iJRASET
International Journal For Research in Applied Science and Engineering Technology



INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 3 **Issue:** IV **Month of publication:** April 2015

DOI:

www.ijraset.com

Call: 08813907089 | E-mail ID: ijraset@gmail.com

International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Preparation of Lead-Polymer Nano composite For Nuclear Shielding Applications

Zeinab M. Shakfa [#1], Ahmed E. Hannora [*2], Mohamed M. Sherif [#3]
[#] *Department of Physics, Faculty of Science, Cairo University, Giza-12613, Egypt.*
[*] *Department of Science and Mathematics, Fac. of Petroleum and Mining Engineering, Suez University* Suez- 43721, Egypt

***Abstract*— Nano Lead was prepared using polyvinyl alcohol as a process control agent in a high energy ball mill. The crystallite size and the micro-strain of the produced nano-Pb/PVA at different milling times were calculated from X-ray diffraction patterns. Field Emission Scanning and High Resolution Transmission electron microscopy were used to characterize the progress of Pb particles morphology through the milling process and to ensure the nanometric size of the milled nano-Pb/PVA then mixed with Polypropylene for gamma ray attenuation.**
***Keywords*—Lead; Polymer nanocomposites; Ball milling; Polyvinyl alcohol; Nanomaterials.**

I. INTRODUCTION

Gamma and X-rays radiation specifically are a primary concern for external exposure because of their high penetrating power and domestic effects on humans. Accumulated doses from ionizing radiation like X-rays and gamma rays can cause cancer, DNA mutations, sterility, etc [1]. With the increased usage of radioactive materials in medicine and industry, shielding is a priority in order to protect both humans and equipments. An example of a widely used shielding material for gamma and X-rays is Lead (Pb). Many dense elements can attenuate gamma and X-rays but Pb is distinguished by its availability and low cost compared with other denser elements like Tungsten and Thorium. Also, Pb has an advantage over various aggregate materials used to shield gamma such as concrete by being more efficient and uniform in density. Some shortages of pure Pb as a nuclear shielder are heaviness, being rigid and poorly portable beside the ability to produce secondary ionizing radiation which requires additional shielding. Many research efforts were done to design more efficient shielding materials that are potentially capable of attenuating gamma and neutrons in addition to being light weight, cost effective, easily portable and flexible.

Polymer composites are a suitable candidate to solve the traditional shielders' problems. The dispersion of Pb additives into high-performance hydrogen-rich polymeric material can provide a radiation shield for use against gamma and X-rays as well as secondary particulate and electromagnetic radiation resulting from nuclear reactions within the shield. Polymer composites are dual-purpose shielders as they combine the Hydrogen-rich polymer which is effective in absorbing neutrons and metallic additives which are effective for gamma shielding. Also, Polymer composites are light, commercially available and produce less secondary radiation when compared with pure metallic shielders. The multifunctionality of polymer composites assigned them to be used in many applications like shielding space crafts and making protective enclosures for humans and devices in hospitals and nuclear power plants [2, 3].

Making the metallic additives of a polymer composite in the nano scale remarkably enhances the final physical properties of the composite. Polymer nanocomposites acquire higher mechanical, thermal, electrical and optical properties comparing to their counterparts in micro scale [4, 5]. The relation between the nanometric size and the efficiency of nuclear shielding needs more investigation in depth. So, in this work we aimed to prepare a polymer nanocomposite with nano-Pb additives for future study of their nuclear shielding properties.

Ordinary chemical methods to prepare nano-Pb produce low yields in addition to being expensive. So, we tried to prepare nano-Pb by the mechanical method in a high energy ball mill (attritor). The previous trials to mechanically mill pure ductile metals like Pb ended up with particle growth and powder flattening to flakes with millimetres long [6-8]. Lead cannot simply be mechanically milled to the nanometric size because the process is impeded by excessive cold welding of the powder particles preventing them from fracturing. Hence, the critical balance between cold welding and fracturing that is necessary for successful mechanical alloying cannot be achieved. Up to our knowledge, no successful trial to prepare nano-size from pure elemental Pb by ball milling was reported in the literature.

