## RESEARCH ARTICLE

## OPTIMIZATION OF POLYMER TOUGHENING PROCESS OF SOLID HYDROXYAPATITE IMPLANT.

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| Manuscript Info | Abstract |
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The experimental work of this study was conducted using a statistical experimental design in conjunction with the Taguchi method for optimization. The most effective factors affecting the toughening of the ceramic pellets were evaluated. The target of this experimental work was to maximize the product strength and minimize the amount of deposited polymer on the product. The Taguchi signal-to-noise (S/N) ratio was used for the optimization process. The experiments were divided into three stages. Stage One was identifying the parameters and the determination of the range of values to be explored; this step is usually done through literature review and simple experiments. Stage Two was the screening experiments (process characterization) to find out the most significant parameters effects on the process; this stage is done through the design of an orthogonal array based on the number of the input factors into the process. Stage Three was the optimization process itself [1-10]. Stages One and Two were reported in a previous study [9]. Stage Three is the subject of this study; for this stage the levels of the candidate factors were narrowed for more precise optimization. Four candidate factors with three levels were tried using an $L_9$ $(3^4)$ orthogonal array. The results were optimized using a statistical experimental design using the analysis of means and orthogonal array. The excess polymer removal technique used and the number of polymer solution coatings showed major effects on the flexural strength. Presumably, because these affect the mass of polymer applied. Optimum values of the flexural strength and mass of polymer were determined using statistical experimental design and the Taguchi method. Using the optimum experimental condition for preparing samples produces high quality in dense implants with 10.49 MPa flexural strength and 80 μm thickness of the deposited polymer, with mass of 21.0 mg / sample.

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## Introduction

Orthopedic practice often requires the use of synthetic bone-graft material to replace missing or severely damaged natural bone [11-23]. Ideally, such material would be biocompatible [21-29], suitably bioactive so that the implanted material may be fully remodeled [18-24], and adequately tolerant of mechanical orthopedic fixation [25-27]. Due to the chemical similarity with bones and perfect biocompatibility, the majority of bone substituting materials are made of

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(or covered by) calcium phosphates [28-30]. However, the mechanical properties of calcium phosphate based bioceramics are poor; the samples are brittle and cannot be easily shaped to the desired dimensions and form [31-33], so the application of calcium phosphates in surgery is reduced.

Polymer coatings are proposed to toughen ceramic implants because of the flexible properties of polymers. A polymer coating applied to the surfaces of a porous but compact ceramic implant would maintain the desirable porous structure. The incorporation of a biodegradable polymer to an implant material would minimize fragmentation following mechanical fixation [34-41], provide further increase in strength and toughness in keeping with the high-load performance requirements, and provide well-characterized binding sites for selected pharmaceutical agents [42-51]. This study deals with preliminary experiments with compact pellets as fore-runners to those with scaffolds in order to determine the principal features of the toughening with biodegradable polymer coatings.

The mechanical properties of the polymer coated ceramic implants are affected by various factors such as the technique used to remove the excess polymer solution, concentration of polymer, heat treatment temperature, number of coatings and so on. In this study, the influence of these factors on the flexural strength and the amount of the deposited polymer in the samples was analyzed. The interrelationships between the above factors are complex, and the analysis of this system to optimize the factors is a time and labor-consuming work. Hence, the analyses using conventional methods are inefficient.

In this study, statistical experimental design and Taguchi's parameter design were carried out simultaneously. The first can be considered as a raw data analysis (it focuses on the measured value itself) and the latter is the S/N data analysis. By raw data analysis, the effects of several factors on the mechanical strength were analyzed. The excess polymer solution removal method, concentration of polymer, heat treatment temperature and the number of coatings were chosen as significant factors. The influence of these factors was analyzed by S/N data analysis. The purpose of this study was to evaluate the effect of several factors on the mechanical strength of the ceramic implant coated with polymer and to optimize these factors using a statistical experimental design and the Taguchi's parameter design.

## Methodology:-
### Processing of Dense Ceramic Pellets
The green ceramic pellets consisted of a mixture of silica-contained hydroxyapatite (Si-HA) powder (with the $SiO_2$: HA ratio = 1:1) and polyvinyl acetate/retained sol binder. The Si-HA was prepared by the sol gel method described in detail elsewhere [52&53]. Pellets weight used was 0.25±0.01 gm of Si-HA. The mixture was then added into an 11 mm diameter die and pressed for approximately 30 seconds under 20.3 Mpsi. Later on, the pellets were sintered at 1250°C for 2 hours. Resulting pellets were approximately 11 mm in diameter, 1.1 mm thick, and 0.25 gm in weight.

### Preparation of PCL solutions
10, 12, and 14% (weight by volume) solutions of polycaprolactone, PCL, (average molecular weight 80,000 Da) were prepared by dissolving of 10, 12, or 14 gm of PCL in 100 ml of dichloromethane, respectively. These particular concentrations were used as recommended in a previous study [9]. The solutions were put into closed glass vessels and shaken over-night to complete dissolution.

