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A Study on Removal of Arsenic from Industrial Waste Water

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Abstract: Arsenic removal from water has been receiving considerable attention in the field of water supply engineering. To develop the optimal coagulation protocol ,the effectiveness of several operation options such as coagulants, coagulant aids and additives ,as well as Adsorption. It is noted from this study that arsenic adsorption isotherm under water operation condition could be describe by Langmuir equation. An efficient flocs separation system subsequent to coagulation was essential to achieve the effectiveness of overall arsenic removal.

I.INTRODUCTION

Arsenic contamination in natural water is a worldwide problem because of its known toxicity and health hazards. The US Environmental Protection Agency (USEPA) has classified arsenic as a Class 'A' human carcinogenic and because of the serious health problems caused by arsenic in 2001, it reduced the maximum arsenic content in drinking water from 50 ppb to10 ppb[1]. Thus several technologies such as oxidation and filtration [2], A wide range of physical and chemical treatment methods have been applied for arsenic removal from contaminated waters fall into the following categories: (a) precipitation–coagulation processes, such as coagulation with iron or aluminum salts or Lime softening (b) membrane processes, such as reverse osmosis, ultrafiltration, Nano filtration, and electro dialysis. (c) adsorption processes, such as adsorption on activated or modified materials (d) ion exchange [3]. In these methods, the adsorption techniques are simple and convenient and easily available and have the potential for regeneration and sludge-free operation. Various adsorbents for arsenic removal have been developed that include such materials as activated carbon[4], agricultural products and by-products e.g. waste rice husk [5], eggshell [6] and shrimp shell [7], industrial by-products/wastes e.g. fly ash [8] and red mud [9]), oxides e.g. iron oxide [10], manganese oxides [11] and alumina [12], bio sorbents [13,14]

II. MATERIALS AND PROCEDURES

A. Arsenic Chemistry and Mobility

Arsenic occurs in Free State. Arsenic is situated in the 33rd spot on the periodic table, right next to Germanium and Selenium. Arsenic has been known for a very long time and the person who may have first isolated it is not known but credit generally is given to Albertus Magnus in about the year 1250. The element, which is classified as a metalloid, is named from the Latin arsenicum and Greek arsenic on which are both oxygen and chlorine to from inorganic pent avelent arsenate and trivalent arsenite. Arsenic can occurs in the environment in several oxidation (-3, 0, +3 and +5) as a sulphide (WHO 2008).

B. Reaction of Arsenic With Hydrogen

Arsenic trioxide can be reduced by hydrogen gas to form arsine. Arsine, AsH_3, is a very toxic gas that is denser than air. When arsine is heated, it breaks down to arsenic and hydrogen. Reaction of arsenic trioxide with hydrogen gas to form water and arsine.

$$As_2O_3(s) + 6H_2(g) \rightarrow 3H_2O(l) + 2AsH_3(g) \quad (1)$$

C. Reaction of arsenic with oxygen

Solid arsenic is oxidized when exposed to oxygen. The surface of the metalloid becomes black. When heated in oxygen gas, arsenic binds with oxygen to form tetra arsenic deca oxide, or arsenious oxide. Reaction of arsenic with oxygen gas to form arsenious oxide.

$$4As(s) + 5O_2(g) \rightarrow As_4O_{10}(s) \quad (2)$$

D. Reaction of Arsenic With Halogens

Arsenic reacts well with halogens with the exception of astatine. Arsenic penta fluoride is a highly toxic colorless gas. When liquid arsenic tri chloride is cooled with free chlorine molecule, arsenic penta chloride can be formed. Solid arsenic with bromine gas forms a light yellow solid called arsenic tri bromide. Reaction of arsenious oxide with hydrochloric acid to form water and arsenic tri chloride:

$As_4O6+12\ HCl\ (aq) \rightarrow 4AsCl_3\ (l) + 6H_2O\ (l)$ **(3)**

E. Reaction of Arsenic with Water

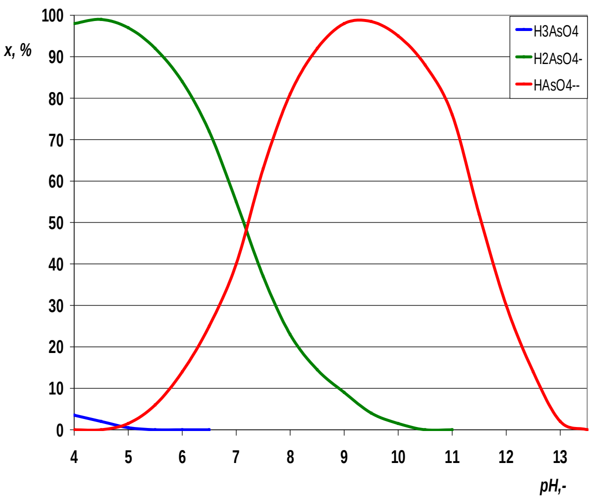
Pure arsenic is insoluble in water; however, many arsenic compounds dissolve easily in water. Arsenic in the form of arsenic tri-sulfide, and arsenic acid are examples of arsenic compounds that have found their ways into water supplies.

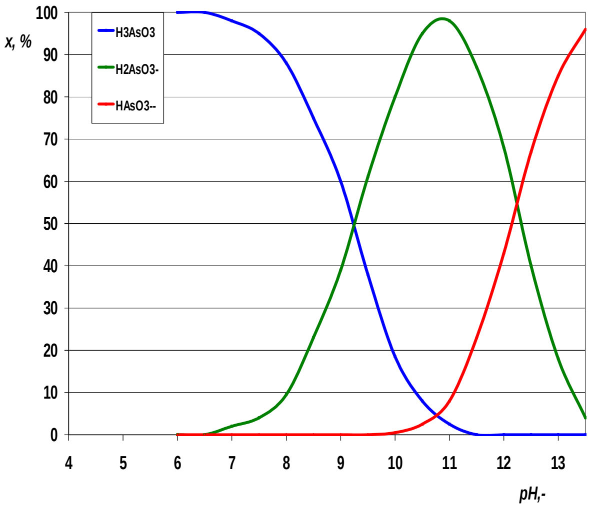
Reaction of arsenic tri sulfide with water to form hydrogen sulfide and arsenious acid:

$As_2S_3(s) + 6H_2O\ (l) \rightarrow 3H_2S\ (g) + 2H_3AsO_3\ (aq)$

F. Solubility Diagram of Arsenic (III) and Arsenic (V)

Arsenate exists in four form in aqueous solution based on pH, H_3AsO_4, H_2AsO_4, $HAsO^{2-}$, and $AsO_4^{3-.}$ Similarly, arsenite exist in five from: $H_4AsO_3^{+}$, H_3AsO_3, H_2AsO^{3-}, $HAsO_3^{2-}$, and AsO_3^{3-}.As (III) was found virtually independent of pH in the absence of other specifically adsorbed anion (smedley and Kinniburing, 2002). More trivalent arsenic is found in reducing groundwater condition than pentavalent arsenic, As (III) exist a non-dissociated at neutral and slightly acidic conditions and only at pH > 8 considerable amount of anionic species are found. As (V) is completely dissociated and present in the form of monovalent, divalent, trivalent anion (Ali and Aboul- Enien).





This Figure, contains solubility diagrams for As(III) and As(V), ionic forms of arsenate dominate at pH >3, while arsenite is neutral at pH <9 and ionic at pH >9.

G. Natural And Manmade Source Of Arsenic

Arsenic is found in earth' crust most commonly in the form of FeAsS or iron arsenide sulphide(EHC).Arsenic is associated with graphite and sulphide rich metal sedimentary rock and high arsenic concentration occurs in area of sulphide mineralization._It has been estimated that about one-third of the atmospheric flux of arsenic is of natural origin.