One approach to reduce excessive cold welding and promote fracturing is to modify the surface of the deforming Pb particles by introducing a process control agent (PCA). PCA impedes the clean metal-to-metal contact necessary for cold welding and therefore prevent agglomeration. The used PCA in this study is polyvinyl alcohol (PVA) and has been chosen for its reported efficiency as a surfactant and it's compatibility with the main polymer (Polypropylene) [9-11]. PVA can help the nano-Pb particles to uniformly disperse in the polypropylene producing an ultra-large interfacial area per unit volume between the nano-

©IJRASET 2015: All Rights are Reserved

Pb and host polymer.

II. MATERIALS AND METHODS

Preparation of the samples used in this study was performed by a high energy ball milling method. Highly purified lead powder (100-mesh - Alfa Aesar), polyvinyl alcohol (PVA) with average Mwt ~ 57000 – 66000 (Alfa Aesar) and Polypropylene (Isotactic with average Mwt ~ 580000 and density ~ 0.9 gm/cm^3) were used as raw materials. To produce the nano sized Pb particles, micro sized Pb powders pre-mixed with PVA were pulverized with a high energy ball mill. The used mill was previously designed, manufactured and modified in order to increase its efficiency [12]. The grinding medium consisted of hardened steel vial and hardened steel balls of diameters of 7 and 9 mm. A water-cooling system surrounds the attritor vial to prevent the excessive increase in the powder temperature. The ratio of ball to powders weight was 10:1 while the weight ratio of Pb: PVA was 1:1. The milling process was carried out under dry Argon atmosphere with a rotation speed about 500 rounds per minute (rpm) at room temperature. The mechanical alloying was performed for different milling time intervals (30, 60 and 90 minutes).

The characteristics of the prepared samples in each step were evaluated by X-ray diffraction (XRD). XRD was performed using D5000 powder diffractometer using Cu K_α radiation (wavelength λ = 0.15406 nm). Field Emission Scanning electron microscopy (FE-SEM) and High Resolution Transmission electron microscopy (HR-TEM) were used to characterize the progress of Pb particles morphology through the milling process and to ensure the nanometric size of the milled nano-Pb/PVA.

To prepare the final composite for irradiation, different ratios of nano-Pb/PVA particles are dispersed in Polypropylene and milled at room temperature (25° C) in air atmosphere for around 10 minutes to ensure the homogenous mixing. The mixtures then cured by Isotactic hot pressing under pressure ~ 5 MPa and temperature from 260 – 270 ° C according to the concentration of Pb.

III.RESULTS AND DISCUSSION

X-ray diffraction patterns of pure Pb, PVA and Pb/PVA mixtures milled for 30, 60 and 90 minutes are shown in figure (1). The intense characteristic peaks of Pb gradually decrease and significantly broaden as the milling time increases because of the increment in lattice strain and grain refinement of Pb particles as a result of the repeated multi-formations of the impact distorting, cold welding and fracturing. Peaks belong to simple oxidized phases of Pb like PbO appeared due to leakage of air into the milling vial or by oxidation from PVA itself.

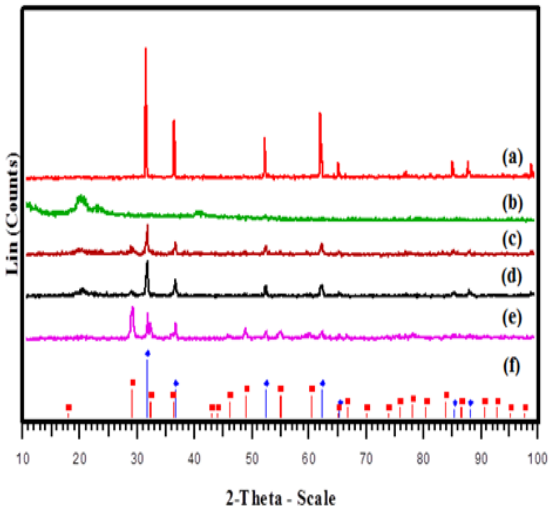


Fig (1). XRD patterns of (a) as received Lead, (b) PVA, (c, d and e) after mechanical milling/alloying for 30, 60 and 90 min. respectively,(f) ICDD (JCPDS) standard for (♦) Lead (01-0972) and (■) Lead Oxide (03-0561).