### Polymer Coating of Samples
A simple dipping method was used for all samples. However, three different techniques for removing excess of the polymer solution were employed. The first one consisted of careful removal of the sample from the polymer solution with no removal of excess solution (no shacking). The second technique used was shaking by hands for 30 seconds (hand shaking). The final technique required immediate rotation of the sample for 30 seconds, in a closed metallic cylinder at a speed of 1000 RPM.
The number of dipping into the polymer solution, the concentration of the polymer solution and heat treatment were three other important variables. 2, 3, and 4 dipping in either 10, 12 or 14% polymer solutions were used. After dipping, each sample was allowed to dry for 15 minutes at room temperature. Following this, the samples were heated in an oven at 50, 60 or 70 °C for 30 minutes.

### Taguchi Design of Experiments [1-5]
Four parameters were examined in the polymer coating process of the pellet samples as shown in Table 1

**Table 1 -**Coating Process Parameters

| | Parameter | Levels | | |
|---|---|---|---|---|
| Symbol | | 1 | 2 | 3 |
| a | A - Removal tech. | No shaking | Hand-shaking | Spin-coating |
| b | B - Number of dipping | 2 | 3 | 4 |
| c | C- Heat treatment | 50 °C | 60°C | 70°C |
| d | D- Conc. of polymer | 10 % | 12 % | 14 % |

The orthogonal array $L_9$ $(3^4)$ was chosen for the optimization of the coating process, as detailed in Table 2. Eight samples were prepared for each trial.

**Table 2-** L9 Orthogonal array

| Experiment # | A<br>Removal | B<br># of Dipping | C<br>Heat Treatment | D<br>PCL Concentration |
|---|---|---|---|---|
| 1 | Hand Shaking | 2 | 50 °C | 10 % |
| 2 | Hand Shaking | 3 | 60 °C | 12 % |
| 3 | Hand Shaking | 4 | 70 °C | 14 % |
| 4 | Spin | 2 | 60 °C | 14 % |
| 5 | Spin | 3 | 70 °C | 10 % |
| 6 | Spin | 4 | 50 °C | 12 % |
| 7 | No Shaking | 2 | 70 °C | 12 % |
| 8 | No Shaking | 3 | 50 °C | 14 % |
| 9 | No Shaking | 4 | 60 °C | 10 % |

**The Signal-to-Noise (S/N) Ratio**
For the optimization process of this experimental work, two cases of Taguchi signal-to-noise (S/N) ratio were chosen, namely "more-is-better" for maximizing the measured response, and less-is-better in the case of minimizing the response as illustrated below:

The case for the optimal response being a maximum is referred to as the more-is-better (MB) case, which is appropriate for examining a process where it is desired that the mechanical strength of the product material should be maximum. The Taguchi S/N ratio here is then [1-4]:

$$\eta_{MB} = -10 \log_{10}\left( \frac{1}{n} \sum_{i=1}^{n} \frac{1}{y_i^2} \right) \qquad (1)$$

When the optimal response is desired to be a minimum, such as the amount of polymer deposited on the sample in this study, we have the less-is-better (LB) case, where the Taguchi S/N function becomes [1-4]:

$$\eta_{LB} = -10 \log_{10}\left( \frac{1}{n} \sum_{i=1}^{n} y_i^2 \right) \qquad (2)$$

Thus in the case of developing an effective but economic process to coat pellets, we are concerned primarily with two "properties" (strength and mass of polymer deposited), and we are investigating various "factors" (processing variables) in order to arrive at the optimal combination of high strength with the amount of polymer mass used at a minimum. In this way, we aim to define the processing conditions to produce pellets with acceptable strength but at minimum materials cost.

**Mass and Polymer Thickness Measurements on Pellets**
Before coating, the initial mass of each dense pellet was determined. After coating, the mass of each pellet was measured again. Therefore, for each experimental trial, the amount of polymer adsorbed on each pellet was known. The surface area of the pellets was estimated as follows:

$$S_{pellet} = 2\pi R^2 + 2\pi RH \qquad (3)$$

where: **R** is the radius of the pellet
        **H** is the thickness of the pellet

We assumed that the polymer coating was homogeneous and even. The density of PCL (1.146 g/cm$^3$) was considered similar for all concentrations. Based on these assumptions, the thickness of the polymer coating formed on the pellets was estimated as follows:

$$h = \frac{mS_{pellet}}{\rho} \qquad (4)$$

where: **h** is the PCL thickness on the surface of a pellet
        **m** is the mass of polymer deposited on pellet
        **ρ** is the density of PCL
An average PCL thickness was recorded for each pellet trial.

**Three Point Bending Test**
A small three-point bending jig was designed and built by joining a small steel rod (3 mm in diameter) perpendicular to a larger rod. The sample holder consisted of a further two rods (also 3 mm in diameter) joined to a metal block and placed 1 cm apart. A groove was made in the centre of the block to allow alignment of the load. Three-point bending tests were carried out on an Instron machine set for flexural strength. The Instron machine was calibrated with a full-scale load of 100 Kg and a crosshead displacement rate of 0.033 mm/min. The sample holder with a pellet was inserted placed centrally in the machine with the loading rod at a small distance from the sample. At this point the cross head was activated and the sample tested until a significant drop in load was indicated. The broken sample was removed from the jig, and the process was repeated with each pellet from all trials.
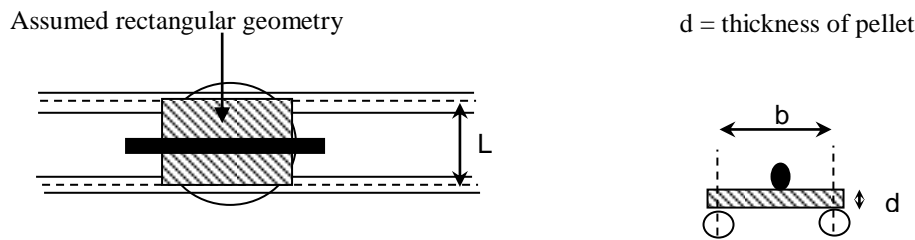


