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Estimation of Radon Mass Exhalation Rate and Radium Content in Soil Samples Collected from Kolasib District of Mizoram, India

Vanramlawma[1], Hmingchungnunga[2], B. Zoliana[3], L. Z. Chhangte[4], B. K. Sahoo[5], B. K. Sapra[6], Z. Pachuau[7]
[1, 2]*Department of Physics, Mizoram University,Aizawl-796004, Mizoram,India.*
[3, 4]*Department of Physics, Govt. Zirtiri Residential Science College, Aizawl-796001, Mizoram,India.*
[5, 6]*Radiological Physics & Advisory Division, Bhabha Atomic Research Centre (BARC), Mumbai–400094,India.*

Abstract: ***Scintillation-based detector Smart RnDuo have been used for the measurement of radon mass exhalation rate in soil samples collected from Kolasib districts of Mizoram, which is located in the North-Eastern region of India. The state of Mizoram is considered to be one of the highest age adjusted lung cancer incidence rate area in India. The radon mass exhalation rate in these samples has been found to vary from 0.92 mBq/kg/hr to 51.97 mBq/kg/hr, with an average value of 26.57 mBq/kg/hr. For measurement of the radium activity concentration in the collected soil samples, 5"X 4" NaI (Tl) gamma spectrometer was employed. It was found that the activity concentration of radium obtained in this study ranges from 12.90 Bq/kg to 43.39 Bq/kg, with an average value of 26.84 Bq/kg. From the data obtained, the radon emanation factor was also calculated. Correlation between radium activity concentration and radon mass exhalation rates obtained in this study shows that they have a positive correlation.***

Keywords: Radionuclides, Radon mass exhalation rate, RnDuo, soil

I. INTRODUCTION

Primordial radionuclides such as 238U and 232Th are found in traces almost in every soil, rocks and water on earth. They usually have a very long half-life and continuously decay into another radioactive element, hence forming a decay chain. Primordial radionuclides like 238U, 232Th and 40K and their corresponding daughter elements are the main source of gamma radiations on earth[1], and their presence in the soil could lead us to identify their origin as well as abundance of their daughter elements like radon, thoron and their progenies.

Radon, a decay product of 238U, is a highly radioactive, colourless, chemically inert gas and is one of the main sources of radiation exposure to human beings[2]. Of the total radiation received by humans, the contribution of radon and its decay products is 51% through inhalation and 0.21% through ingestion[3-4]. Due to its short half-life, radon continuously disintegrates into its progeny, thereby releasing alpha particles. The three main progenies of radon, namely, polonium, lead and bismuth have extremely short half-life and this makes radon extremely hazardous [5-6].

Radon is being continuously formed in the soil and released in air. Radon formed in the soil enters into the pore spaces of soil grains by a process called emanation and then escapes into the environment by a process known as exhalation[7-8]. Exhalation mainly depends on the type of soil, its grain size distribution, porosity and the water content of the soil. Radon and its isotopes have been identified as the second leading cause of lung cancer after tobacco smoking [9-10]. Hence, estimation of radon mass exhalation rate is important to give a useful reference against radiation safety measures and to create a baseline for further studies.

II. STUDY AREA

Figure 1 show the geographical sites where the soil samples were collected. The sampling area extends from from 23˚58'56.5'' to 24˚30'43.5'' latitude and from 92˚35'54.4'' to 92˚46'23.7'' longitude in Kolasib district of Mizoram, India. The soil samples were collected from 27 different locations within the study area. As Mizoram has one of the highest age rate for lung cancer incidence in males and females amongst all sites of India[11], it is important to study of the concentration of carcinogenic elements like radon and its parent element ^{226}Ra in this area.

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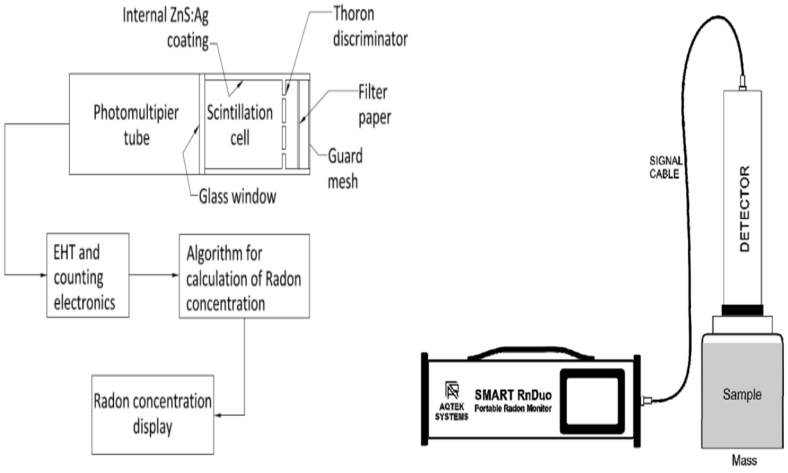
Fig. 1: Geographical Map of the studied region of Kolasib district, Mizoram.