Volcanic action is the most important natural source of arsenic, followed by low-temperature volatilization. Inorganic arsenic of geological origin is found in groundwater used as drinking-water in several parts of the world, Organic arsenic compounds such as arsenobetaine, arsenocholine, tetramethylarsonium salts, arsenosugars and arsenic-containing lipids are mainly found in marine organisms although some of these compounds have also been found in terrestrial species. The man made source of arsenic is released by semiconductor, and metallurgical industries and when Elemental arsenic is produced by reduction of arsenic trioxide (As_2O_3) with charcoal. As_2O_3 is produced as a by-product of metal smelting operations. Mining, and smelting of non-ferrous metal and burning of fossil fuel are the major industrial process that contributed to anthropogenic arsenic contamination of air water soil. The normal level of arsenic which is found in soil is about 5-6 mg/kg (Lioa et.al 2005). While the average toxic level of arsenic is reported at 40 mg/kg dry soil (sheppard 1992),arsenic in soil exists mostly as inorganic from while organic from is occasionally found.

III. CONVENTIONAL METHODS OF ARSENIC REMOVAL

There are many methods of arsenic removal such as adsorption, membrane separation, precipitation, lime softening, ion exchange and coagulation or flocculation.

A. *Adsorption*

Adsorption is the phenomenon in which molecule of gas, liquid or solid become attached to the solid surface of atoms, ions, or molecules from a gas, liquid, or dissolved solid to a surface. (I" Glossary). Intermolecular forces attract the molecules, causing them to cling to the surface. Adsorption process is operative in most natural physical, chemical and biological system. Adsorption capacity depends on the adsorbent properties, adsorbate chemical properties, temperature, pH, doges of adsorbent, and size of the adsorbent etc.

Adsorption process is leading to find improved and tailor-made material, which will meet several requirements such as regeneration capability, easy availability, and cost effectiveness etc. Numerous low cast adsorbents have so for studies for the removal of arsenic from water and wastewater. Coconut shell, coconut husk, dry plant, different type of mud, many variety of tree bark, fly ash many type of bio-adsorbent only few listed. Removal of heavy metals by these materials might be attributed to their protein, Carbohydrates and phenolic compound contents.

Which have metal-binding functional such as carboxyl, hydroxyl, Sulphate, arsenate and amino group (Mohan pitman2007a).In this technique the active surface of the adsorbent, its surfaces energy and the pH of the solution highly influence the removal efficiencies. Adsorption may be classified two types; first we explained depending upon the concentration of adsorbent, concentration adsorbent depending upon positive adsorption and negative adsorption, positive adsorption might be defined if the concentration is more on the surface as compared to its concentration on bulk phase. i.e. (when a concentrated solution of KCL is shaken with blood charcoal).Negative adsorption may be defined if the concentration is less on the surface as compared to its concentration in bulk phase. i.e. (When dilute solution of KCL is shaken with blood charcoal).Second type may be defined depending upon the nature of force existing between adsorbate and adsorbent, existing forces defined Vander walls forces and chemical forces.

1) *Factor Affecting the Adsorption*

- *a)* Nature of adsorbate and adsorbent.
- *b)* The Surface area of adsorbent.
- *c)* Activation of adsorbent.
- *d)* Experimental Condition; temperature, pressure, pH, dosage, time etc.

pH is the most important parameter in the adsorption process for arsenic removal. The initial pH of adsorption process is related to the adsorption mechanism on to the adsorbent surface from water and reflects the nature of the physicochemical interaction of species in solution and the adsorptive sites of adsorbent. (Chiban et al., 2011b) it will also adsorb the observed the adsorption capacity increased with increasing the contact time until equilibrium time was reached. Adsorption process is generally exothermic in nature and the extant and rate of adsorption in most of the cases decreases with increasing temperature. Various adsorption studies frequently investigated in the literature when temperature increased the adsorption capacity increased with temperature, due to an increase in number of adsorption site of the adsorbent, the process was claimed to be endothermic.

