

# Research Journal of Pharmaceutical, Biological and Chemical

## Sciences

### Deposition of ZnO Nano Particles on Stainless Steel Orthodontic Wires by Chemical Solution Method for Friction Reduction Propose.

M Kachoei<sup>1</sup>, B Divband<sup>2, 3\*</sup>, F Eskandarinejad<sup>1</sup>, and M Khatamian<sup>3</sup>.

<sup>1</sup>Department of Orthodontics, Faculty of Dentistry, Tabriz University of Medical Sciences <sup>2</sup>Research Center for Pharmaceutical Nanotechnology, Tabriz University of Medical Sciences, Tabriz, Iran <sup>3</sup>Inorganic Chemistry Department, Faculty of Chemistry, University of Tabriz, C.P. 51664, Tabriz, Iran.

#### ABSTRACT

In this paper, we report deposition of ZnO nanoparticles on stainless steel orthodontic wires by chemical solution method for the first time. The prepared samples were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM) coupled with energy dispersive x-ray analysis (EDX), visibleultraviolet diffuse reflectance spectroscopy (DRS) and transmission electron microscope (TEM) to evaluate particle structure, size distribution and composition. ZnO nanoparticles are almost hexagonal (spherical) in shape. The mean particles size of them is 40-45 nm. Coating of SS wires with ZnO nanoparticles has effect in decreasing of frictional forces which is an important problem in orthodontic treatments. Amount of friction force between rectangular SS wires and orthodontic brackets showed 51% decrease in zno coated group relative to non coated group of wires. Also after deposition of nano zno on round SS wires 39% decrease in friction force was observed. ZnO nano particles play as a significant solid lubricant between orthodontic brackets and wires.

Keywords: ZnO nanoparticles; Chemical solution deposition, Stainless steel wires, Friction reduction



\*Corresponding author baharakdivband@yahoo.com,



#### INTRODUCTION

Compact and uniform films and also self-assembled nanostructures of ZnO can be synthesized by various vapor-phase and solution-phase techniques. In comparison to the vapor phase techniques, the low-temperature synthesis from chemical solutions is simpler and more cost-effective. In chemical solution deposition, films are grown on the substrate through a controlled heterogeneous precipitation of ZnO. In successive chemical solution deposition (SCSD) process, a thin layer of zinc complex ions is adsorbed on the substrate by the immersion of substrate into the complex solution kept at room temperature. This step is followed by the immersion of the wet substrate in boiling water where the chemical reaction between water and the adsorbed ions leads to the growth of a few nm-thick and adherent ZnO film. The cycle of ion adsorption followed by chemical reaction in hot water is repeated to achieve the desired thickness [1–8].

ZnO seems to have the richest family of nanostructures (nanospheres, nanocombs, nanorings, nanohelixes/ nanosprings, nanobelts, nanowires and nanocages) among all materials, both in structures and in properties. These could have novel applications in dentistry and orthodontic treatments. Orthodontic treatment is directly related to tooth movements. One common procedure to translate a tooth in the dental arch is sliding it along an arch-wire, which is associated with advantages such as decreased clinical treatment time, patient satisfaction, and three-dimensional control of tooth movements [9,10]. In orthodontic treatments, when the tooth-bonded brackets move along the wire, friction results from the load naturally occurred on contact points [11]. Friction accompanies all sliding techniques and is considered an uncontrolled factor [9,12]. Some techniques have been developed to overcome this problem, such as use of wires of different metals, shapes, and sizes, as well as application of extra-oral forces or use of temporary implants [13,14]. Furthermore, the use of nanoparticles, invented in the 1990s, has been emphasized to decrease frictional forces between two metallic surfaces as excellent solid lubricants [15-17]. In order to utilize this characteristic of nanoparticles to decrease friction during orthodontic treatments, orthodontic wires or brackets must be coated with the nanoparticles [15]. Redlich et al. (2008) coated 0.019 inch × 0.025 inch orthodontic wires with inorganic fullerene-like nanoparticles of tungsten disulfide (WS<sub>2</sub>) and showed significantly reduced frictional forces on wires [15]. Naveh et al. (1995) reported reduced friction after coating Nickel-Titanium (NiTi) wires with nanoparticles of WS<sub>2</sub> in the laboratory [18]. Furthermore, stainless steel (SS) orthodontic wires were subjected to significantly decreased frictional forces when coated with nanoparticles of Carbone Nitride (CN<sub>x</sub>) as suggested by Wei et al. [19], Goto et al. (2008) demonstrated a reduction in the frictional coefficient of zinc oxide (ZnO)-coated SS substrates in vacuum [20]. Appropriate benefits of nanoparticles are related to, Rolling effects that cause two surfaces to slide on each other due to the spherical shape of the particles, Nanoparticles serve as spacers, preventing the contact between the two opposing surfaces, Third-body material transfer, which only occurs when the nanoparticles are released from the coated surfaces by electrolysis and transfer to the opposing metal (bracket) [15-17, 21]. Obviously, future clinical use of coated wires will depend on safe biocompatibility tests according to accepted procedures. Here, we report on the composition, and structure of films grown by SCSD process on stainless steel (SS) orthodontic wires. The aim of the present study was to assess the effect of ZnO spherical nanoparticle coatings on reducing frictional forces in sliding tooth movements.