III. METHODOLOGY

A. Measurement of Radon Mass Exhalation Rate

The measurement of radon mass exhalation rate in collected soil samples was carried out by using a scintillation based radon monitor Smart RnDuo. RnDuo is an advanced portable continuous radon /thoron monitor, whose detection principle is based on detection of alpha emitted from radon and its decay products formed inside a scintillation cell volume, and has multiple applications in radon and thoron studies [12]. The instrument consists of a progeny filter and a thoron discriminator which eliminates radon/thoron progenies and thoron from entering the scintillation chamber.

The alpha scintillations from radon and its decay products formed inside the cell are continuously counted by the PMT and the associated counting electronics. The instrument has an in-built algorithm to continuously measure radon concentration. The measured radon concentration is then processed by a microprocessor unit as per the developed algorithm to display the concentration of radon (Fig. 2).



(a) (b)

Fig. 2: (a) Schematics of radon measurement process in RnDuo.(b) Measurement of radon mass exhalation from soil.

For measuring radon mass exhalation rate, the sample was first weigh and its volume taken, and then put inside the radon mass exhalation chamber. The detector is then mounted on top of the exhalation chamber (Fig. 2b), and build up data of radon was retrieved every 60 minutes for a period of about 10- 24 hours. Least square fitting of the data obtained was carried out using the equation [13]:

$$C(t) = \left(\frac{J_m M}{V}\right) t + C_o \quad (1)$$

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Where C (t) is ^{222}Rn concentration (Bq m^{-3}) at time t, C_o is the ^{222}Rn concentration (Bq m^{-3}) present in the chamber volume at t = 0, M is the total mass of the dry sample (kg), V is the effective volume (volume of detector + porous volume of sample + residual air volume of mass exhalation chamber) (m^3).

The porous volume (V_p) can be estimated using the following equation

$$V_P = V_s - \left(\frac{M}{\rho_g} \right) \quad (2)$$

Where V_s is the sample volume in the mass exhalation chamber, ρ_g is the specific gravity of the sample and t is the measurement time (h).

On least square fitting of the data to the above equation, one may obtain J_M from the fitted parameters with the information of the mass M of the sample.

B. Measurement of Natural Radioactivity Using NaI (Tl) Detector

The measurement of activity concentration of ^{228}Ra in the soil samples collected was carried out by using Thallium (Tl) activated 5" X 4" Sodium Iodide (NaI) detector enclosed in a cylindrical lead and iron shield. The detector was coupled with a PC based multi-channel analyzer, GSPEC-SA (Ver. 2.5X). For efficiency calibration, standard source of ^{238}U (provided by BARC, Mumbai) was analyzed for a period of 10800 sec (3 hours). From the gamma energy peak obtained, the efficiency was calculated using the formula:

$$\eta(\%) = \frac{Area / \sec}{dps} \times \frac{100}{Ab\%} \times 100 \quad (3)$$

Where, $\eta(\%)$ = Percent Efficiency

Area/sec = Net peak area per second (background subtracted)

dps = Source strength

Ab% = Gamma ray abundance factor

Before the start of the experiment, the collected samples were dried to a temperature of about 110^{o}C and were then grinded and sieved using a 500 μm mesh. Each sample were then sealed inside an air-tight container of 250 ml and was kept undisturbed for a minimum period of 30 days to attain radioactive equilibrium.

Before measurement, a three-point energy calibration was carried out using sources containing a mixture of ^{60}Co and ^{137}Cs sources[14]. The energy calibration allows the establishment of the relationship between the channel numbers of the analyzer and the known energy of the photons [15].