To optimize the design of an adsorption system for the adsorption of adsorbates, it is important to establish the relationship between equilibrium capacity and equilibrium concentration. The overall adsorption process can be modeled by monocomponent and multicomponent isotherm. Various isotherm equation have been used described the equilibrium characteristics of adsorption. Various isotherm equations like those of Frenudlich, Langmuir,

B. *Langmuir Isotherms*

Langmuir theory (I. Langmuir,) the adsorption of gases on plane surface glass, mica and platinum, J.AmChem.Soc.40 (9) (1918)1361-1404.

The adsorption takes place at specific homogenous sites within the adsorbent,that adsorption is a first order chemical process and monolayer of adsorbed material is formed onto series of distinct sites on the surface of adsorbent. (Monda et.al 2009 AIche).

$$Qe = {}^{q_{max}K_LC_e}/_{(1+K_LC_e)} \qquad (4)$$

Where q_{max} (mg/g) and K_L (l/mg) are the Langmuir constants related to the capacity and energy of Adsorption.

C. Extended Version Of Langmuir Isotherm

Let's we assume the surface site are uniform, and that all the adsorb ate molecules (ions) in the solution compete for the same surface sites, (R.T.yang gas sepration by adsorption process,butter worths,bosteon,MA,1987) Langmuir equation for multicomponent system as.

$$q_{e,i} = q_{max} K_i C_{e,i} \Big/ 1 + \sum_{j=1}^{N} K_j C_{e,j} \qquad (5)$$

The value of q_{max} and K_i obtained from optimizing fitting the above equation.

D. Freundlich Isotherm

Freundlich Isotherm developed on the basis of the formation monolayer due to adsorption on to rough heterogeneous surface and degree of favorability of adsorption.

$$q_e = K_f C_e^{1/n} \qquad (6)$$

Where K_f ((mg/g)(/mg/g)$^{1/n}$ is called the adsorption capacity and 1/n define the degree of favorability of adsorption.

E. Extended Version of Freundlich Isotherm

The Freundlich isotherm from binery mixture can also called Extended version of Freundlich isotherm as given below(W.Fritz,E.U. Schluender, Simultaneous adsorption equilibria of organic solutesin dilute aquesesolutionon activate carbon, chem.Eng.Sci. 29(1974)1279-1282.

$$q_{e_1} = K_{F,1} C_{e,1}^{n_1+x_1} \Big/ C_{e,1}^{x1} + y_1 C_{e,1}^{z1} \qquad (7)$$

$$q_{e_2} = K_{F,2} C_{e,2}^{n_2+x_2} \Big/ C_{e,2}^{x2} + y_2 C_{e,1}^{Z2} \qquad (8)$$

Where $K_{F,1}$, $K_{F,2}$, and n_1 and n_2 can be estimated from the corresponding individual Freundlich isotherm equation and the other six parameter(x_1; y_1;$z_1$and x_2; y_2;z_2) are the multi-component Freundlich adsorption constant for the second components.(G.Mckay, B. AlDuri, Preduction of multicomponent adsorption equilibrium data using empirical corelations Chem.Eng.J.41(1989) 9-23 of parameter in the isotherm equation.

F. Adsorption Kinetic Model

Different type of kinetic model, such as pseudo-first order, pseudo-second order, Weber and Morris, Bangham, Elovich and inter particle diffusion mode, are generally used to describe the non-equilibrium stage of equilibrium(shashi.et.al 2013)Original forms of these model describe below.

G. Pseudo-First Order Model

Pseudo first order model describe the rate of adsorption of pollutants on the adsorbent, the pseudo first order equation is

$$\frac{dy}{dx} = k_f(q_e - q_t) \log(q_e - q_t) = \log q_e - \frac{k_f}{2.303} t \qquad (9)$$

Where K_f is the pseudo first order rate constant (min^{-1}), q_t is the amount of adsorb ate adsorbed at a time t(mg/g), $q_e$is the adsorption capacity in equilibrium (mg/g), t is the contact time.