#### EXPERIMENTAL

#### Materials

In this in vitro study, 80 orthodontic wires of 0.016 inch and 0.019 inch × 0.025 inch (American Orthodontics, USA) were used with and without ZnO nanoparticle coating. The studied devices included 40 SS brackets of the upper right centrals in 0.022 standard system (Ultratrimm, Dentaurum, Germany), 40 SS brackets of the upper right centrals in 0.018 standard system (Ultratrimm, Dentaurum, Germany), forty 0.016 inch SS straight wires (American Orthodontics, USA) with and without spherical ZnO nanoparticle coating as well as 40 rectangular 0.019 inch × 0.025 inch SS straight wires (American Orthodontics, USA) with and without spherical ZnO nanoparticle coating.

#### Measurements

X-ray diffraction patterns (XRD) were collected using a Siemens D500 diffractometer with Cu k $\alpha$  radiation ( $\lambda$ =1.5418 A° and  $\theta$ =4-80°) at room temperature. Scanning electron microscope (Philips XL30)



equipped with energy dispersive X-ray (EDX) facility was used to capture SEM images and to perform elemental analysis. The products have been characterized by transmission electron microscope (TEM) and energy-dispersive X-ray analysis (EDX). TEM studies, combined with EDAX were carried out on a Zeiss LEO 912 Omega instrument, operating at 120 kV. TEM specimens were made by evaporating one drop of solution of sample in ethanol onto carboncoated copper grids. Grids were blotted dry on filter paper and investigated without further treatment.

Furthermore, a universal testing machine (Huns field Test Equipment, H5K Model; England) was used to exert tensile and sliding movements between the wires and brackets.

#### **Experimental procedure**

In order to coat wires with ZnO nanoparticles, the bath was prepared by adding concentrated  $NH_3$  solution (35.28wt.%) as the complexing agent, to 100 ml of aqueous  $ZnSO_4$  .7H<sub>2</sub>O (0.5M) solution. Due to the formation of  $Zn(OH)_2$ , the mixture became milky in appearance. Further addition of ammonia dissolved  $Zn(OH)_2$  and yielded a clear zincate solution. The solution was then diluted to a volume of 500 ml. The concentration of ammonia in the freshly prepared bath was about 1.7M and the pH was 11.0. The wires were cleaned in an ultrasonic bath of ethanol for 30 min at 30°C just before the deposition.

#### **RESULTS AND DISCUSSION**

#### **Deposition mechanism**

In the SCSD technique, which is also known as SILAR (successive ion layer adsorption and reaction), ZnO is deposited as the result of a chemical reaction between boiling water and a thin layer of a zinc complex solution that the substrate carries.  $NH_3$  is used as the complexing agent and the reactions leading to the formation of zincate ions are,

$Zn^{2+}(aq)+4NH_{3}(aq)\leftrightarrow Zn(NH_{3})_{2}^{+4}(aq)$	(1)
$Zn(NH_3)_2^{+4} + 4HO^- \leftrightarrow ZnO_2^{-2} + 2H_2O + 4NH_3$ ,	(2)

The reaction between the boiling water and the zincate ions adsorbed on the substrate leads to the deposition of ZnO through,

$$ZnO_2^{-2} + H_2O \leftrightarrow ZnO + 2HO^{-}$$
 (3)

At a reaction temperature exceeding 50  $\degree$ C the zinc hydroxide film transforms to ZnO. The exact route and mechanism by which ZnO is deposited on the substrate, in a hot-water bath, is not very well known and needs further studies.