The activity concentration of ^{226}Ra for each samples were then obtained by analyzing with GSPEC-SA Multichannel Analyzer for a period of 50,000 seconds. The weights of the samples were measured and the activity concentration of radium (C_{Ra}) was obtained (in Bq/kg) for each samples using the formula:

$$C_{Ra} = \frac{N}{T} \times \frac{100}{\gamma\%} \times \frac{100}{\eta\%} \times \frac{1}{W_t} \quad (4)$$

Where, $\frac{N}{T}$ = Background subtracted net photo peak counts in time 'T'.

γ = abundance of gamma ray under consideration.

η = absolute detection efficiency obtained from the energy efficiency calibration.

Wt = weight of the sample

C. Emanation Factor

The fraction of radon atoms generated that escape the solid phase in which they are formed and become free to migrate through the bulk medium is defined as the emanation factor of radon. The emanation factor (*f*) can be calculated using the formula [16]:

$$f = \frac{Jm}{CRa}\lambda \quad (5)$$

C_{Ra} - Radium activity concentration (Bq/kg)

λ – Decay constant of radon (in hr.)

J_m – Mass exhalation rate of radon (mBq/kg/hr)

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IV. RESULTS AND DISCUSSIONS

Table 1 gives the results of measurement of radon mass exhalation rates, radium activity concentration and radon emanation factor in soil samples collected from a total of 27 locations within the specified study area. It was found that the measured radium activity concentration obtained in this report was found to be in the range of 12.90 Bq/kg to 43.39 Bq/kg, having an average value of 26.84 Bq/kg. The radium activity concentration in all the sample were found to be quite comparable with the worldwide average value of 35 Bq/kg, as proposed by UNSCEAR[2]. Also, they were much lower than the critical value of 1000 Bq/kg set by IAEA[17].

The radon mass exhalaltion was also measured and was found to be in the range of 0.92 mBq/kg/hr to 51.97 mBq/kg/hr, with an average value of 26.57 mBq/kg/hr. These results were quite comparable to the results obtained [18] in other districts of Mizoram. From the measured values of radium activity concentration and radon mass exhalation rates, the radon emanation factor was calculated and was found to be in the range of 0.002 to 0.25, with an average of 0.13. The radon emanation factor usually ranges from about 0.05 to 0.70 in typical rocks and soils [19], and the result obtained in this report were quite comparable to the given range.

Figue 3 shows the correlation between the measured values of radium activity concentration and the radon mass exhalation rate in the collected samples. The correlation coefficient was obtained by plotting the radium activity concentration along X- axis and the radon mass exhalation rate along Y-axis, and the correlation coefficient was found to be R^2= 0.309. This result shows that there is a positive correlation between the two parameters, although the correlation is not quite strong. Although the radon mass exhalation rate depends on the radium activity concentration[20], the possible reason for this weak correlation may be because of other factors such as soil moisture content, porosity, grain size distribution etc, which radon mass exhalation is also dependent upon.

