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# Some biomedical applications of Polyoxometalates: A review

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**Abstract -** *Polyoxometalates refer to a class of oligomeric cluster of transition metal oxoanion having well defined infra structure with a large variation in size, metal-oxygen framework, elemental composition and unmatched properties. They are considered as one of the most attractive and growing area of research and development especially in the field of catalysis, material chemistry and biochemistry. Along with many valuable properties like high biological activity and low cost feature, some polyoxometalates and their derivatives also show anti-bacterial, anti-viral, anti-tumor and anti-cancer activities. Few are observed to decrease the symptoms of diabetes. All these together accounts for the inexhaustible encouragement of researchers towards synthesis and study of polyoxometalates. This review is based on various scientific literatures and summarizes some bio-medical applications of polyoxometalates including their role in protein crystallization and in medicines.*

***Keywords: polyoxometalate, protein crystallization, anti-tumor, Alzheimer's disease, anti-viral.***

## 1. INTRODUCTION

Polyoxometalates (POMs) are distinct assembly of early transition metal (especially Mo, V and W) oxide with diverse structure and versatile properties, due to which they show potential applications in various domains such as catalysis [1], analytical chemistry, nanotechnology, chemical sensing, medicine [2] and material science[3]. Synthesis of POM clusters is very simple and requires a small number of steps or even just one step. Geometry, composition and charge of POM clusters can be changed through synthetic variables like concentration and type of metal oxoanion, presence of additional ligand, ionic strength, pH, type of heteroatom, reducing agent, temperature, methodology etc. Polyoxometalates show a large variety of shapes, elemental composition and size which may vary from small $[Mo_6O_{19}]^{n-}$[4] to nanosized species like $[H_xMo_{368}O_{1032}(H_2O)_{240}(SO_4)_{48}]^{48-}$ [5] . The ability of POM based hybrid materials to form species of size in the range of 10-100 nanometer, made them a connecting link between macro and nano world. The discrete polyanions of POMs are water soluble. They are non–sensitive to water and air. Also, they are more stable in aqueous acidic solution. This class of compound has the unique quality to undergo condensation reaction under aqueous acidic condition and the ability to gain and lose a specific number of electrons without changing the structural arrangement [6]. Lower charge density of POMs than the traditional anions (eg- $ClO_4^-$ , $NO_3^-$ ) makes them an ideal candidate for outer-sphere electron transfer reagent. There are two main types of POMs-isopolyoxoanions and heteropolyoxoanions with general formula $[M_aO_b]^{n-}$ and $[X_cM_aO_b]^{n-}$ respectively . Due to their easily modifiable [7] structure and electronic properties, heteropolyoxometalates are more studied and applied in the field of medicine than isopolyoxometalates. On the basis of various scientific literature published in different part of the world over several decades and extensive research on patent growth, following are the most challenging areas where they show strong impact.

## 2. IMPACT OF POM ON PROTEIN CRYSTALLIZATION

### 2.1 Protein Crystallization

The process of formation of an ideal protein crystal from a random state of its macromolecular sol is called protein crystallization. Quality of the crystal is minutely controlled by many extensive and intensive factors. Crystallization of protein is essential in order to study not only the 3D structure of protein but also to understand the functions of protein, as well as their interaction with drugs at a molecular level. Structure of protein also has direct impact on protein engineering and medicine design. Structural biology widely uses X-ray crystallography for studying 3D structure of biomolecules. Thanks to the fast technical progress in synchrotron facilities and development of software suites which helped in solving phase problem. Even then the task of applying X-ray crystallography on protein is not so easy because the major obstacle is to obtain a high quality single crystal. Protein crystallization is a time taking process because it is done by trial and error method. Also, success of the

process depends on many factors like concentration, pH, buffer, temperature etc. along with intensive nature of protein. Hence in this period of time biomolecular crystallization is a bottleneck in structural genomics and researchers have been putting a constant effort in this field. Observations show that rate of crystallization process can be increased by using some substances called additives. These additives form some cross- links between the repulsive amino acids and helps in their aggregation by decreasing repulsion between them. This brings some physical, chemical or conformational changes and promotes the building up of Bravais lattice.

