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Research Article

Evaluation the Effect of Cellulose Nanocrystalline Particles on Flexural Strength and Surface Hardness of Autoploymerized Temporary Fixed Restoration Resin

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ABSTRACT:

Background: Autopolymerized acrylic resin is commonly used material for construction of temporary restoration; however it has insufficient mechanical properties. The purpose of this study was to evaluate the effect of adding cellulose nano-particles on flexural strength and surface hardness of auto-polymerised acrylic resin.

Materials and Method: Following the acid hydrolysis of microcrystalline cellulose particles to achieve cellulose nanocrystals (CNC) particles, 40 specimens were fabricated for each test that divided to four groups (n=10) coded A-D. group A was the control group (without adding CNC). specimens in other three groups (B-D) were reinforced with CNC at loading of 1,2.5,5 wt%. flexural strength was assessed with a three-point bending test using a universal testing machine. surface hardness testing was conducted using a Vickers hardness tester. One way ANOVA and Tucky test were used to analyzed the data.scanning electron microscopy (SEM) observed was applied to evaluate the fracture pattern.

Results: A significant increase in flexural strength was observed in group with 2.5% cellulose nanofillers (P=2.5%),however no significant increase was observed in surface hardness. The SEM observation demonstrated brittle fracture in pure PMMA. By adding 2.5% wt of cellulose nanofillers, the characteristics of fracture is almost ductile, but in 5% wt of nanofillers,several cracks and void was observed at fracture surface.

Conclusion: Reinforcement of the Autopolymerized acrylic resin with 2.5% wt cellulose nanofillers significantly increased its flexural strength with no adverse effect on the surface hardness.

Key wprds: provisional restoration, Autopolymerized acrylic resin, microcrystalline cellulose, nanocrystalline cellulose, flexural strength, surface hardness.

[I] INTRUDUCTION

Fabrication of provisional restoration in order to protect the prepared teeth until the final restoration is completed is an important step in the fixed prosthesis treatment. Provisional restorations should have a physical and biological property, esthetic and durability for good performance, especially when the final restoration delivered to the patient is delayed.

Patients with full mouth reconstruction require long-term use of temporary restoration used to

evaluate the effectiveness of a treatment plan or to analyze the form and function of future permanent prosthesis. Also, long-term temporary restoration is required in patients treated with implants who need a long time for osseointegration. [1] One of the most widely used materials in Prosthetic Dentistry is poly methyl methacrylate autopolymerized acrylic resin (PMMA), which is commonly used in temporary restorations.[2] PMMA is a resin polymer whose mechanical and chemicophysical properties have been extensively researched. Ease of use, low cost, ease of repair and polishing ability have caused PMMA to be used for fabricating temporary prosthesis. However, there are problems with the mechanical properties of PMMA.[3] Insufficient flexural strength of autopolymerized resin can lead to fractures or cracks caused by bending forces caused by chewing. Great efforts have been made to improve the mechanical properties of acrylic resin materials by adding various materials such as metal fillers like silver, aluminum and titanium, but an adverse effect is their unseemly color. Other materials used to strengthen acrylic resin are fibers including aramid, carbon and polyethylene fiber which strengthen the acryl; however, the lack of esthetic, tough polishing, tissue irritation and difficulty of connecting to the resin matrix are the problems with this category of materials. [4] Recent advances in nanotechnology have led to changes in various fields such as composite materials. The strength of materials increases with fine particles to a critical size which is reported to be between 10 and 15 nm for the majority of materials.[5] Nanocrystalline cellulose derived from plants and wood pulp can increase the strength and hardness of the material that is in their composition. Adding a little of this compound can increase resistance to stress applied to polymeric compositions. Therefore, due to these properties, nanocrystalline cellulose has become a material with high power. [6] The effect of adding nanocrystalline cellulose with different concentrations on the mechanical and bio-activity properties of various polymers in the industrial, medical and pharmaceutical uses has been investigated in many studies. [7,8] Given that no study has been considered the addition of nanocrystalline cellulose particles to resin material used for making temporary fixed restorations, the aim of this study is to evaluate the effects of adding particles of nanocrystalline cellulose, on the flexural strength and surface hardness of autopolymerized acrylic resin for making temporary fixed restorations.