**Figure -** Three Point Bending Jig and Assumptions

Load versus displacement graphs were plotted for each sample from the data obtained during testing. The maximum load / load at fracture for each sample were determined from the maximum point on the graphs. Using calculations based on a rectangular geometry, the flexural strengths of the samples were determined:

$$\sigma = stress = \frac{Mc}{I} \qquad (5)$$

where: **M** = maximum bending moment
        **c** = distance from centre of specimen to outer fibers
        **I** = moment of inertia of cross section
        **F** = applied load

|  | M | c | I | σ |
|---|---|---|---|---|
| Rectangular | $\dfrac{FL}{4}$ | $\dfrac{d}{2}$ | $\dfrac{bd^3}{12}$ | $\dfrac{3FL}{2bd^2}$ |

$$\sigma_{fs} = \frac{3F_f L}{2bd^2} \tag{6}$$

where: $\mathbf{F_f}$ is the load at fracture, $\mathbf{L}$ is the distance between support points and the other parameters are indicated in Figure 2.

**Scanning Electron Microscopy**
The thickness of the polymeric coatings formed on the dense pellets (one pellet from each trial was studied) was measured by scanning electron microscopy after three-point bending test had been performed. Firstly, the samples were sputtered with gold to provide the surface conductivity necessary for use in the SEM. The samples were placed in the SEM and studied under various magnifications. Finally, images were photographed representing the surface features of the specimens.

## Experimental Results and Discussion
To find the optimal factor levels of the system a series of experiments were conducted. This has been done following an $L_9$ orthogonal array that uses narrower ranges of the factor levels near the initially predicted optimal settings. The most significant factors are the polymer removal method, number of coatings, heat treatment temperature and the polymer concentration. An "analysis of means", ANOM, was used to determine the effects of parameter changes on the mechanical strength and the deposited mass of polymer for the coated pellets. The results were used to determine the effect of each of the parameters on the measured responses. Tables 3 – 5 show the raw experimental data and the mean values of the measured responses, as well as the parameter effects on the flexural strength and the amount of polymer deposited in the samples after the coating process.

The effects of the parameters on the flexural strength and $\eta_{MB}$ for the pellet samples are shown in Figure 2. Of the parameters tested, the polymer concentration and the polymer removal technique have the largest effects on the flexural strength with the number of coatings and the heat treatment temperature having smaller effect. The predicted maximum flexural strength should be obtained for a sample prepared with "no shaking" technique, coated three times, heat treated at 70 °C and 14% polymer concentration. An examination of the plot for $\eta_{MB}$ (bottom plot in Figure 2) for the flexural strength shows that the effects on this response are very similar to those for the actual value of the flexural strength. Comparison of the parameter effects on the flexural strength and $\eta_{MB}$ shows that parameter effects are remarkably similar for the response, indicating that strength rather than variability dominated $\eta_{MB}$.

The effects of the parameters on the deposited mass of polymer on the pellet samples and $\eta_{LB}$ are shown in Figure 3. In this case, the spinning technique, the number of coatings, and the polymer concentration have large effects, while the heat treatment shows a small effect. Examination of the plot $\eta_{LB}$ (bottom plot of Figure 3) for the mass of polymer shows that the effects on the response are similar to the actual response (top plot of Figure 3).The predicted minimum mass of polymer should be obtained for samples prepared with the spinning technique, coated twice, heat-treated at 50°C and 10% of polymer concentration. The response dominates the variability in determining the signal-to-noise ratio, as shown by the fact that parameter levels that result in lower mass of polymer also results in high value of $\eta_{LB}$, since it is always desirable to maximize the signal-to-noise ratio. Figure 4 shows the surface SEM micrographs of the experimental trials for the pellet samples. Trials 1, 8, 9 show a porous structure of the polymer coating. The heat treatment of these trials was below the melting point of the polymer; as a result, the coating structure was not even, as can be seen in trials 2, 3, 7 where the samples of these trials were heat treated at temperatures 60 & 70 °C. Trials 4, 5, 6 show the minimum amount of polymer deposited on the surface due to the removal technique used in these trials, i.e. the spinning technique. Figure 5 shows the SEM micrographs of the cross-section of all the trials. The thickness of the deposited polymer on the surfaces of the pellets was calculated for the optimum experimental conditions to be approximately 80 μm.