### 2.2 Role of polyoxometalates in protein crystallization

Polyoxometalates have drawn the attention of researchers for protein crystallization after their use for the crystallization and structure clarification of ribosome, which was later awarded with the Nobel prize in 2009 [8].Success rate of protein crystallization depends directly on interaction between protein and additives used along with many other known and unknown variables. Selection of the correct additive with positive effect on crystallization process is again a tough and time taking task because of its empirical nature. POMs are chosen as an additive because of its negative charge due to which it has the ability to connect with +vely charged monomeric units. As a result of this, repulsion between them decreases. Hence rigidity increases which favours crystallization[9]. Recently co-crystallization of latent aurone synthase with $Na_6$ $[TeW_6O_{24}].22H_2O$[10] was done to get best diffracting crystal in order to elucidate its structure. Interaction between POM and amino acids may be (i)electrostatic attraction (arises due to opposite nature of charges of POM and amino acid) (ii) H-bonding[11]-[12] ,especially in case of amino acid with proton donar in the side chain (e.g.- arginine, histidine, lysine) (iii)covalent bond or (iv) van der waal forces [13]. However, on the basis of various experiments, it has been observed that the most common binding force is electrostatic attraction. Zhang et al [14] supported this by showing that the POM-protein interaction is generally exothermic(ΔH= -ve) in nature. Also, the interaction between amino acids and POM increases with increasing negative charge on POM. In case of lacunary POM, for example- ($\alpha_2$-$[P_2W_{17}O_{61}]^{10-}$) interaction with human serum albumin (HSA) probably occurs due to favourable entropy change and not due to enthalpy change which is +ve (endothermic process). Ability of POMs to form cross-links depends on many factors like its charge, size, shape, symmetry etc. POMs with higher charge and more internal symmetry have more binding ability. Further, denser crystal formation takes place if shape and size of POM is complementary with the protein monomeric units. On the basis of above deliberation the following conclusions can be drawn on the role of POMs as an additive (in protein crystallization)-

- They promote crystallization by stabilizing enzyme conformation.
- They block the movement of protein and thus rigidify them.
- They act as a connecting link between two similarly charged amino-acids, thus enhance crystal packing and stability.

## 3. MEDICINAL VALUE OF POLYOXOMETALATES

### 3.1 Inhibitory effect of POM on Alzheimer's disease

A neurodegenerative disorder called Alzheimer's disease is responsible for ageing, memory loss and cognitive decline. This disease results in gradual death of brain cells as a result of which a number of nerve cells and their connections[15] decreases and size of brain shrinks[16] .Danger of this disease increases with ageing and is becoming one of the major causes of death in older people in United States[17]. Ageing[18], family history[19] ,genetic factor , presence of certain genes like apolipo protein E (APOE) gene[20] are some unavoidable factors which increase the risk of this disease. It is believed that one of the several factors responsible for causing the Alzheimer's disease is the development of some tangles and plaques within and between the affected nerve cells. It has been observed that many tangles made up of tau protein are found within the neurons and a number of plaques made up of β-amyloid are formed between the dying nerve cells. Cholinesterase inhibitors, which include Tacrine(Cognex), Donepezil,Alantamine and Exelon ,as well as Memantine (Nemenda) are some drugs which may be used. At present, there is no way to prevent or slow Alzheimer's disease. Mechanism of Alzheimer's disease pathogenesis is very complex and has not been understood fully. However, on the basis of various observations it has been shown that formation of amyloid fibrils by the polymerisation of Aβ (amyloid β-peptides) is of extreme importance [21]-[23]. Along with their multidisciplinary properties and applications, some of the heteropolyoxometalate, specially POMs with Well-Dawson structure have been reported to have strong inhibitory effect on Aβ-aggregation. The binding between polyoxometalates