[II] MATERIALS AND METHODS

The materials used in this study included autopolymerized acrylic resin, PMMA (Tempron, GC corporation, Tokyo, Japan), microcrystalline cellulose particles (cellulose microcrystalline, (C4H1005) n, Merck, Germany), sulfuric acid 95-98% (sulfuric acid 95 -98%, Merck, Germany) and distilled water.(Table 1)

Cellulose nanoparticles are mainly classified into three branches of (NFC) nano-fibrils of cellulose, (NCC) nanocrystalline cellulose (BNC) bacterial nano cellulose which in turn are dependent on the cellulosic sources and production conditions, based on size, function and method of preparation. [9] In this study, for the production of nanocrystalline cellulose, acid hydrolysis of available commercial microcrystalline cellulose particles with dimensions of 10 to 15 microns was used. [10]

In the process of acid hydrolysis, 150 grams of cellulose micro crystal particles was combined with 300 ml of 95-98% sulfuric acid and was heated to a temperature of 50 ° C. The resulting composition was then kept at this temperature for 6 hours and, 700 ml of distilled water was added to the solution, while stirring continued. The resulting solution was then cooled to 27 ° C and underwent centrifugation (1200 rpm, 45 min) at the temperature of 25° C. Supernatants were collected from the surface of solution after centrifugation and again 600 ml of distilled water was added to the solution and three centrifugation cycles were repeated. Finally, filtered solution and nano-crystal particles were extracted and then, SEM analysis was performed to evaluate nano

state of the particles.(Fig.1) 40 specimens were prepared for each test, the specimens were divided into 4 groups (n = 10) and coded A -D. Group A was the control group in which no change was made to the composition of resin samples. specimens of the 3 remaining groups (B-D), were respectively strengthened by nanocrystalline cellulose particles at a ratio of 1, 2.5 and 5 wt%. Measurements were performed by electronic scale (Sartorius, Germany) with the precision of 0.0001 grams. Nano-crystalline cellulose particles with the specified ratios were mixed immediately with the acrylic powder after combining with the monomer using the sonication method in order to reduce particle aggregation and phase separation. The mixing process of acrylic resin was conducted based on the guidelines with the mass ratio of 1/2 powder to monomer for 30 seconds by metal spatula in a clean and dry container on the vibrator. This combination was casted in a tow-piece aluminum mold and after 5 minutes, the samples were removed from the mold.(Fig.2)

Acrylic specimens were prepared for flexural strength testing with dimensions of 50 *10 * 3.3 mm and surface hardness testing with dimensions of 30 * 10 * 3.3 After polishing the samples with silicon carbide polisher paper of grade 200, 400, 600, 800, 1000 and 1200 grits, the size of the specimens was measured by caliper and then kept incubated for 72 h at 37 ° C in deionized water. The three-point flexural strength test was conducted with a universal testing machine (Zwick Roall) at a speed of 5 mm / min, and the flexural strength was calculated using the equation $S = 3 FL / 2bd^2$ where F is force of wedge to the middle of the specimen, L is the distance between supports which was set at 40 mm for all specimen, and b and d, are the width and thickness of the specimen which were 10 and 3.3 mm, respectively. After the fracture of the specimen under the applied forces, SEM images from each group were selectively prepared by electron microscopy. To test the surface hardness of the micro Vickers test by digital Micro-vickers hardness tester (MHI, Koopa Co, Iran) the impact of diamond pyramid on specimen surfaces with a square form with 50 grams of force applied for a period of 10 seconds was determined by digital micro-hardness computer. For each specimen three different points (one in the center and two at each end border) were selected and an average of three points for each specimen were considered as surface hardness. The results of one way ANOVA and Tukey test were obtained by SPSS software.

[III] RESULTS

Flexural strength values of each group specimens, with mean and standard deviation for each of the four tested groups are shown in Table 2.

According to the recorded values, average flexural strength of weight groups 1, 2.5and 5 wt% of cellulose nanocrystalline were increased by 2.5%, 15.95% and 6.5%, respectively, compared to the control group. The highest amount of flexural strength was observed in the 2.5 wt% group. One-way analysis of variance (One way ANOVA) was used for flexural strength of the 4 groups and the following results were obtained and reported in Table 3.

P-value is less than the value related to the significant difference, i.e.,0.05, so the minimum average of one group and the other group was significantly different. To find a significant difference between the groups with different weight percentages of cellulose nanocrystalline (B, C, D) with the pure PMMA (A), Tukey HSD test was performed in accordance with Table 4.