H. Pseudo- second Order

This pseudo-second order model represented (Ho and McKay, 1999),

$$\frac{dy}{dx} = k_s(q_e - q_t)^2 \frac{t}{q_t} = \frac{1}{k_s q_e^2} + \frac{1}{q_e} \qquad (10)$$

Where k_s are pseudo second order constant (g/mgs); at t→0 $K_s q_e^2$ represented the rate of adsorption (mg/gs).The main difference between a First Order Reaction & a Pseudo First Order reaction is that the pseudo first order reaction is a reaction that should actually happen by some higher order (more than 1) but occurs by first order because the concentration of one reactant used is very high & another reactant used is very low. The concentration of the reactant which is very low can be neglected& thus the concentration of the reactant which has higher concentration is alone taken.

I. *Thermodynamic Parameter*

The influence of temperature on the sorption of arsenic it was observed that the rise in temperature increase the adsorption capacity on the surface of adsorbent. With increase in temperature the mobility of arsenic ions increase and the retarding forces acting on the diffusing ions decrease, thereby increase the sportive capacity of adsorbent. (V.C.et.al (2005)]. The sorption at higher temperature indicates the endothermic adsorption process. Show in earlier the increase in sorption capacity with an increase in temperature may be attributed to chemisorption. Various thermodynamic quantities namely heat of adsorption (ΔH), change in entropy(ΔS), and Gibbs free energy (ΔG) were computed from the equilibrium constant K_D values.

J. *Estimation of Thermodynamic Parameter*

The Gibbs free energy change of the adsorption process is related to the equilibrium constant by the classical van't Hoff equation:

$$\Delta G_0 = -RT\ln K_D \qquad (11)$$

The Gibbs free energy changes are also related to the entropy change and heat of adsorption at constant temperature

$$\Delta G_0 = \Delta H - T\Delta S_0 \qquad (12)$$

Combining the above two equation, one gets:

$$lnk_D = \frac{-\Delta G}{RT} = \frac{\Delta S_0}{T} - \frac{\Delta H_0}{R} \qquad (13)$$

Where ΔG_0 is the free energy change in (KJ/mole), ΔH_0 is the change in enthalpy (KJ/mole), ΔS_0 entropy change (KJ/mole K), T is the absolute temperature (K), R is the universal gas constant and $K_D = \frac{Q_e}{C_e}$ is the single point or linear sorption distribution coefficient. Thus the,ΔH_0 is the enthalpy change (KJ/mol) which can be determined from the slop of liner Van't Hoff plot, i.e. ln K_D verses (1/T), using the equation:

$$\Delta H_0 = \left[R \frac{dlnK_D}{d(1/T)}\right] \qquad (14)$$

This ΔH_0 corresponds to the isosteric heat of adsorption ($\Delta H_{st,\,0}$) with zero surface coverage. (Suzuki. M, 1982)

K. *Ion Exchange*

In this method arsenic ions held electro statically on the surface of strong base anion exchange. Resins are exchange for ions similar charge in the solution from the resin. When arsenic dissolves in water they typically form ions. Ions are electrically charged portions of a compound. There is a balance of positively and negatively charged ions in natural waters. When arsenic is dissolved in water, the water is typically crystal clear. If the water is cloudy or otherwise discolored it is likely that some, or all of the contaminants are in a solid form. Solid particles are not intended to be removed by ion exchange and solids will clog the treatment media. The electrical charge on an ion can be either positive (+) or negative (-). Valence is the term that describes the category of the electrical charge on a dissolved ion such as positive or negative. If the contaminant has a positive charge, it would be called a cation, and would be removed by use of an IE media called cation exchange resin. If the contaminant has a negative charge, it would be called an anion, and the appropriate treatment media would be called an anion exchange resin. For arsenic removal, an ion exchange resin, usually loaded with chloride ions on "exchange site" is placed in vessel. The arsenic containing water is passed through the vessel and the arsenic "exchanges" for the chloride ions. The water exiting in the vessel lower in arsenic but higher in chloride than the water entering the vessel. Eventually, the resin becomes "exhausted"; that is, all or most of the "exchange sites" that's were loaded with arsenic or other anion.(Jacksona and miller 2000).The factor affecting the use of the ion exchange process include contact time and spent regenerate disposal.