and Aβ is electrostatic attraction . It has been reported that POM and Aβ interaction and hence Aβ inhibition effect can be increased by using POM-Dawson derivatives(POMds) with different histidine chelating metals like Mn, Co, Fe, Ni, Cu etc. Thus Ni and Co metallated POM derivatives(POMds)can be applied for the treatment of this disease. Studies[24]-[25] show that formation of Aβ-aggregates decreases in the order of – POMds-Dawson-Ni > POMds-Dawson -Co > POM-Dawson Higher inhibitory rate of POMds-Dawson-Ni and POMds-Dawson-Co is probably due to their chelation effect . With increasing affinity between POMds and Aβ monomer, concentration of monomer decreases which retards the rate of aggregation. It has been observed that POMds-Dawson-Ni and POMds-Dawson- Co also inhibit the Aβ oligomer formation which is the main cause of cytotoxicity. They also depress the peroxidase like activity mediated by Aβ. This is why synthesis and screening of POMs with Aβ-aggregation inhibitory effect have drawn much attention as therapeutic and preventive agent of Alzheimer's disease.

### 3.2Inhibitory effect of polyoxometalates on microbial infection and tumor growth

Bacterial infections, viral infections and tumor growth are important health issues nowadays.

Table 1 shows therapeutic value of some polyoxometalates.

| Formula of POMs | Inhibitory activity |
|---|---|
| $K_5H_2[FeW_{11}TiO_{40}].17H_2O$ , $K_5H[H_2ZnW_{11}TiO_{40}].35H_2O$ [37] | Against Ova- induced lung inflammation |
| $K_7Na_3[Cu_4(H_2O)_2(PW_9O_{34})_2].20H_2O$ [38] | Inhibits viability of Osteosarcoma cell |
| $\{Ag_3(bpy)_6[PW_{12}O_{40}]\}$, $\{Ag_5(H_2biim)_2(HbiimNO_2)_2[PW_{12}O_{40}]\}$ and $\{Ag_7(pytz)_4[PW_{12}O_{40}]\}$ [39] | Anti-bacterial activity specially against gram –ve bacteria |
| $Cs_2K_4Na[SiW_9Nb_3O_{40}].H_2O$ [40] | Anti-HBV (Hepatitis B virus) infection |
| $[(Na)(Sb_3O_7)_2(SbW_7O_{24})_3]^{13-}$ [41] | Anti –HIV activity |
| $K_{27}[KAs_4W_{40}O_{140}]$ , $K_{18}[KSb_9W_{21}O_{86}]$ [42] | Against H. pylori (Helicobecter pylori) |

[bpy =2,2-bipyridine, $H_2$biim =2,2-biimidazole, pytz = 4-(1H-tetrazol-5-yl)pyridine ]

This urges the development of new drugs with more effective and low toxic effect. In the past few years, some researchers have reported that POM and their derivatives have come forward as strong candidate in the field of medicine for making alternative anti-tumor , anti -microbial and anti-viral drugs[26]-[35].Their low toxicity and high solubility in water and pH dependent stability and structure [7] make them an easy and ideal target for biochemical reaction. POM with different addenda atoms has different physiological effects. Some of them are used in making anti-viral drugs, some in anti-bacterial drugs while some show high activity in tumor growth inhibition. E.g.- polyoxovanadates show anti-bacterial and anti- tumor effect. They also interact with some proteins like myosine, actin, $Ca^{2+}$ ATPase and effect mitochondrial function [36].