Table **3**- Mean values of the flexural strength and mass deposited for the pellets

| | | | | | Flexural Strength (MPa) | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Sample # | Uncoated | Trial 1 | Trial 2 | Trial 3 | Trial 4 | Trial 5 | Trial 6 | Trial 7 | Trial 8 | Trial 9 |
| 1 | 3.492 | 5.795 | 16.090 | 16.400 | 4.519 | 8.152 | 4.927 | 5.696 | 24.840 | 7.478 |
| 2 | 4.537 | 9.830 | 9.165 | ----- | 4.696 | 3.529 | 4.592 | ------ | 17.210 | 7.727 |
| 3 | 4.479 | 7.722 | 7.668 | 11.750 | ------ | 8.898 | 2.619 | 4.922 | 26.370 | 7.170 |
| 4 | 4.049 | 4.945 | 8.433 | 25.640 | ------ | 5.071 | 4.456 | 5.646 | ------ | 6.202 |
| 5 | 4.053 | 4.945 | 5.316 | 12.480 | 4.013 | 7.677 | 2.827 | 7.415 | 13.460 | 7.722 |
| 6 | 2.995 | 6.401 | 6.243 | 16.750 | 4.465 | 4.153 | 1.814 | 10.420 | 8.166 | 4.619 |
| 7 | 4.144 | 6.998 | 10.410 | 13.130 | 3.257 | 5.406 | 6.329 | 10.690 | 10.720 | 8.410 |
| 8 | ---- | 3.533 | 4.298 | 13.020 | 4.081 | 3.791 | 3.882 | 7.469 | 9.052 | 7.772 |
| Mean | 3.964 | 6.271 | 8.453 | 15.596 | 4.172 | 5.835 | 3.931 | 7.465 | 15.688 | 7.138 |
| | | | | | Polymer Deposited mass (gm) | | | | | |
| Sample # | Uncoated | Trial 1 | Trial 2 | Trial 3 | Trial 4 | Trial 5 | Trial 6 | Trial 7 | Trial 8 | Trial 9 |
| 1 | -- | 0.010 | 0.020 | 0.040 | 0.005 | 0.004 | 0.007 | 0.012 | 0.035 | 0.019 |
| 2 | -- | 0.010 | 0.019 | 0.042 | 0.004 | 0.005 | 0.006 | 0.019 | 0.043 | 0.022 |
| 3 | -- | 0.010 | 0.027 | 0.042 | 0.005 | 0.005 | 0.007 | 0.016 | 0.039 | 0.026 |
| 4 | -- | 0.011 | 0.027 | 0.052 | 0.005 | 0.007 | 0.005 | 0.017 | 0.036 | 0.025 |
| 5 | -- | 0.010 | 0.027 | 0.035 | 0.006 | 0.004 | 0.006 | 0.015 | 0.039 | 0.026 |
| 6 | -- | 0.014 | 0.025 | 0.046 | 0.005 | 0.006 | 0.006 | 0.014 | 0.028 | 0.023 |
| 7 | -- | 0.017 | 0.025 | 0.046 | 0.006 | 0.007 | 0.008 | 0.013 | 0.033 | 0.028 |
| 8 | -- | 0.010 | 0.026 | 0.040 | 0.005 | 0.005 | 0.005 | 0.015 | 0.038 | 0.030 |
| Mean | -- | 0.012 | 0.025 | 0.043 | 0.005 | 0.005 | 0.006 | 0.015 | 0.036 | 0.025 |

Table **4-**Mean of the means of the flexural strength and deposited mass of polymer for pellets

| Trials & Samples | | Flexural strength (MPa) | | Mass of Polymer (gm) | |
|---|---|---|---|---|---|
| Trial # | # of samples | Mean | $\eta_{MB}$ | Mean | $\eta_{LB}$ |
| Uncoated | 8 | 3.964 | | 0.000 | |
| 1 | 8 | 6.271 | 27.42 | 0.012 | -21.41 |
| 2 | 8 | 8.453 | 29.04 | 0.025 | -27.85 |
| 3 | 8 | 15.596 | 35.44 | 0.043 | -32.70 |
| 4 | 8 | 4.172 | 24.24 | 0.005 | -14.25 |
| 5 | 8 | 5.835 | 25.54 | 0.005 | -14.79 |
| 6 | 8 | 3.931 | 22.22 | 0.006 | -16.02 |
| 7 | 8 | 7.465 | 28.94 | 0.015 | -23.68 |
| 8 | 8 | 15.688 | 34.14 | 0.036 | -31.27 |
| 9 | 8 | 7.139 | 29.04 | 0.025 | -27.99 |
| Mean | 8 | 8.283 | 28.45 | 0.0191 | -23.33 |

Table **5-**Parameter effects on the flexural strength and mass of polymer measurements and the signal-to-noise (S/N) ratio for pellets

| | | Flexural Strength (MPa) | | | Mass of polymer (gm) | | | |
|---|---|---|---|---|---|---|---|---|
| | # | Value | Effect | $\eta_{MB}$ dB | # | Value | Effect | $\eta_{MB}$ dB |
| Removal technique | 8 | H. shake | 1.814 | 2.26 | 8 | H. shake | 6.35 | -4.32 |
| | 8 | Spin | 1.823 | 2.18 | 8 | Spin | 7.18 | -3.99 |
| | 8 | No Shake | -3.637 | -4.45 | 8 | No Shake | -13.53 | 8.31 |
| Number of coatings | 8 | 2 | -2.314 | -1.58 | 8 | 2 | -8.53 | 3.55 |
| | 8 | 3 | 1.709 | 1.13 | 8 | 3 | 2.97 | -1.31 |
| | 8 | 4 | 0.605 | 0.45 | 8 | 4 | 5.56 | -2.24 |
| Heat Treatment | 8 | 50°C | 0.347 | -0.52 | 8 | 50°C | -1.07 | 0.43 |
| | 8 | 60°C | -1.696 | -1.00 | 8 | 60°C | -0.94 | -0.04 |
| | 8 | 70°C | 1.349 | 1.53 | 8 | 70°C | 2.01 | -0.39 |
| Concentration | 8 | 10% | -1.869 | -1.11 | 8 | 10% | -5.19 | 1.93 |