The Scherrer's formula estimates the size (*d*) of very small crystals from the measured width (*B*) of their diffraction curves [13]. After 30 minutes of mechanical milling, the crystallite size of Pb decreased from 721.2 nm to about 100.8 nm. The reduction in the average crystallite size is a result of the domination of fracture and de-agglomeration processes over the cold welding process by the assistance of PVA as a process control agent. PVA by being absorbed on the surfaces of nano-Pb particles helps to inhibit excessive cold welding by lowering the surface tension and preventing the clean metal-to-metal surface contact needed for Pb

©IJRASET 2015: All Rights are Reserved

particles to weld. After 60 min of milling, the decrement of crystallite size becomes more steadily till reaching 68.6 nm. At this stage, the cold welding process becomes more significant as the milling time increase but still inhibited by PVA. As milling time increase to 90 minutes, the crystallite size increase to about ~ 115 nm. This can be attributed to the agglomeration of the nano structured Pb particles and the increment in cold welding as milling time increase. The shortening of PVA in capping new nano-Pb particles and the domination of cold welding over fracturing at this stage can be explained by the consumption of the initial semi-crystalline phase of PVA into amorphous coating on the already produced nano-Pb particles specially with the nearly disappearance of its broad peak in the XRD pattern at 90 min of milling.

During the milling process, Pb particles face intensive plastic deformations such as dislocations and lattice distortions. The collision of Pb with milling tools causes the production of new grain boundaries subdividing the original grains into smaller grains or crystallites leading to total grain size reduction and micro-strain increment in the milled material. In the first 30 minutes, the strain increased from about 0.0414 before milling to 0.0549 due to the creation of plastic defects in the mixture by fracturing. As the crystallite size decreased steadily in the second half hour of milling, the strain "unlike expected" also decreased to 0.0545. This can be explained by the strain relaxation resulting from the subsequent annihilation and recombination of the produced dislocations to small angle grain boundaries separating the individual grains. These new sub grains are already in the nanometric size and can be the reason of the observed size and strain reduction at the same time. As milling progresses to 90 minutes, the strain increases with milling time till 0.0560 due to the domination of cold welding process.

From XRD analysis, the optimum time of milling to get the finest crystallite size is 60 minutes. The as received Pb and milled Pb/PVA for 60 minutes were investigated by FE-SEM and HR-TEM as shown in fig (2).

