous wound dressing is polyvinyl alcohol. For an improved blood clotting wound healing rate, PVA is typically mixed with additional substances or medications to create hydrogel dressings [27]. Because PVA hydrogels lack appropriate elasticity, membrane stiffness, and hydrophilic qualities, they are frequently combined with other polysaccharide-based hydrogels, including starch and alginate. Crosslinked polymers, either natural or synthetic, can be used to prepare hydrogel wound dressing films. Further, natural or synthetic ingredients, such as polyvinylpyrrolidine, polyvinyl alcohol, polyurethane, and poly(methacrylates), are used to make different kinds of hydrogel dressings [9, 28]. ABCHs are multipurpose substances used in drug delivery, tissue engineering, and wound healing. Based on their biodegradable and stimuli-responsive qualities, they provide controlled drug release, biocompatible scaffolding for tissue regeneration, and efficient wound dressings.

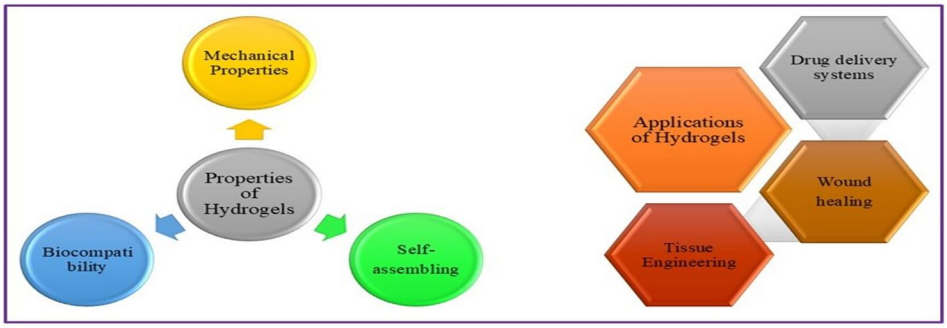

Figure 3. Structural properties and applications of hydrogels.

VI. CONCLUSION

In this chapter, the mechanical features of hydrogels that spontaneously swell are explained by their association of amphiphilic polymers. These hydrogels are utilized in the fields of pharmaceuticals, biocompatibility, and other environmental applications. The most suitable properties of hydrogels employed along with medicines, crosslinking mechanism, fabrication processes, design considerations of hydrogel engineering, and wound healing have been covered in this chapter, along with an overview of the many types of hydrogels on wound inflammation. Subsequently, it is essential to examine novel possibilities for amphiphilic block copolymers in order to broaden the range of hydrogel applications. Hydrogels made of hydrophilic and hydrophobic polymers have found extensive use in the biomedical field because of their good mechanical qualities, biocompatibility, and biodegradability. Hydrogels based on amphiphilic block copolymers are networks that have both hydrophilic and hydrophobic segments. These networks typically develop through effective free radical crosslinking polymerisation with hydrophilic and hydrophobic monomers. Utilised in drug delivery, tissue engineering, and wound healing, ABCHs are hydrogels composed of hydrophilic and hydrophobic monomers that have adjustable strength, compatibility, and self-assembly.

VII. ACKNOWLEDGEMENT

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A. Data Availability

Data will be made available on request.

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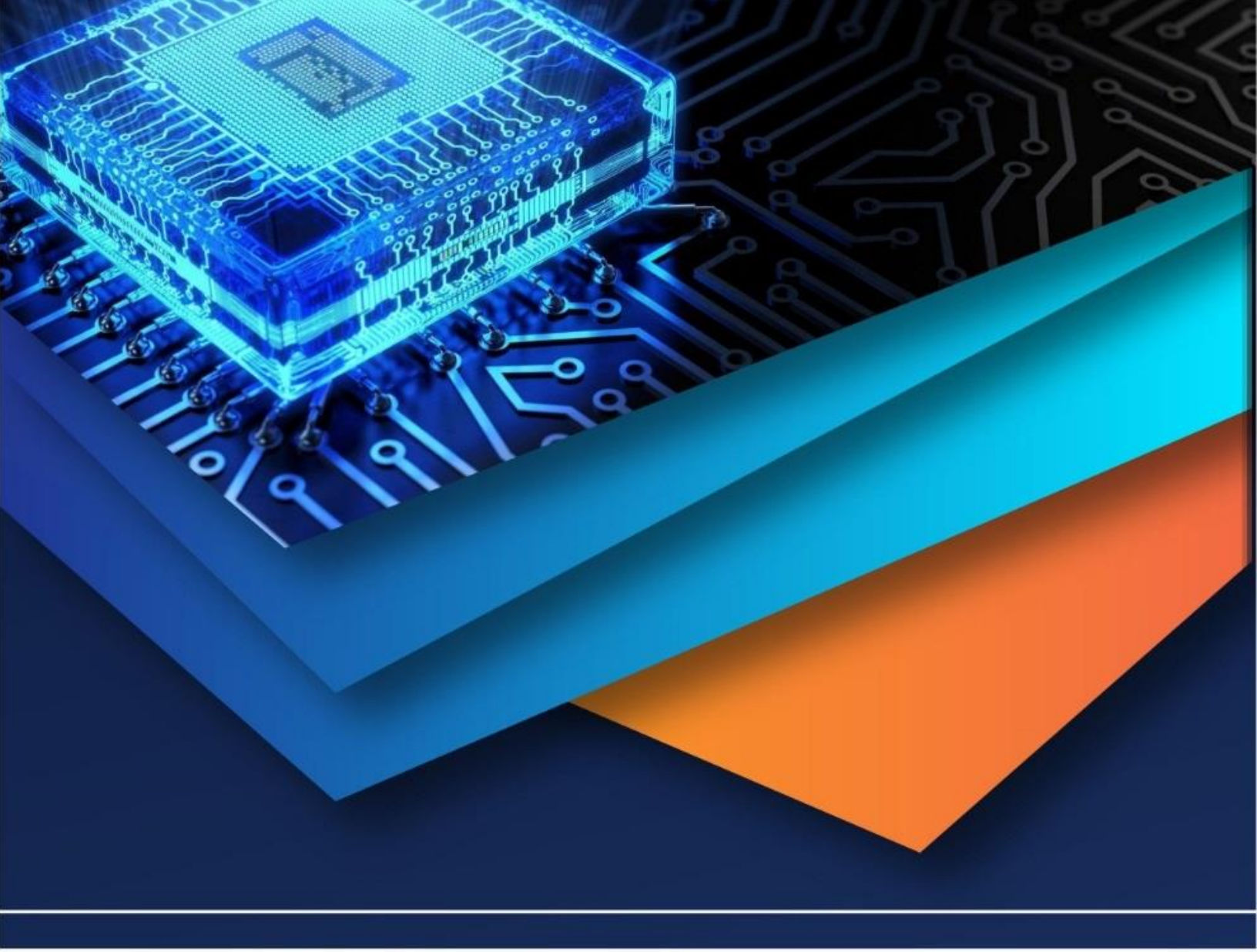

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