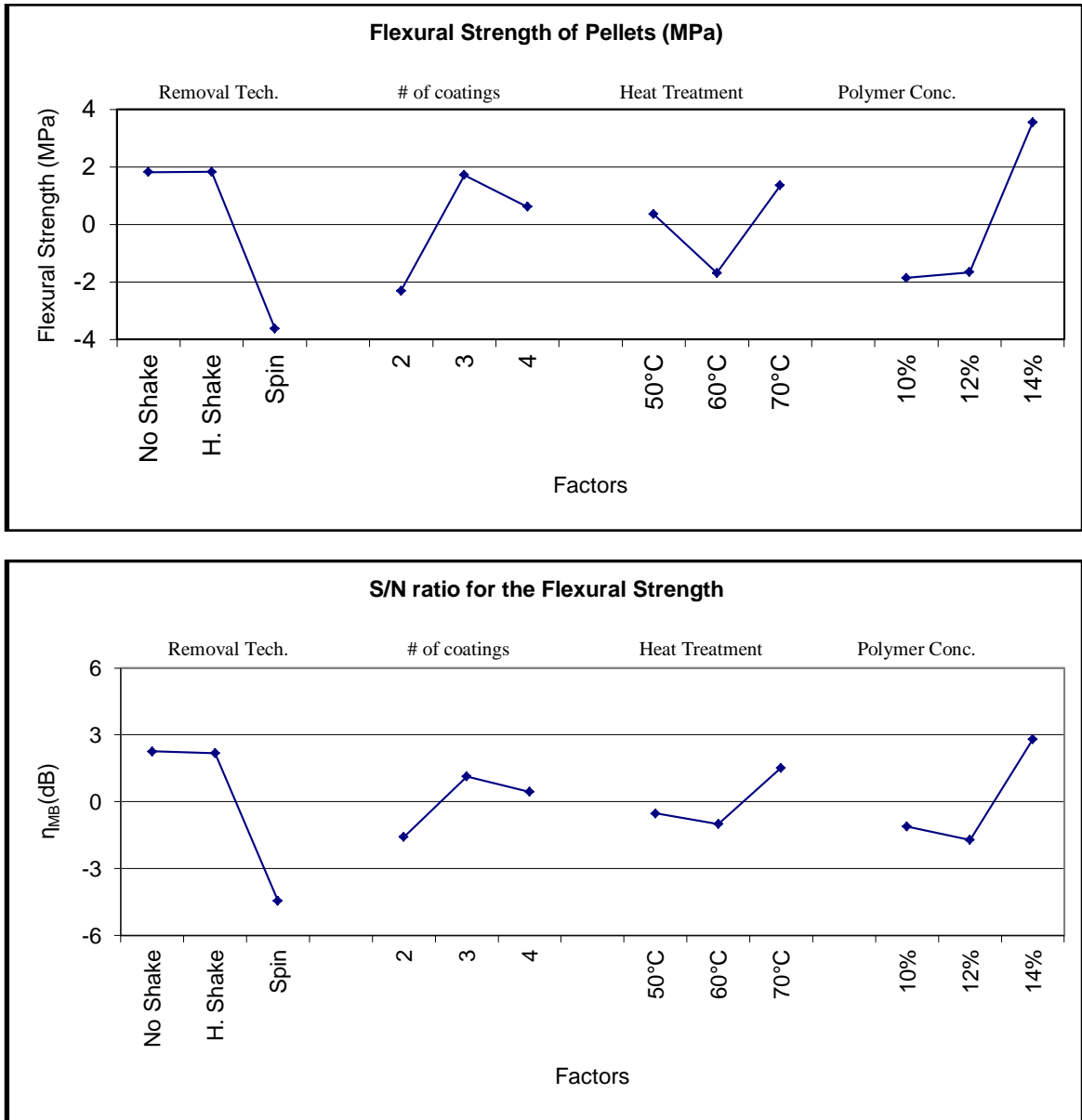| of PCL | 8 | 12% | -1.667 | -1.71 | 8 | 12% | -3.82 | 0.81 |
| solution | 8 | 14% | 3.535 | 2.82 | 8 | 14% | 9.01 | -2.75 |