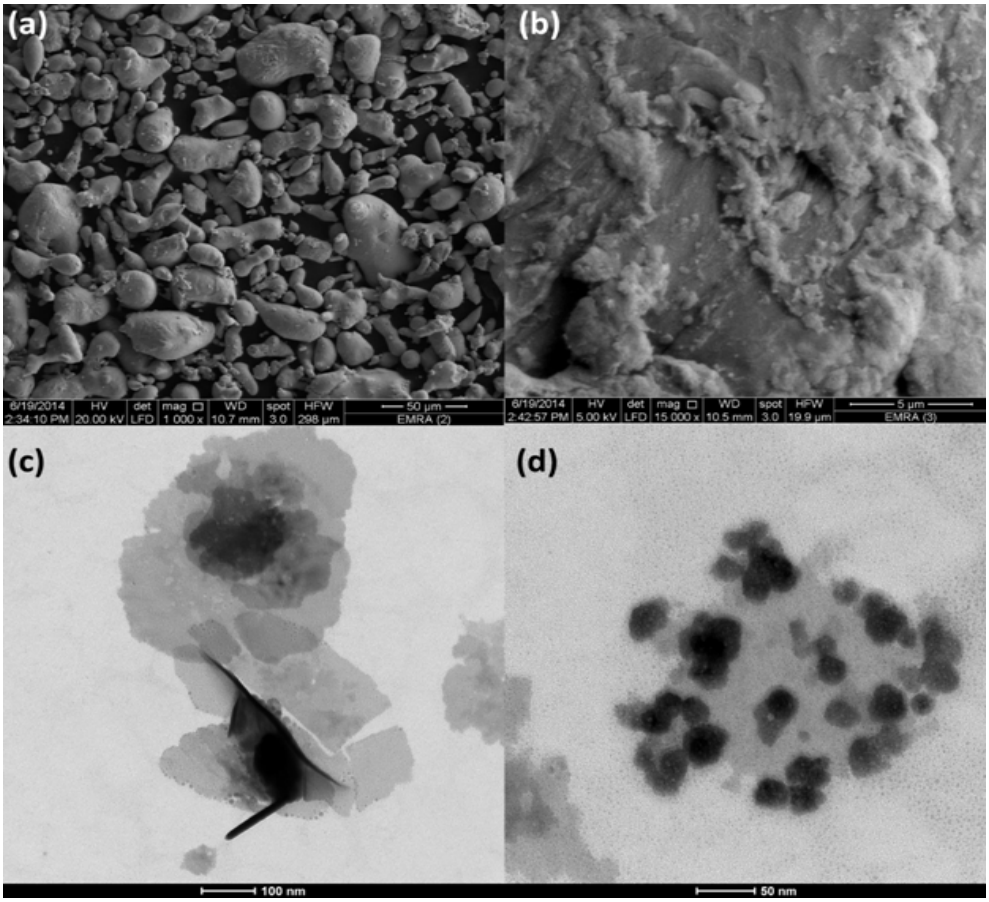


Fig (2) Electron microscopy images (a) FE-SEM of as received Lead,
(b) FE-SEM of 60 min milled Pb/PVA, (c, d) HR-TEM of 60 min milled Pb/PVA.

Fig (2-a) shows the FE-SEM image of row Pb powder with irregular shapes and particle sizes in order of microns. After milling time 60 minutes, Pb/PVA formed tiny thin flakes with diameters in order of few millimetres. These flakes are formed due to the

©IJRASET 2015: All Rights are Reserved

fusion of PVA under the instantaneous high temperature released from the strong impact with the balls during milling (local melting) and then rapidly cooled on the vial walls. Figure (2-b) shows the surface morphology of one of these flakes with only PVA layer without ensured appearance of Pb particles. The quasi-spherical and flaky shapes of the milled Pb particles noticed in HR-TEM images are absent in FE-SEM images so brighter spots are closer to be electrostatic discharges than being Pb particles. In addition, Dark and bright areas in FE-SEM have the same topography which indicates that these areas belong to the same material (PVA). The domination of PVA on the surface of these flakes indicates the successful capping of the milled Pb particles with the polymeric surfactant.

Bright field images of HR-TEM are shown with different magnifications in figure (2-c, d). Pb particles with crystallite size less than 100 nm can be recognized as separated particles coated by PVA or in the form aggregates and the diameter and shapes of these particles are not uniform throughout. Fig (2-c) shows a rose like patterns of centered Pb particles in order of 100 – 200 nm with different shapes separated and surrounded by PVA coating. Pb particles have morphologies between spherical and asymmetric flattened flakey shapes. This flakey shape is due to the ductility of Pb that causes Pb particles to undergo sphere-to-flake welding under the ball-to-ball or ball-to-vial collisions. The flattening and the relatively large size of these flakes indicate the significant presence of the cold welding process at this stage of milling. Fig (2-d) shows an aggregate of finer spherical Pb particles in order of tens nanometers separately adhered on the surface of a bigger ball of PVA ensuring the success of PVA in capping the nano-Pb particles and prevent most of them from re-welding again.

The experiment presents inexpensive top-down technique to commercially produce nano-Pb which could be used in various fields including defence, aerospace, and medicine and shielding large nuclear facilities.