The mean values of surface hardness of each specimen are shown with a standard deviation of each of the four tested groups in Table 5.

One way ANOVA was used to determine the surface hardness of the 4 groups and the results are reported in Table 6.

Given that the P- value is more than 0.05, there was no significant difference between the groups.

[IV] DISCUSSION

The results of the present work showed that Cellulose nanocrystal particles in all weight percentage of 1%, 2.5% and 5% increased flexural

strength compared to the control group where the most notable increase was observed in the 2.5% weight group of cellulose nanocrystal particles.

Flexural strength is a combination of compressive, tensile and shear strength, all of which represent stiffness and resistance to matter fracture. [11] In order to improve and optimize the mechanical properties of PMMA, three ways can be used to replace the PMMA with alternative materials, chemical modification of PMMA and PMMA reinforcement with other materials. The most widely used renewable materials are acrylic polymers modified with rubber. It is worth noting that the low flexural strength, fatigue failure and high costs have limited the use of these substances. [12] As an example of the changes in chemical composition, the application of metal compounds and ceramic fillers such as aluminum dioxide can be used to improve their mechanical properties. Ceramic fillers composition have been studied in dental materials which reflect biocompatibility and improved mechanical properties of the resulting composition. [13] In a study by Zhang,the reinforcing effect of denture base acrylic resin was investigated by nano-ziconia particles, where the highest flexural strength was observed by adding 1.5% weight of nano-ziconia particles and the highest surface hardness was observed by adding 1.5% and 2% weight of nano-ziconia particles. [14] Ellawa reported an increase in the flexural strength of high impact PMMA acrylic resin with AL2O3 particles at a ratio of 5 to 20 wt%. [13] Vojdani reported that the addition of 2.5% of AL2O3 particles can significantly increase the flexural strength of acrylic resin. [15] Cellulose is one of the most important natural polymers and a biocompatible material used on an industrial scale as an interminable raw material. It has been used in the form of wood and plant fibers as a source of energy, construction materials and clothing.[16] Several studies have shown that cellulose nanocrystalline can have many applications in medicine and industry such as regenerative drugs, optical appliance, automotive appliance and industrial composite materials. These nanocrystals which show a crystalline structure and high surface area crystals with a high tensile strength and a high Module of elasticity, can be used as an appropriate reinforcing filler for a variety of polymer and composite and thermoplastic materials. [17] Since the strength of materials with fine particles with a critical size for most materials increases between 10 and 15 nm, nano-sized particles were used in the present study. [18] Cellulose Nanocrystalline particles used in the study were produced by hydrolysis of commercial microcrystalline cellulose available in an acidic environment that was added to strengthen the pure PMMA polymer. Due to increased levels of nanoparticles and their high energy level, these particles tend to adhere to each other, which leads to loss of contact between the nanoparticles and the polymer and thus, reiforcing effect of the nano-filler is reduced. [18] In this way, finding the right amount of weight percentage of filler in combination with pure polymer is very important for its optimization. In the present study and the studies reviewed here, one of the reasons for improved mechanical properties, particularly flexural strength is that by adding crystalline fillers to PMMA when the force applied to PMMA is accompanied by the propagation of cracks, the nanoparticles absorb the crack's energy, thereby preventing the spread of the crack. [15] This will increase the flexural strength.

In this study, the flexural strength in Group 1% wt cellulose nanocrystal particles showed no significant difference compared to the control group; therefore, strengthening the acrylic resin with 1wt% cellulose nanocrystalline particles outweighs the benefits. In Group 1% wt dispersed particle concentration was still not enough to be able to effectively prevent the growth of cracks. Also in the assessment of SEM images of fracture surface, plenty of brittle fracture features were shown.(Fig.3)

In this study, the 2.5% wt group showed the highest flexural strength compared to the control group. The SEM analysis of this group showed

that the fracture surface was significantly rougher than pure PMMA and PMMA with 1 wt% of Nanofiller.(Fig.4,5) This means that in this group, the fracture surface features was significantly distanced from a crisp state because a sufficient amount of distributed Nanofiller is able to prevent the growth of cracks.