**Figure 2-**Parameter effects on the flexural strength (top) and $\eta_{MB}$ (bottom) for the pellet samples.

*Note: the zero value on the Y-axis represents the flexural strength mean-of-means value, which is calculated to be 8.28 MPa. The graphs show the effect and S/N ratio of the parameters on this value.*
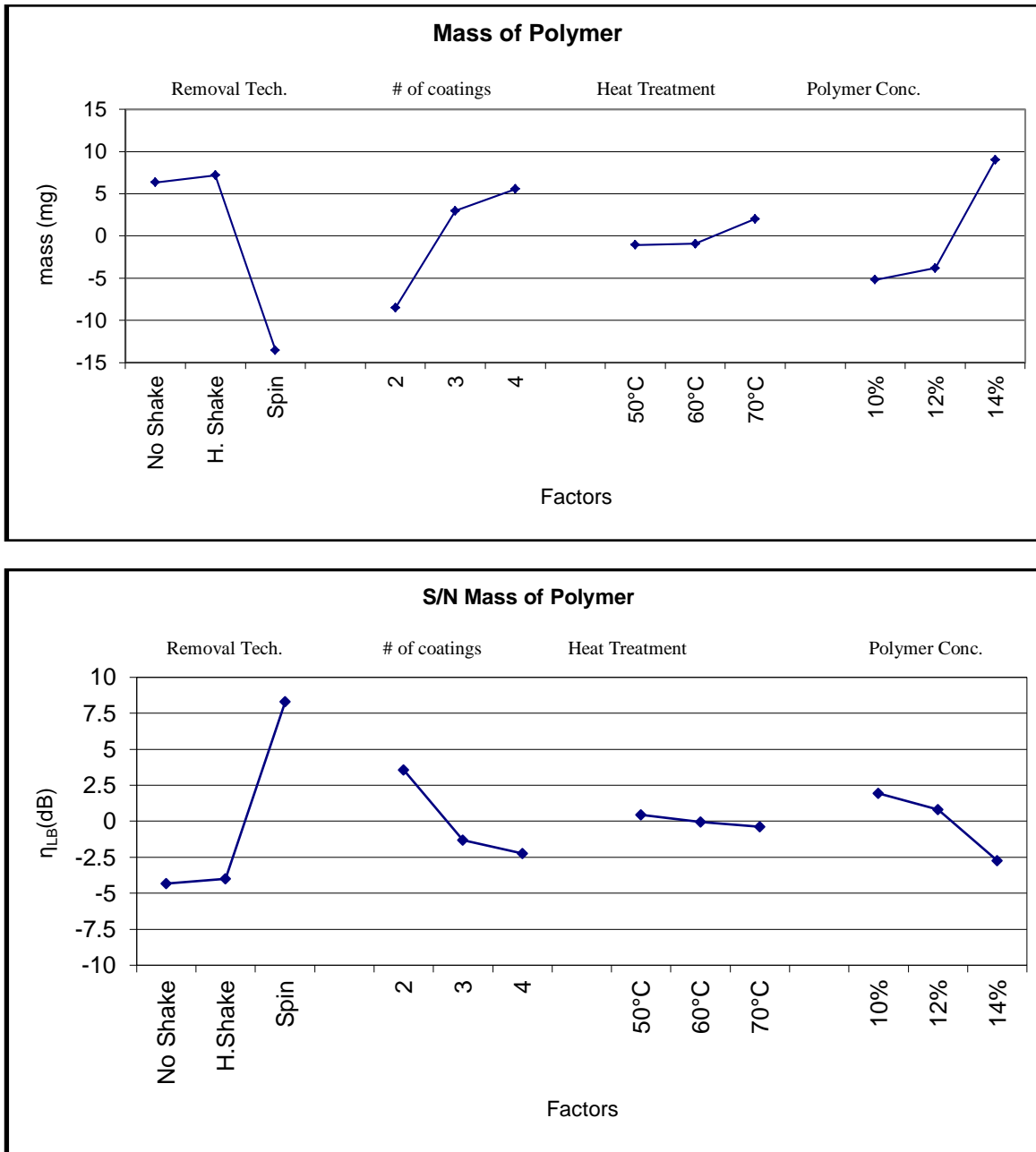
**Figure 3-**Parameter effects on the polymer mass (top) and $\eta_{LB}$ (bottom) for the pellet samples.