L. *Membrane Filtration*

Membrane filtration covers all engineering approaches for the transport of substances between two fractions with the help of permeable membranes. In general, mechanical separation processes for separating gaseous or liquid streams use membrane technology. Some synthetic membrane is permeable to certain dissolved compound but exclude others. Because of their selective permeability for different compound, such membrane can act as a molecular filter to remove dissolved arsenic, along with many other dissolved and particulate compounds. Pressure differences are the driving force for separation. (Mohamed chiban 2013). The removal efficiency of membrane filtration depend on the pore size in the membrane and particle size of the of the arsenic species. As advancements are made in membrane production and module design, capital and operating costs continue to decline. The membrane processes discussed here are four types Microfiltration (MF), Ultrafiltration (UF), Nano filtration (NF), and reverse Osmosis (RO).Generally all the membrane material is manufactured from a synthetic polymer, although other forms, including ceramic and metallic “membranes,” may be available. Generally all membranes manufactured for drinking water are made of polymeric material, since they are significantly less expensive than membranes constructed of other materials.

M. Lime Softening

It is similar to precipitation process where limes (lime, lime-soda ash, or caustic soda) for used removal of any heavy metals. In this process the pH is sufficiently raised by 1N of sodium hydroxide and if required to used magnesium hydroxide. Generally we know that the pH of water lie between6.5 to 7. In lime softening process to raise the pH approximately 10 to11. Some advantages of lime softening process remove dissolved minerals and scale forming tendencies, household cleaning agent, removing arsenic, radium and uranium, certain organic compound and total organic carbon (TOC).

N. Electro-Coagulation and Flougulation

Electrocoagulation is the sewage treatment technique of gives the promising result EC understandably has several similarities with the chemical coagulation but also significant differences, such as side reactions.(I. Heidmann, W.Calmano, Removal of Zn(II), Cu(II), Ni(II), Ag(I) and Cr(VI) present in aqueous solutions by aluminum electrocoagulation, J.Hazard. Mater.152(2008)934–941). In electro- coagulation multiple reaction occurring simultaneous anode and cathode, Aluminum is more suitable electrode material for electro coagulation. The resulting metal concentration is the main parameter affecting destabilization of pollutants .The effect of the main parameters- current density, initial pH, and electric charge per volume, temperature and electrolysis cell.

M. Yan, D. Wang, J. Qu, J. Ni, C.W.K. Chow, Enhanced coagulation for highalkalinity and micro-polluted water: The third way through coagulant optimization, Water Res. 42 (2008) 2278–2286.

The amount of metal cation dissolved during the reactions at the anode can be calculated according to Faraday’s law:

$$m = \frac{ItM_w}{zF}$$

Where I is the current, t is the time in (s), Mw is molecular weight of the substance (g/mol), F is Faraday’s constant (96485 C/mol), z is the number of electrons involved in the reaction (2 for Fe2+ and 3 for Fe3+ and Al3+) and m is the quantity of metal dissolved (g). Several studies have reported current yields higher than 100% for the dissolving of electrodes.

IV. CONCLUSION

In present review,we tried to elaborate on different natural and man made sources of As in water .In order to combat Arsenic problem,various remediation methods based on conventional and modern technology for removal of As in several parts of the world have been critically reviewed.Most of the existing technologies for removal of As involve the direct removal of As^5 or converting As^3 to As^5 .The awareness of population is deemed equally important in maintaining in choosing mitigation.