In this study, groups of 5 wt% cellulose nanocrystal particles showed a slight increase in flexural strength compared to the control group. In SEM assessment, fracture surface numerous bubbles and cracks were propagated.(Fig.6)

In Vojdani's study, a 5.8% decrease was observed in flexural strength by adding 5 wt% of AL2O3 particles to acrylic resin PMMA that resulted in a decrease in the bond strength between resin molecular networks. Increased filler volume in the polymer network as a confounding factor, void formation and agglomeration of filler particles were cited as possible reasons for this decline. [15]

In the present study the possible reasons for the decrease in the flexural strength of Group 5 wt% compared to 2.5 wt%, include reduction in the volume of the resin matrix that is load-bearing, changes in elastic module of resin and the form of crack propagation in the specimen due to the increased volume of filler, air trap, the formation of bubbles as the starting point of the fracture and finally the lack of sufficient Wetting of resin matrix and filler particles as well as a possible increase in particle accumulation.

In various studies, Micro-Vickers hardness test through average penetration rate of Micro-Vickers digital device indicator in three points of each sample has been used to evaluate surface hardness of the specimens.[3,15] This method has been used in present study.

In studies conducted by duraid Mohammed in 2012 [19] , Ahmed in 2014 [20] where zirconia-oxide nano particles were used and Vojdani in 2012 [15] where particles of AL2O3 were added, an increase in surface hardness was observed with an increased percentage of Nanofiller.

Improved surface hardness by adding metal oxide Nanofiller particles may be due to their strong ionic bond. In this study, Group 1 wt% and 2.5 wt% showed little increase in surface hardness and Group 5 wt% showed a slight decrease in surface hardness compared to the control group, but differences was not a significant. In the present study, cellulose nanocrystal particles have been used. Cellulose, which has a fragile nature, has the form of hexagonal glucose whose monomers are connected by glycoside bonds. Cellulose is a linear polymer in which multiple hydroxide groups tighten the chain by establishing a hydrogen bond between each other. The decrease observed in the surface hardness of cellulose nanocrystalline particles in Group 5 wt% in this study may be attributed to this character of Nanofiller. Considering the lack of a significant difference in surface hardness in Group 1 wt% and 2.5 wt% and a decrease in surface hardness in Group 5 wt% compared to the control group, in addition to the significant increase in the flexural strength of Group 2.5 wt%, it appears that addition of a 2.5 wt% weight of cellulose nanocrystal particles to PMMA compound is more economical than other mentioned percentages and can increases the flexural strength of the specimen compared to purely commercial specimen.

[V] CONCLUSION

Considering the limitations in this research it can be concluded that, adding cellulose nanocrystalline , like other mentioned strengthening fillers by certain weight percent is associated with improved mechanical properties, particularly flexural strength and above that weight percentage, the added cellulose nanocrystalline particles shows a decrease in mechanical properties as was observed for surface hardness following the addition of 5% of nanocrystalline cellulose particles.

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Table 1: materials used in the study

Material	Composition	Manufacture
Tempron	powder: polymethyl metacrylate liquid: methyl metacrylate	GC,Japan
Cellulose microcrystalline	cellulose microcrystalline powder with 10-15 micrometer particles	Merc,Germany
Sulfuric acid	Sulfuric acid 95-98%	Merc,Germany
Distilled water	Distilled water	Razi,Iran

Table 2: mean,SD,maximum, minimum for flexural strength

Groups	Numbers of samples	Mean	SD	MAX	MIN
A(pure PMMA)	10	61.OO	8.3	70.30	50.20
B(PMMA with 1% wt CNC)	10	62.52	6.3	75.25	52.25
C(PMMA with 2.5% wt CNC)	10	70.73	3.6	75.01	63.51
D(PMMA with 5%wt CNC)	10	64.96	3.7	69.25	60.25

Table 3:Inferential statistical results for flexural strength

ANOVA	Sum of squares	Df	Mean squares	F	SIG(P Value)
Between groups	547.969	3	182.656	5.291	0.04
Within groups	1242.853	36	34.524		
Total	1790.822	39			

Table 4: Tukey HSD for flexural strength

Groups	Mean difference	Pvalue
A & B	1.51	0.9
A & C	9.72	0.04
A & D	3.95	0.4
C & B	8.20	0.01
C & D	3.25	0.2
B & D	2.15	0.6

Table 5: mean,SD,maximum, minimum for Surface hardness

Groups	Numbers of samples	Mean	SD	MAX	MIN
A(pure PMMA)	10	21.02	4.5	21.4	12.5
B(PMMA with 1% wt CNC)	10	21.59	5.1	26.5	11.4
C(PMMA with 2.5% wt CNC)	10	23.76	2.5	39.3	20.4
D(PMMA with 5%wt CNC)	10	19.43	2.7	20.8	15.3