*Note: the zero value on the Y-axis represents the polymer mass mean of means value, which is calculated to be 19.1 mg. The graphs show the effect and S/N ratio of the parameters on this value.*
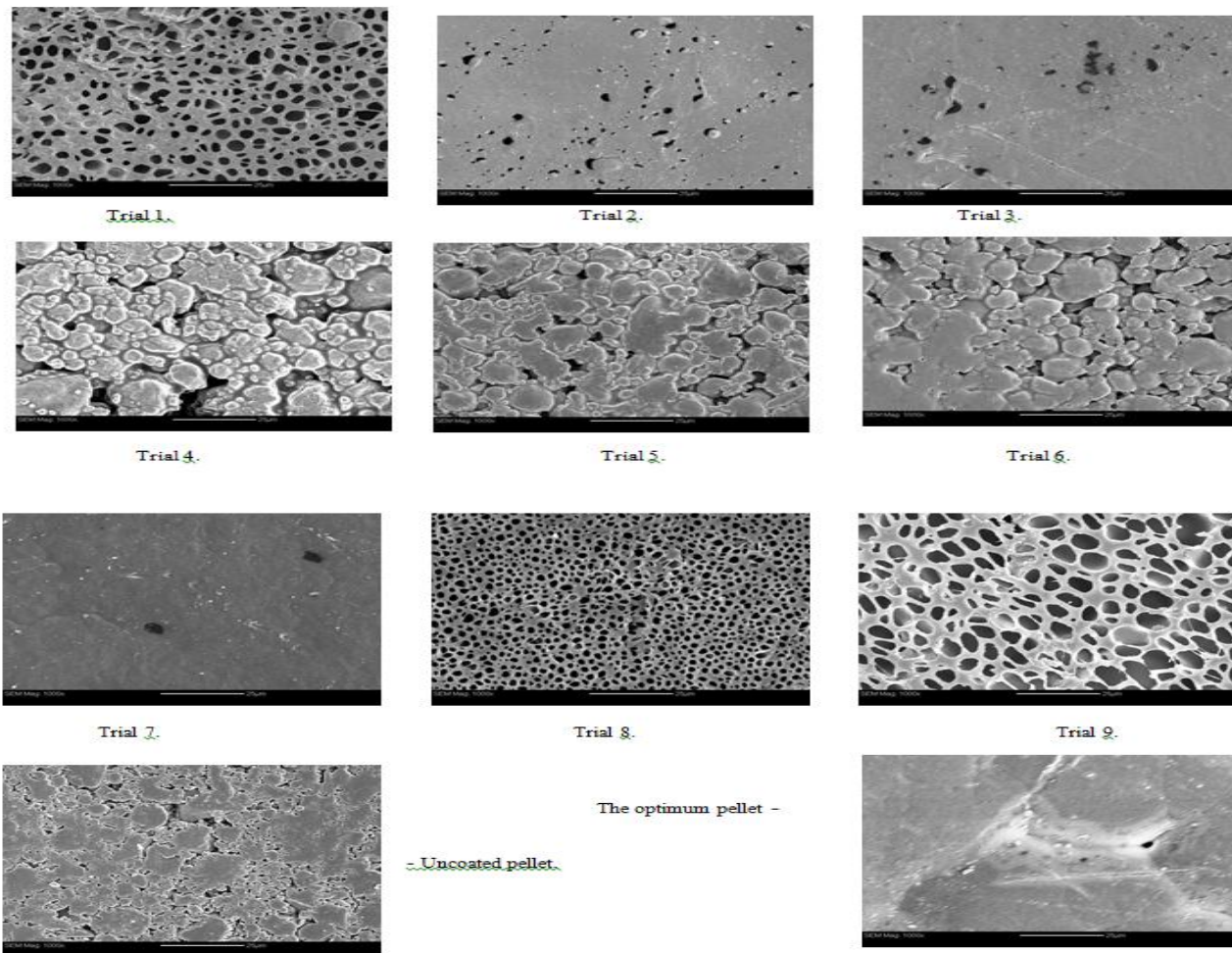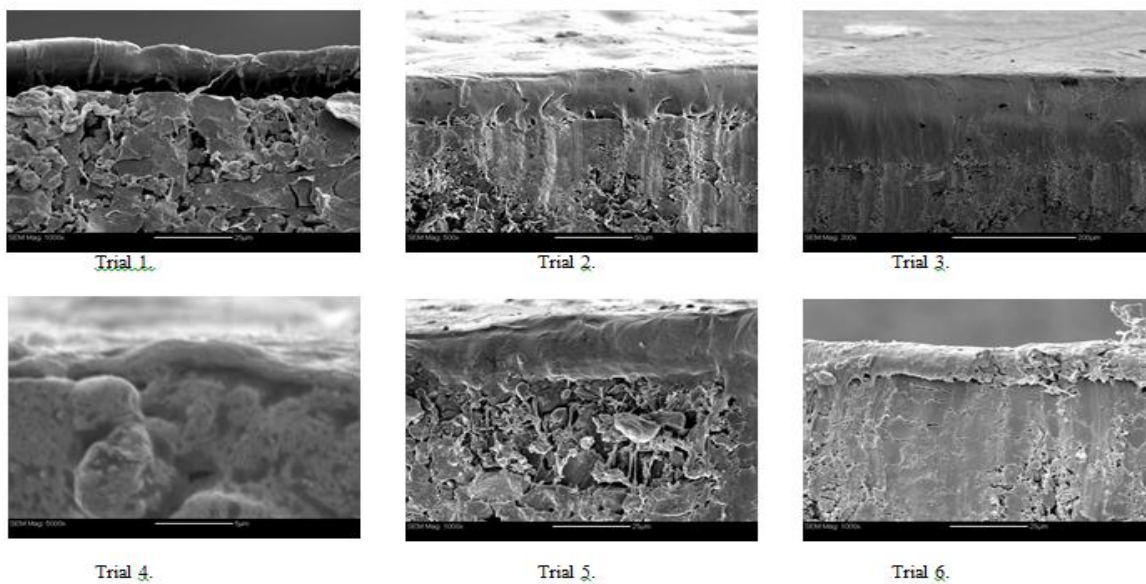
Trial 1.

Trial 2.

Trial 3.

Trial 4.

Trial 5.

Trial 6.

Trial 7.

Trial 8.

Trial 9.

The optimum pellet –

– Uncoated pellet.

**Figure 4-**SEM surface micrographs for the all pellet samples trials.



Trial 1.

Trial 2.