A. Abbreviation

1) As: Arsenic
2) As$^{3:}$ Arsenite
3) As$^{5:}$ Arsenate

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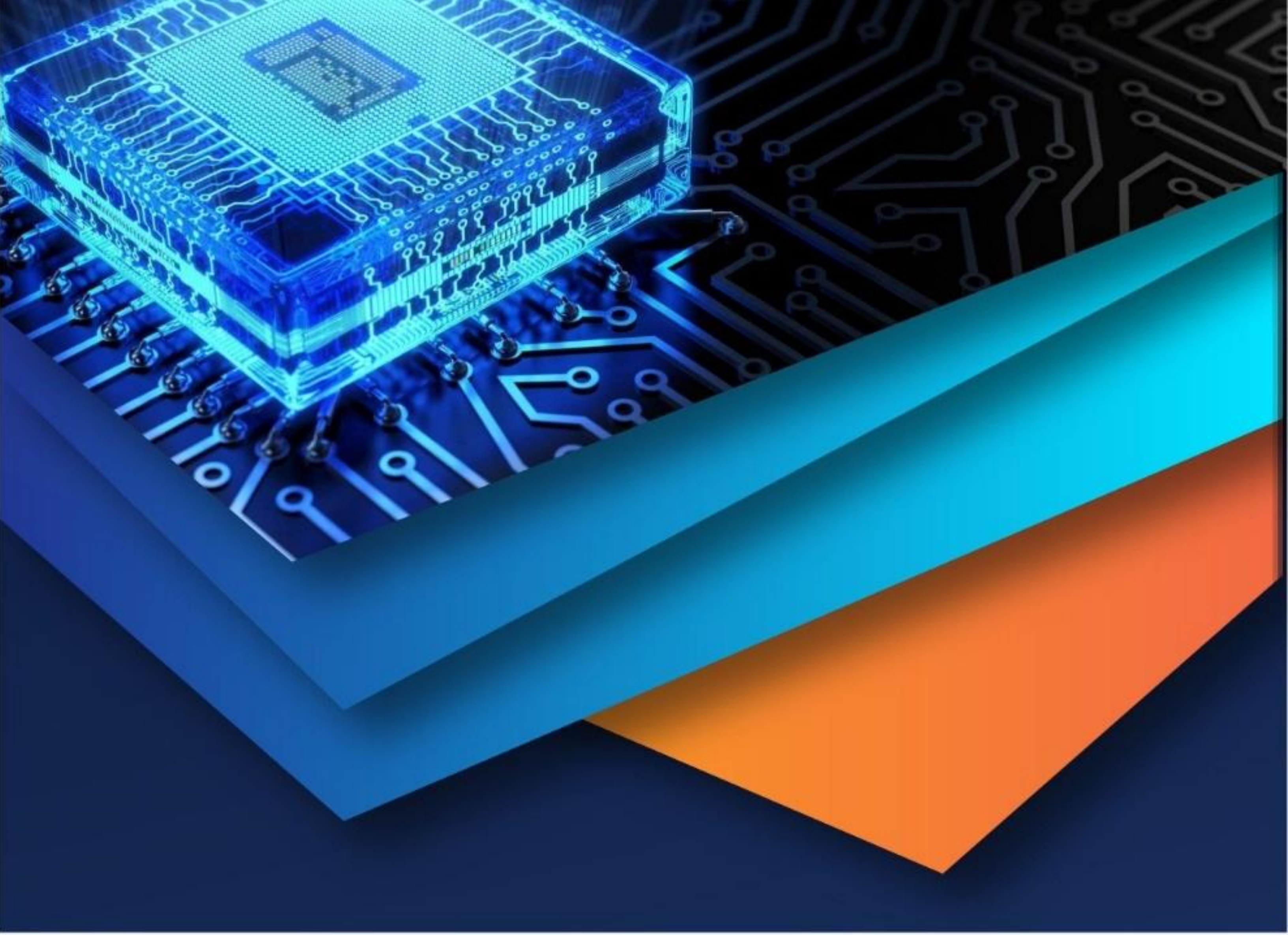

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