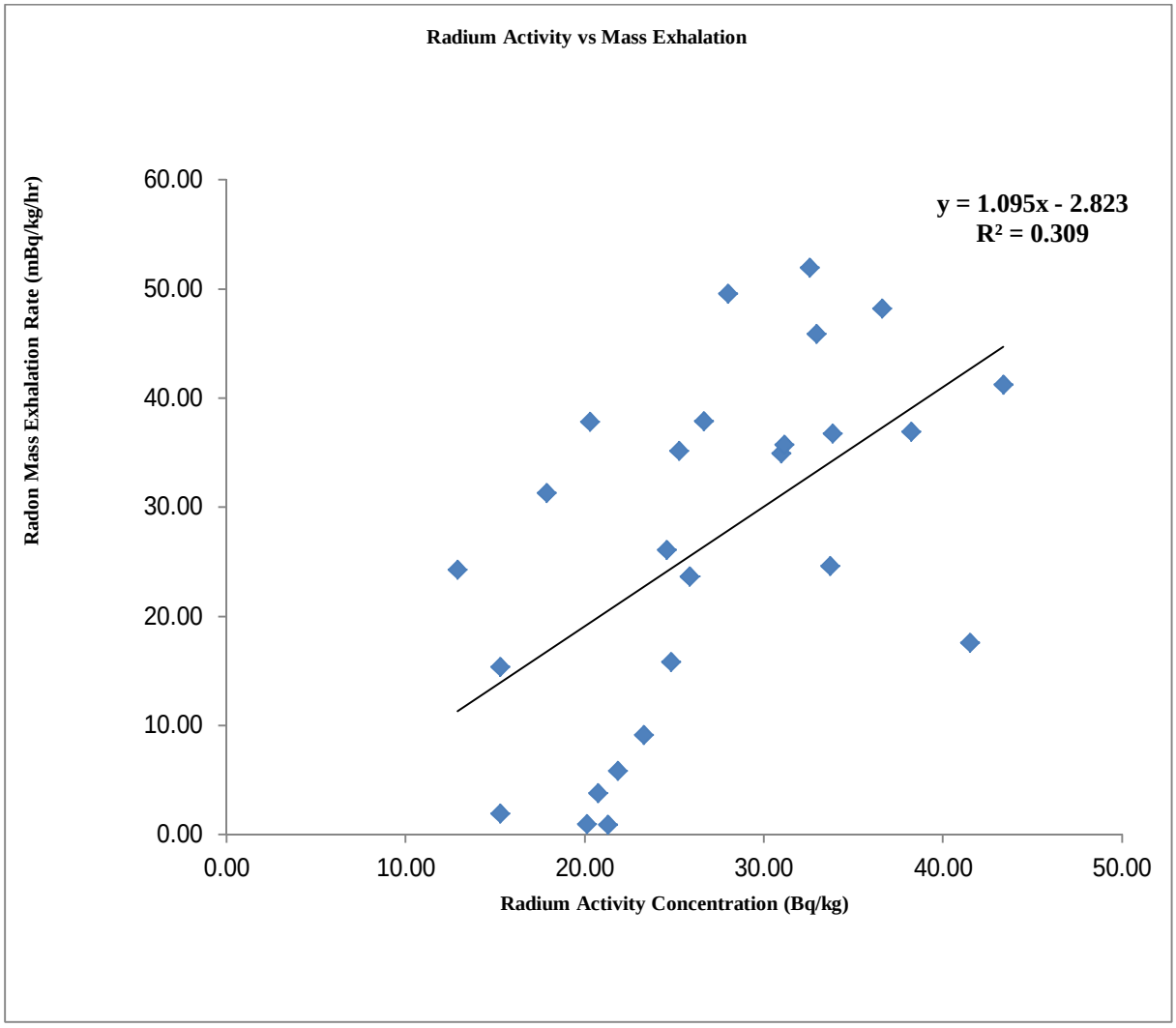


Fig. 3: Correlation between Radium Activity Concentration and Radon Mass Exhalation Rate.

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Table 1: Measurement of radon mass exhalation rate, radium activity concentration and emanation factor of radon in soil samples collected

Sl. No.	Location Codes	Radium Activity Concentration (C_{Ra}) (Bq/kg)	Radium Activity Concentration (C_{Ra}) in (mBq/kg)	Mass Exhalation Rate (J_m) (mBq/kg/hr)	Emanation Factor (f)
1	K-1	43.39	43386.40	41.24	0.13
2	K-2	24.58	24578.34	26.09	0.14
3	K-3	20.28	20275.27	37.86	0.25
4	K-4	20.73	20728.21	3.81	0.02
5	K-5	30.97	30965.29	34.94	0.15
6	K-6	28.00	28003.76	49.56	0.24
7	K-7	24.83	24825.05	15.87	0.09
8	K-8	25.83	25829.95	23.67	0.12
9	K-9	25.83	25829.95	23.67	0.12
10	K-10	23.28	23284.73	9.16	0.05
11	K-11	31.13	31132.96	35.76	0.15
12	K-12	25.27	25272.67	35.18	0.19
13	K-13	36.60	36602.60	48.24	0.18
14	K-14	32.92	32920.78	45.91	0.19
15	K-15	12.90	12899.49	24.27	0.25
16	K-16	15.28	15275.39	15.37	0.14
17	K-17	20.10	20101.04	0.99	0.01
18	K-18	15.28	15280.87	1.93	0.02
19	K-19	32.54	32544.82	51.97	0.21
20	K-20	33.83	33833.31	36.78	0.15
21	K-21	38.23	38226.92	36.93	0.13
22	K-22	26.65	26647.15	37.89	0.19
23	K-23	17.87	17870.13	31.32	0.24
24	K-24	33.69	33687.68	24.61	0.10
25	K-25	21.82	21822.64	5.87	0.04
26	K-26	41.49	41491.89	17.63	0.06
27	K-27	21.28	21280.20	0.92	0.01
Average		26.84	26836.94	26.57	0.13
Minimum		12.90	12899.49	0.92	0.02
Maximum		43.39	43386.40	51.97	0.25

V. CONCLUSIONS

The measured values of radon mass exhalation rate in Kolasib district ranged from 0.92 mBq/kg/hr to 51.97 mBq/kg/hr, with an average value of 26.57 mBq/kg/hr. These values were comparable to the results obtained in other districts of Mizoram. The radium activity concentration was found to ranged between 12.90 Bq/kg to 43.39 Bq/kg, with an average value of 26.84 Bq/kg. These values were found to be much lower than the critical value set by IAEA[17]. Also, correlation between radon mass exhalation rate and radium activity concentration shows that they have a positive correlation.