Different time intervals were selected for coating the wires with ZnO nanoparticles (10, 15, 20, 30, 40, 50, and 60 min).

#### Structure and morphology

The size and morphology of the samples are illustrated in Fig.1. SEM images of the SS wires confirmed ZnO nanoparticle coating in this method are uniform and in spherical shape. From particle size analyzer results, the synthesized ZnO nanoparticles on the wires have narrow size distribution with a mean diameter of 42 nm.





Figure 1: SEM of a wire coated with zinc oxide a) after 10 min, b) after 15 min, c) after 30 min, d) EDX after 30 min

Furthermore, a pilot study and assessment of frictional forces showed that the time interval of 20 min was the best choice for nanoparticle coatings, so in figure 2, The SEM images of a wire coated with zinc oxide before and after friction test were illustrated. As shown in figure 2 after friction test, most of the nano particles are not be separated from the wires and this fact is very pleasure. So nano particles are stable after friction test and this fact will cause reduction of frictional force between orthodontic wires and brackets on contact points when the tooth-bonded brackets move along the wire.



Figure 2: SEM of a wire coated with zinc oxide (20 min); a: before friction test (× 1500), b: before friction test (× 3000), c: after friction test (× 1500), d: after friction test (× 3000)

May – June

2015



The diffraction peaks for ZnO lifted off its SS substrate were in good agreement with those given in the standard data (PCPDF, 79-0207) and showed a good crystallinity. This means that as prepared materials had crystallized in a hexagonal wortzite ZnO. On the other hand, it is clear to see that the width of the reflections is considerably broadened, indicating that a small crystalline domain size. The average crystallite size of nano ZnO was determined as 45  $\pm$  5 nm from the line width broadening of the XRD peak corresponding to (002) reflection, using the Scherrer equation.



Figure 3: XRD pattern of ZnO lifted off its SS substrate

The TEM image of ZnO nanoparticles lifted off its SS substrate was illustrated in figure 4. ZnO nanoparticles are almost hexagonal (spherical) in shape and the mean particle size of them are less than 50 nm.



Figure 4: TEM image of ZnO lifted off its SS substrate

The diffuse reflectance spectra (DRS) of ZnO films deposited on SS wires shows in Fig. 5. The plot in the inset of Fig. 5 measures a bandgap of 3.38 eV for the film deposited on SS wires. This value is in excellent agreement with the Aexciton bandgap of ZnO that is 3.40 eV at 395 K [22-26].

May – June

2015

RJPBCS

**6(3)** 

Page No. 108





Figure 5: DRS spectra of ZnO film lifted off its SS substrate. The film bandgap energy (inset plot)

#### Friction Force evaluation

The friction force calculations during the sliding procedure were performed after nanoparticle coatings and preparation of wires. To simulate the type of movement that occurs during orthodontic treatment, i.e. sliding of a tooth across an archwire, we used Upper right central incisors (0.018 and 0.022 orthodontic systems) SS standard brackets(Dentarum ,Germany) were bonded with cyanoacrylate glue to aluminum plates by a bracket-mounting device. This apparatus insured the accurate and similar positioning of the brackets on the plates. Then the plate was fastened with screws to a notch in a special device, built for this study; it was then attached to the base of an Instron testing machine (Universal Testing Machine 4502, High Wycombe, United Kingdom) [27]. In order to create an identical condition for all the specimens, the brackets were changed after each wire sliding. The aluminum plate was positioned in three different notches angulated at 0°, 5° and 10° to the long axis of the device, using a special screw holding instrument for the simulation of the 2<sup>nd</sup>-order bends. Segment of 15-cm archwire was ligated to the brackets with an elastomeric module (orthotechnology, USA). The upper end of the wire was inserted into a tension load cell of the testing machine, and a 150-g weight was attached to the lower end of the wire .Each wire was drawn through the bracket at a constant speed of 0/5 mm/s for a distance of 12 mm while, the frictional forces were calculated by means of the universal testing machine [Fig. 6]

All the friction forces measured during this study were of kinetic type as no calculations were carried out at the baseline; however, after 0.1 s the friction values were calculated.]The mean and standard deviation of friction forces in each system, angulation and nanoparticle coating status were computed. The results were analyzed with Student t and "ANOVA (Analysis of Variance)" multiple comparison tests and significance was set at p < 0.05.