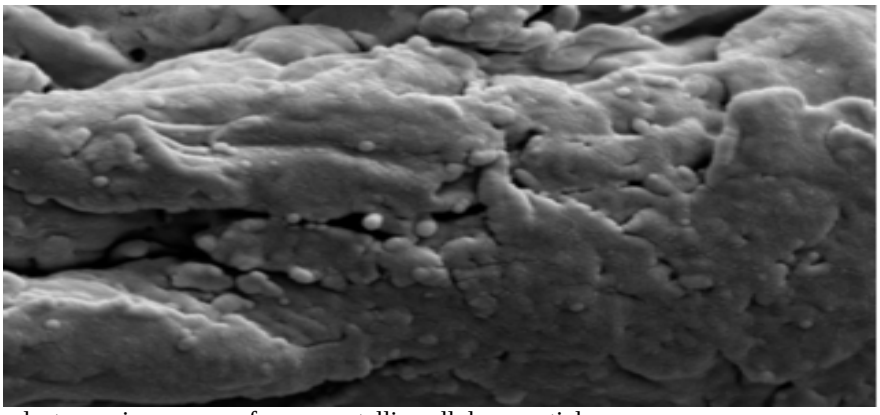

Figure 1:scaning electron microscopy of nanocrystallin cellulose particles

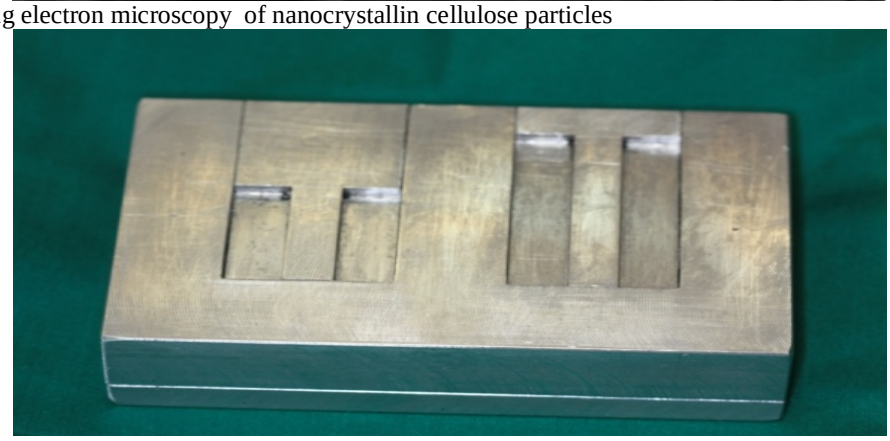

Figure 2: Aluminum mold

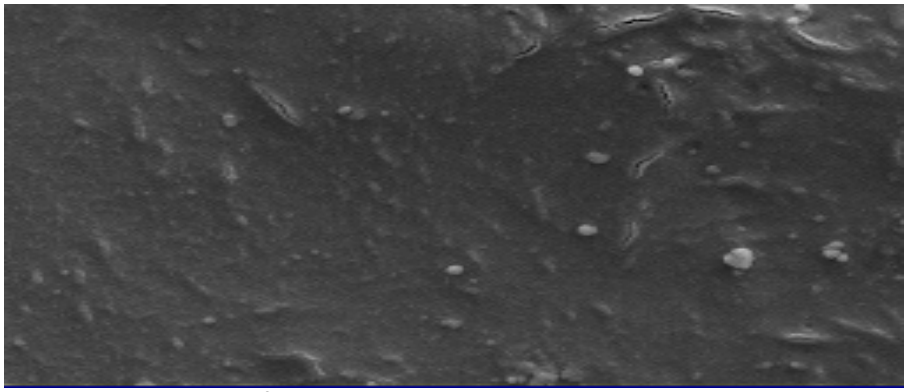

Figure3:scanning electron microscopy of PMMA with 1% wt nanocrystallin celloluse particle