IV. CONCLUSIONS

The production of nano-Pb by ball milling process is possible in spite of the ductility of Pb. The usage of PVA as a process control agent retards cold welding by capping the formed nano-Pb particles. The broadening of XRD diffraction peaks with milling time is due to the reduction in the crystallite size and the induced strain. The optimum milling time to reach finer size is 60 minutes with average crystallite size ~ 68 nm. The nanometric size and the capping of PVA on the nano-Pb particles are confirmed by XRD, FE-SEM and HR-TEM. HR-TEM images of capped lead showed the tendency of some particles to flatten with milling and the tendency of the finer nano-Pb particles to aggregate separately on the surface of larger particle of PVA.

V. ACKNOWLEDGMENT

The authors thank Mrs. Mai Seif for assistance with ball milling and samples preparation. Equipment support from Suez University, Faculty of Petroleum and Mining Engineering in Egypt are acknowledged.

REFERENCES

[1] W.R. Leo, Techniques for Nuclear and Particle Physics Experiments: A How-To Approach,2nd ed.,vol.1, Springer – Verlag , Heidelberg, Germany:1994.

[2] S. Nambiar, J. T. Yeow, “Polymer-composite materials for radiation protection,” ACS Applied Materials and Interfaces, vol. 4, pp. 5717–5726, November 2012.

[3] C. Harrison, S. Weaver, C. Bertelsen, E. Burgett, N. Hertel, E. Grulke, “Polyethylene/Boron Nitride Composites for Space Radiation Shielding,” Journal of Applied Polymer Science, Vol. 109, pp. 2529– 2538, 2008.

[4] L. Nicolais, G. Carotenuto, Metal – Polymer Nanocomposites, 1st ed., vol. 1. John Wiley & Sons, Inc, 2005.

[5] V. Harish, N. Nagaiah, T. Niranjana Prabhu, K. T. Varughese, “Preparation and characterization of lead monoxide filled unsaturated polyester based polymer composites for gamma radiation shielding applications,” Journal of Applied Polymer Science, vol. 112, pp. 1503-1508, May 2009.

[6] J.Y. Huang, Y.K. Wu, H.Q. Ye., “Ball milling of ductile metals,” Materials Science and Engineering (Elsiever), vol. A199, pp. 165-172, 1995.

[7] M. Babu, L. Jin-He , L. K. Jin, C. Y. Kuk, C. Suryanarayana , H. Soon-Jik, “Effects of atmosphere and milling time on the coarsening of copper powders during mechanical milling,” Powder Technology-Elsevier, vol. 256, pp. 251–256, February 2014.

[8] A.M. Harris, G.B. Schaffer, N.W. Page, “The morphological evolution of hollow shells during the mechanical milling of ductile metals,” Scripta Materialia, vol. 34, pp. 67-73, December 1999.

[9] Y. R. Uhm, J. Kim, J. Jung, C. K. Rhee, “Preparation and characteristics of nano-B4C/PVA particles and ultra high molecular weight polyethylene composites,” Korean Journal of Chemical Engineering, vol. 27, pp. 1338-1342, December 2009.