#### 3.2.1 Anti-viral and anti-bacterial effect of Polyoxometalates

Nowadays, one of the major health related problems with constant concern is the development of drug resistant pathogens and microbes through gene mutation. POMs are important metal oxoanionic clusters with broad range of anti-microbial activity. Recent interest in POMs for the medical world [43]-[45] is due to their negative charge and large size which make them able to interact with cell surface by penetrating cell membrane except in some cases where they pass over the barrier and diffuse into the cell [46]-[47]. Recently anti-bacterial activity of thallium based POM $[Tl_2\{B\text{-}\beta\text{-}SiW_8O_{30}(OH)\}_2]^{12-}$ is studied [48]. An anti -bacterial capsule has been synthesized by the supra molecular assembly of polyoxomolybdate $[H_3PMo_{12}O_{40}]$,Chitosan and cetyltrimethylammonium bromide (CTAB). In past years, anti-HIV property has been reported for more than 50 polyoxometalates. Polytungstate like $[NMe_3H_7][CH_3C_5H_4TiP_2W_{17}O_{61}]$ and $Na_{16}[Mn_4(H_2O)_2(P_2W_{15}O_{56})_2]$ are active as anti-viral agents. Polyoxometalates show activity against both RNA as well as DNA viruses[49]-[52], like human immunodeficiency RNA virus (HIV),severe acute respiratory syndrome RNA virus(SARS),influenza RNA virus and herpes DNA simplex virus(HSV).

#### 3.2.2 Anti-tumor activity of Polyoxometalates

The process of formation of new blood vessel from the pre existing one is called angiogenesis. This involves migration, growth and differentiation of endothelial cell (lines the interior surface of blood vessel). It is essential for some normal physiological process like reproduction, growth and repair [53]. Angiogenesis

starts with the stimulation of some signalling molecules like vascular endothelial growth factor(VEGF),fibroblast growth factor etc., which then binds to the receptor present on the surface of endothelial cell .The process ends by some other chemical signal called angiogenic inhibitors .In a normal body, there is an intricate balance between stimulatory and inhibitory effect. Excessive angiogenesis is crucial for tumor growth because new blood vessel provides essential nutrients and oxygen to the developing tumors and cause the formation of metastases(new colonies of cancer cell) .Generally, serum of a cancer patient has a very high level of alkaline phosphatase(ALP) . Depending on the type of cancer, the main approved medicines with anti-angiogenic activity are bevacizumab[54], sunitinib malate and sorafenib. They work by blocking VEGF ligands or VEGF receptors(VEGFR) signaling pathways[55].However these VEGF agents can't be used for a long time due to serious side effects like hemorrhage, gastrointestinal perforation, hypertensive crisis, wound dehiscence etc. Another limitation includes their non – specific nature to the VEGFR. Cisplatin and gemcitabline are other effective and approved anti-tumour drugs but they are highly toxic. Studies[56] show that some polyoxometalates have inhibitory effects on tumor growth and angiogenesis.E.g.- decavanadate and other polyoxometalates have been assessed to have inhibitory effect on several alkaline phosphatase (more than 70% inhibition in case of intestinal ALP). This suppresses the abnormal cellular growth responsible for tumor formation. Apoptosis is an important and highly regulated biological process in a multi cellular organism which results in cell shrinkage , chromatin condensation ,nuclear fragmentation and ultimately cell death .Now it is already known that one of the major causes of tumor initiation is suppression in apoptosis due to some oncogenic mutation. Some polyoxometalates have the ability to induce cell apoptosis . So they could be an alternative and potential candidate for cancer therapy. But the major obstacle in the way of using hetropolyoxoanions for chemotherapy is their delayed excretion from body which may interfere with the metabolic process and in the long run may cause toxic effect .Recently [57] an approach has been made to develop organo -based polyoxometalate compound namely[$Mo_6O_{18}$ ($\equiv NC_6H_4$ -2-$CH_3$ -6-CON (Cy)-CO-NH-Cy]$^{2-}$ (POM-AMB-acy) by the synergistic effect between organic moiety and the lindqvist isopolyoxoanion which showed high activity against malignant glioma cell and also passed over the blood brain barrier(BBB). The compound is degradable and consisted of hexamolybdate moiety (POM) and N-acylureido group(acy) linked by 2 -amino -3 -methyl benzoxyl group (AMB).

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