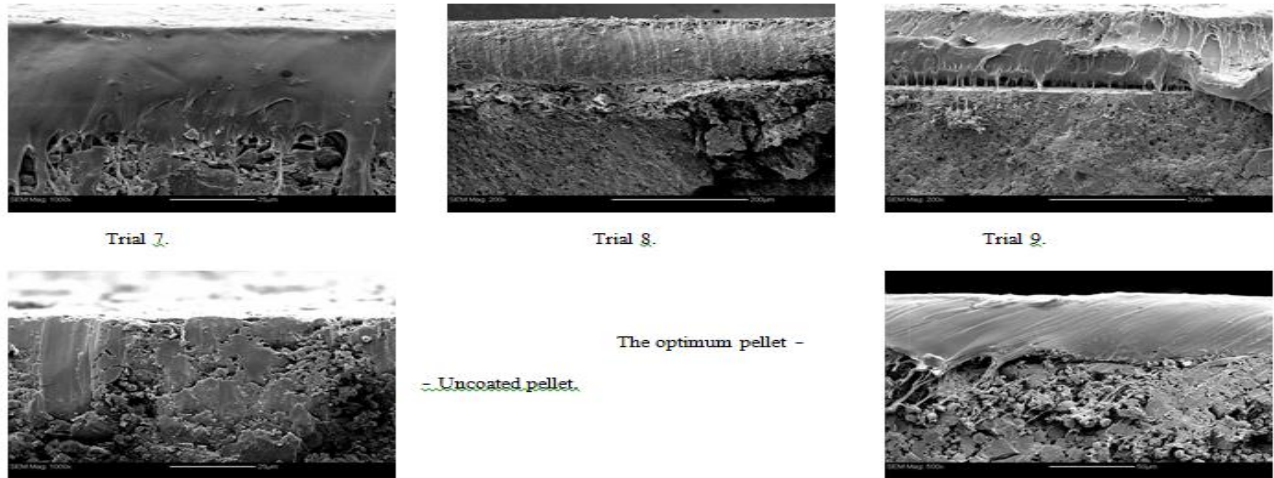Trial 3.

Trial 4.

Trial 5.

Trial 6.

**Figure 5 -**SEM Micrographs of the cross-section of pellet samples coated with polymer for the all trials.

**The Optimum Experimental Conditions**

The experimental conditions having the maximum signal-to-noise ratio are considered the optimum, as the variability of any characteristic is inversely proportional to the S/N ratio [1-8]. Consequently, the Taguchi method can determine the condition of least variability by the signal-to-noise ratio. Table 6 shows the optimum experimental conditions for the pellet samples for the flexural strength and the amount of polymer deposited on the samples. A confirming experiment should be conducted with the experimental conditions shown in Table 6. Table 7 illustrates the results of the confirming experiments. It is seen that all of the predicted results are reasonably close to the measured ones, indicating that the system behaved in a linear manner. In most of the nine cases, the predicted experimental condition yielded extremely good result, and these results were quite close to the predicted one. Therefore, the experimental conditions of Table 5 are the optimum conditions, which produce samples with 10.49 MPa flexural strength and mass of polymer 21.0 mg.

Table **6** -The optimum experimental conditions

| Response | Experimental Conditions | | Response | Experimental Conditions | |
|---|---|---|---|---|---|
| | *Parameter* | *Level* | | *Parameter* | *Level* |
| **Flexural strength** | Removal | no shaking | **Mass of the polymer** | Removal | spinning |
| | # of coats | 3 | | # of coats | 2 |
| | Heat treatment | 70°C | | Heat treatment | 50°C |
| | Polymer Conc. | 14% | | Polymer Conc. | 10% |

**Table 7 -**Parameters and levels of the confirming experiments

| **Parameters & Levels** | | |
|---|---|---|
| *Response* | *Exp. Conditions* | *Value* |
| | | **MPa** |
| **Flexural Strength** | | **8.28** |
| • Actual | $a_1b_2c_3d_3$ | 8.28 |
| • Maximum | $a_1b_2c_3d_3$ | 13.10 |
| • Measured | $a_1b_2c_3d_3$ | 10.49 |
| • Minimum | $a_1b_2c_3d_3$ | 2.33 |
| **Mass of Polymer** | | **mg** |
| • Actual | | **19.1** |
| • Minimum | $a_3b_1c_1d_1$ | 19.1 |
| • Measured | $a_3b_1c_1d_1$ | 4.5 |
| • Maximum | $a_3b_1c_1d_1$ | 21.0 |
| | $a_3b_1c_1d_1$ | 35.3 |

In addition, the yield strength / mass of CaP (MPa/gm of CaP) was calculated and found to be 57.26 MPa/gm as a maximum flexural strength and 9 MPa/gm as a minimum flexural strength the samples could withstand before fracture. The yield strength / total mass of CaP + PCL, (MPa/gm) was calculated to be 49.42 MPa/gm as maximum flexural strength and 10.47 MPa/gm as a minimum strength. The total mass of polymer attached to the surface of the pellets was estimated to be 35.3 mg as a maximum value, 4.5 mg as a minimum value and it is measured to 21.0 mg.

## Conclusion:-

The use of a statistical experimental design in conjunction with the Taguchi methodology of optimization, was found to be an effective method of optimizing the polymer coating process for the dense implants. The optimum experimental conditions were determined with high reproducibility, Table 5. Table 6 shows the results of the confirming experiments, the predicted, and the measured values of the responses. High quality dense implants were produced with a mean value of 10.49 MPa flexural strength and approximately 21.0 mg of polymer deposited on the samples. The flexural strength mean value of the uncoated pellets was 3.96 MPa.

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