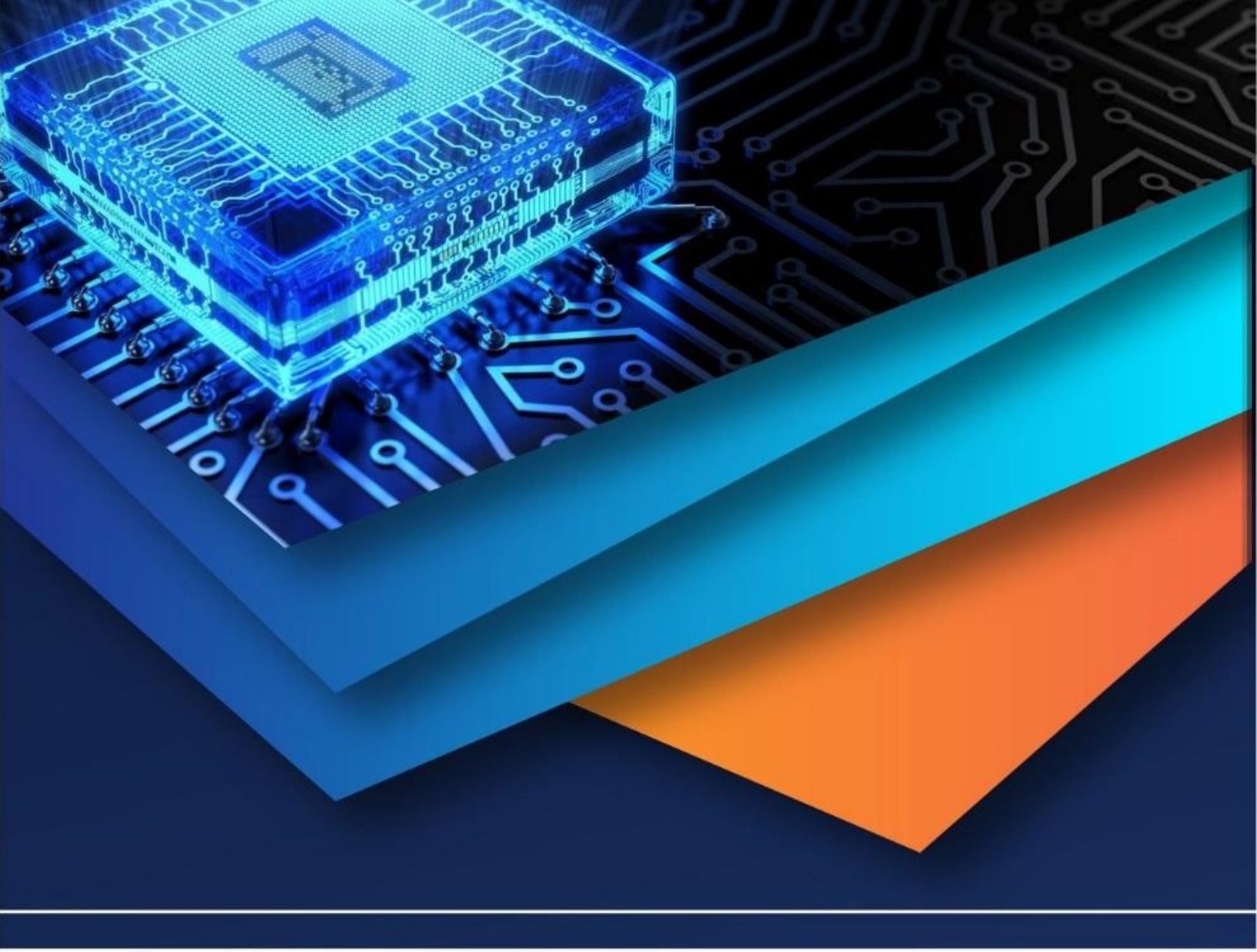
[10] B. Zhao, J. Wang, Z. Li, P. Liu, D. Chen, Y. Zhang, “ Mechanical Strength Improvement of Polypropylene Threads Modified by PVA/CNT Composite Coatings,” Materials Letters, Vol. 62, pp. 4380-4382, Nov. 2008.

[11] G A. Ujhelyiová, Z. Strecká, E. Bolhová, M. Dulíková, P. Bugaj, “Polypropylene Fibres Modifed by Polyvinyl Alcohol and Nanoadditive,” Fibers & Textiles in Eastern Europe, vol. 64, pp. 64-65, April 2007.

[12] A.Hannora, Study of Ti-Al Based Alloys” M.Sc. thesis. Faculty of Science, Suez Canal University, Suez, Egypt, April, 2001.

[13] V. K. Pecharsky, P. Y. Zavalij, Fundamentals Of Powder Diffraction And Structural Characterization Of Material, 1st ed.,vol.1, Library of Congress Cataloging-in-Publication Data, 2005.

©IJRASET 2015: All Rights are Reserved



10.22214/IJRASET

45.98

IMPACT FACTOR: 7.129

IMPACT FACTOR: 7.429

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Call : 08813907089 (24*7 Support on Whatsapp)