Along with the hypothesis suggesting frictional force reduction between orthodontic wires and brackets after coating with nanoparticles. The results showed a significant decrease in kinetic friction resistance to sliding in the ZnO-coated wires at different angles and both orthodontic systems of 0.018 and 0.022.

May – June

2015

RJPBCS

Page No. 109





Figure 6: Friction measurements by means of a universal testing machine

Mean friction force was estimated to be 1.5668±0.107 N and 2.56±0.3401 N for the coated and uncoated wires in all angles of the brackets and wires; suggesting significant reductions of the friction(39%) following ZnO nanoparticles coating (Student t: p<0.0001).

The frictional force significantly decreased to 32.48%, 41.48% and 41.23% at 0°, 5° and 10° angles, respectively, following ZnO nanoparticle coating on 0.016 inch wires compared to the same uncoated wires. In addition, the mean friction resistance significantly decreased by 52.17%, 52.96%, and 48.99% at 0°, 5° and 10° angles, respectively, after ZnO nanoparticle coating on 0.019 inch × 0.025 inch wires compared to the same uncoated wires.

The mean frictional forces in 0.019 inch  $\times$  0.025 inch wires showed 51% of reduction after nanoparticle coating (fig 7). Furthermore, the mean frictional forces in 0.016 inch wires showed 39% of reduction after coating (fig 8).



Figure 7: Mean friction force of 0.019 inch × 0.025 inch coated and uncoated wires

May – June

2015





Figure 8: Mean friction force of 0.016 inch coated and uncoated wires

Appropriate benefits of the nanoparticles are related to the followings:

- Rolling effects that cause two surfaces to slide on each other due to the particles sphere-like shape.
- Nanoparticles serve as spacers, preventing the contact between the two mating surfaces.
- Third body material transfer, which only occurs when the nanoparticles release from the coating surfaces by electroless procedure and transfer to the opposing metal (bracket)

At the first phase, when there is no angle between the bracket slot and the wire, that is, the bracket slot translates parallel to the wire, nanoparticles act as spacers decreasing the number of asperities which come into contact with each other leading in a decreased friction coefficient. As the angle grows between the slot and wire, the force increases at the edges of the slot and causes more friction resistance on the uncoated wire, But in coated wire some nanoparticles seem to exfoliate and slowly disintegrate when subjected to load application, releasing nanoparticles to the sheared interface causing solid lubrication effect.

Furthermore, when the materials are made of stainless steel like the uncoated wires, the friction coefficient is more which increases through the time, possibly due to the tribochemical reactions leading to oxidation and adhesion between the rubbed surfaces [27]. ZnO nanoparticles act as a protection against the oxidation of the metal surfaces and decreases friction resistance consequently.

#### CONCLUSION

ZnO nanoparticles were successfully deposited on stainless steel orthodontic wires by chemical solution method. ZnO nanoparticles are spherical in shape which is disirable for decreasing friction force. These nanoparticles are act as a lubricant and might offer a novel opportunity to significantly reduce friction during tooth movement and the consequent better anchorage control, reduced treatment time and risk of root resorption.

#### REFERENCES

- [1] Ristov M., Sinadinovski G.J., Grozdanov I., Mitreski M., Thin Solid Films 1987; 149: 65
- [2] A.P. Chatterjee, P. Mitra, A.K. Mukhopadhyay, J. Mater. Sci. 34, 4225 (1999)
- [3] A.E. Jimenez-Gonzailes, P.K. Nair, Semicond. Sci. Technol. 10, 1277 (1995)
- [4] X.D. Gao, X.M. Li, W.D. Yu, J. Solid State Chem. 177, 3830 (2004)

May – June

2015

RJPBCS

6(3)