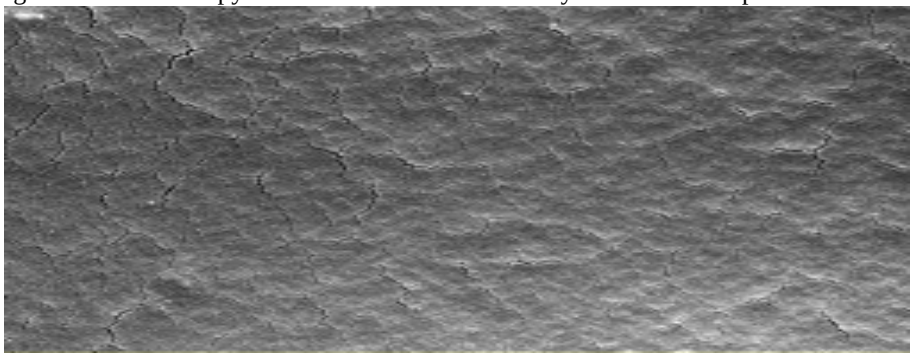

figure 4: scanning electron microscopy of PMMA without nanocrystallin cellulose particle

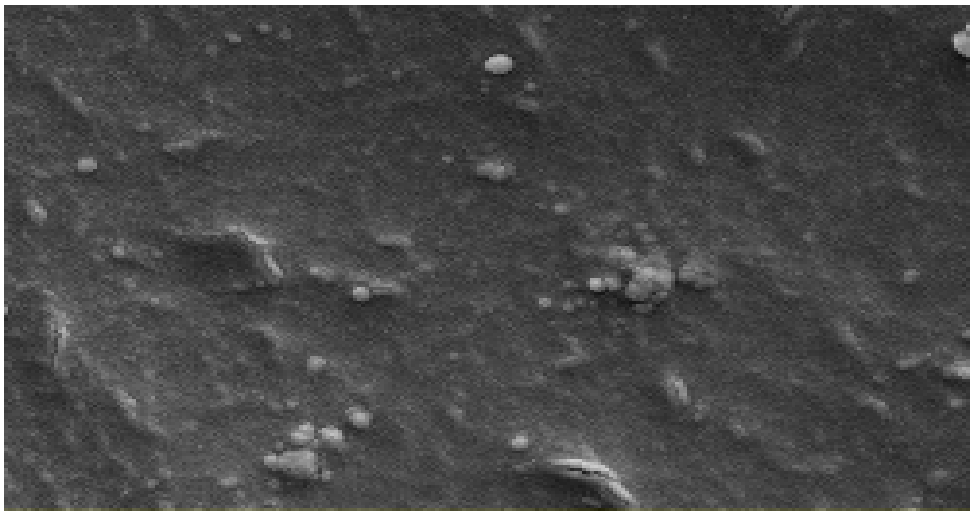

Figure5:scanning electron microscopy of PMMA with 2.5% wt nanocrystallin cellulose particle

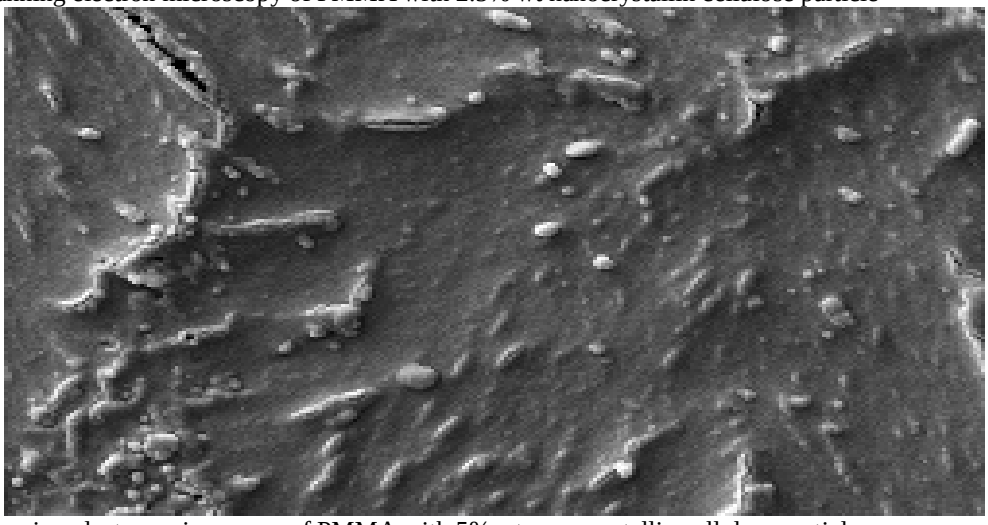

Figure6:scanning electron microscopy of PMMA with 5% wt nanocrystallin celloluse particle