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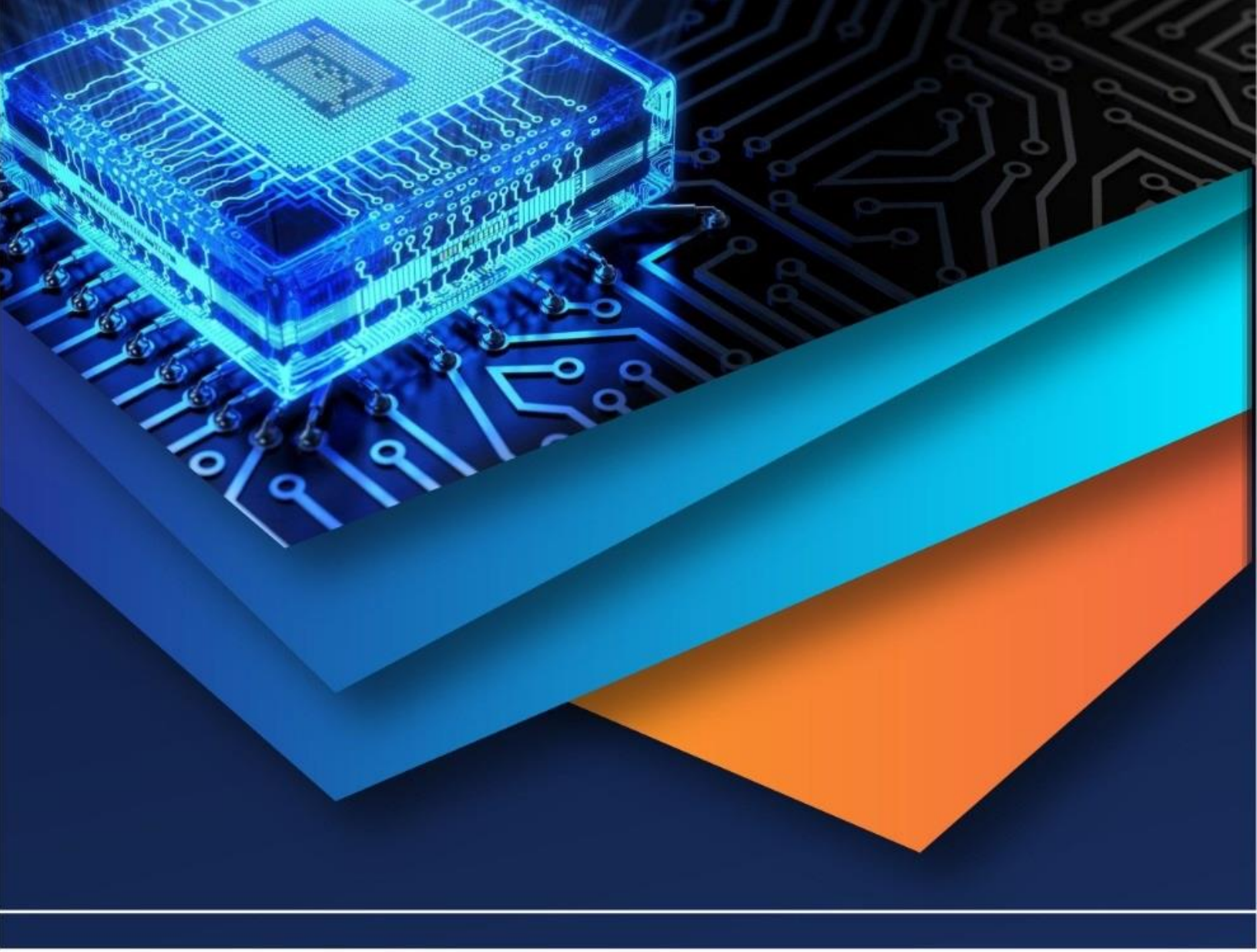
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