Page No. 111



- [5] K. Ramamoorthy, M. Arivanandhan, K. Sankaranarayanan, C. Sanjeeviraja, Mater. Chem.Phys. 85, 257 (2004)
- [6] P. Mitra, J. Khan, Mater. Chem. Phys. 98, 279 (2006)
- [7] A.E. Rakhshani, Appl. Phys. A81, 1497 (2005)
- [8] A.E. Rakhshani, J. Kokaj, J. Mathew, B. Peradeep, Appl. Phys. A 86, 377 (2007)
- [9] Nikolai RJ. Bioengineering analysis of orthodontic. 1st ed., Ch. 2 and 3. Philadelphia: Lea and Febiger; 1985. p. 131-55.
- [10] Bednar JR, Gruendeman GW, Sandrik JL. A comparative study of frictional forces between orthodontic brackets and arch wires. Am J Orthod Dentofacial Orthop 1991; 100: 513-22.
- [11] Tidy DC. Frictional forces in fixed appliances. Am J Orthod Dentofacial Orthop 1989;96:249-54.
- [12] Chen WX, Tu JP, Xu ZD, Tenne R, Rosenstveig R, Chen WL, et al. Wear and friction of Ni-P electroless composite coating including inorganic fullerene-WS<sub>2</sub> nanoparticles. Adv Eng Mater 2002;4: 686-90.
- [13] Thorstenson GA, Kusy RP. Resistance to sliding of self-ligating brackets versus conventional stainless steel twin brackets with second-order angulation in the dry and wet (saliva) states. Am J Orthod Dentofacial Orthop 2001;120:361-70.
- [14] Cash A, Curtis R, Garrigia-Majo D, McDonald F. A comparative study of the static and kinetic frictional resistance of titanium molybdenum alloy archwires in stainless steel rackets. Eur J Orthod 2004;26:105-11.
- [15] Redlich M, Katz A, Rapoport L, Wagner HD, Feldman Y, Tenne R. Improved orthodontic stainless steel wires coated with inorganic fullerene-like nanoparticles of WS<sub>2</sub> impregnated in electroless nickelphosphorous film. Dent Mater 2008;24:1640-6.
- [16] Tenne R, Margulis L, Genut M, Hodes G. Polyhedral and cylindrrcal structurs of WS<sub>2</sub>. Nature 1992; 360:444-5.
- [17] Feldman Y, Wasserman E, Srolovitz DJ, Tenne R. High-Rate, Gas-Phase Growth of MoS<sub>2</sub> Nested Inorganic Fullerenes and Nanotubes. Science 1995;267:222-5.
- [18] Naveh GR, Redlich M, Rapport L, Feldman Y, Tenne R. Inorganic fullerene-like tungsten disulfide nanocoating for friction reduction of Nickel-Titanium alloy. Nanomedicine 2009;4:943-950
- [19] Wei S, Shao T, Ding P. Study of CNx film on 316 L stainless steel for orthodontic application. Diam Relat Mater 2010;19:648-56.
- [20] Goto M, Kasahara A, Tosa M. Reduction in frictional force of ZnO coatings in a vacuum. Jpn J Applied Phys 2008;47:8914-6.
- [21] Drummond C, Israelachvili J. Dynamic phase transitions in confined lubricant fluids under shear. Phys Rev E Stat Nonlin Soft Matter Phys 2001;63:041506.
- [22] J.F. Muth, R.M. Kolbas, A.K. Sharma, S. Oktyabrsky, J. Natayan: J. Appl. Phys. 85, 7884 (1999).
- [23] M. Khatamian, B. Divband, A. Jodaei, Degradation of 4-nitrophenol (4-NP) using ZnO nanoparticles supported on zeolites and modeling of experimental results by artificial neural networks. Materials Chemistry and Physics. 134 (2012) 31--37.
- [24] M. Khatamian, A.A. Khandar, B. Divband, M. Haghighi, S. Ebrahimiasl, Heterogeneous Photocatalytic Degradation of 4-Nitrophenol in Aqueous Suspension by Ln (La<sup>3+</sup>, Nd<sup>3+</sup> or Sm<sup>3+</sup>) doped ZnO Nanoparticles. 365 (2012) 120-127.
- [25] C.W. Teng, J.F. Muth, U. Ozgur, M.J. Bergmann, H.O. Everitt, A.K. Sharma, C. Jin, J. Narayan: Appl. Phys. Lett. 76, (2000), 979.
- [26] B. Divband, M. Khatamian, G.R. Kazemi Eslamian, M. Darbandi, Synthesis of Ag/ZnO nanostructures by different methods and investigation of their photocatalytic efficiency for 4-nitrophenol degradation, Applied Surface Science 284 (2013) 80– 86
- [27] M. Redlich, Y. Mayer, D. Harari, I. Lewinstein. In vitro study of frictional forces during sliding mechanics of "reduced-friction" brackets. Am J Orthod Dentofacial Orthop, 2003; 124: 69-73.