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Short Communication

Corrosion Resistance of Passive Films on Orthodontic Bands in Fluoride-Containing Artificial Saliva

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The purpose of this study was to evaluate the influence of fluoride ions on the corrosion behavior of orthodontic bands in artificial saliva. Thirty as–received orthodontic bands were divided into five groups randomly, and then immersed in fluoride-containing artificial saliva with different NaF concentrations (0wt.%, 0.1wt.%, 0.3wt.%, 0.5wt.% and 1wt.%) separately at 37℃ for 30 days. Potentiodynamic polarization, electrochemical impedance spectroscopy(EIS) and Mott-Schottky measurements were used to study the corrosion behaviour of orthodontic bands. Surface characterization was observed with scanning electron microscope (SEM) and energy dispersive X-ray spectrum (EDS). Statistical analysis was carried out using 1-way analysis of variance (ANOVA). The pitting corrosion could be observed on the orthodontic bands in fluoride-containing artificial saliva. With the increase of concentration of fluoride, corrosion current density increased, corrosion potiential, pitting potiential and the impedance of passive film decreased. Passive films of the bands formed at high concentrations of NaF had more disordered structures and higher defect densities. There were significant differences for pitting potential, corrosion current density, polarization resistance and carrier density of the experimental groups compared with control group ($P<0.05$). Fluorine ion participated in the development of pitting corrosion of orthodontic bands in artificial saliva. Severer corrosion occurred with the increase of concentration of fluoride. For more efficient treatment, the amount and the frequency of using fluorine-containing products should be considered in clinic.

Keywords: Orthodontic band; Fluoride; Potentiodynamic polarization; EIS; Passive film

1. INTRODUCTION

Numerous metallic materials are applied in orthodontics, such as bands, brackets, archwires. As we know, orthodontic appliances in oral environment are affected by saliva [1,2], appliance loading, masticatory forces, temperature fluctuations [3,4] and varieties of food [5,6]. Various microorganisms, enzymes and many aggressive ions can cause corrosion during long time exposure [7,8], and result in the loss of the essential properties of appliances. The release of some metal constituents [9,10] can cause allergic reactions [11,12], therefore, the corrosion performance of orthodontic materials in oral environment is an important factor to determine its biocompatibility. Up to now, there are two aspects to evaluate the corrosion resistance of metal alloys. The ion release [13] is usually analyzed with atomic absorption spectrometry. Electrochemical corrosion experiments in artificial saliva is to assay, for example, IR drop, the corrosion potential, passive current density, polarization resistance and pitting potential [14].

The corrosion of certain metals and alloys have been related with the presence of fluoride ions[15]. Fluorides are widely used in stomatology by means of toothpastes, orthodontic gels, mouth rinses and other therapeutic dental products in order to prevent dental caries [16], since fluoride ion can promote the formation of calcium fluoride that stimulate remineralization. Moreover, fluorides can be taken in through tea, fluoridated water and salt etc. It has been found that fluoride presents a negative effect on the corrosion resistance of orthodontic wires [14-19] ,brackets [20,21] and miniscrew implant [22,23]. In orthodontic treatment, orthodontic bands have been massively used and the residence time of the bands in human mouth are in long duration. However, up to now, the influence of fluorides on corrosion behaviors of orthodontic bands are limited in published literatures.

The metal band is widely used in orthodontics due to its advantage of lower shedding than buccal tube. They are routine employed accompanying with facebow, maxillary expansion appliances, intermaxillary traction and so on. In clinical, the fracture of orthodontic band occurs occasionally, it may be due to stress and other corrosion. Orthodontic band is mainly used on the first molar which is the most prone to dental caries. The rougher metal surface is, the more likely dental plaques, one of the factors of dental caries, accumulate. Therefore, the corrosion resistance of orthodontic band is vital to oral health.

Fluoride is also existed in glass ion binder and zinc polycarbonoxylate cement which are often applied in orthodontic band bonding, therefore, orthodontic bands are easily exposed to fluoride medium. The hypothesis is that the F^- has a negative effect on the corrosion resistance of commercial band. The electrochemical corrosion behaviour of the orthodontic band is estimated by electrochemical measurements and immersion test in artificial saliva with various NaF concentrations. The findings are expected to offer guidance for the development and manufacturing of orthodontic bands in clinical treatment.

2. MATERIALS AND METHODS

2.1. Materials and solution

The orthodontic bands (SWA, Xinya, Hangzhou, China), the subject of this research, were from the same batch number. These as-received bands were machined into specimens with dimensions of 5mm×10×mm×0.2mm for electrochemical measurements, and the exposed measurement area was 5×10 mm retained by the epoxy resin. Prior to the experiment, the samples was polished with silicon carbide (SiC) water polishing papers down to 2000#, and then ultrasonically cleaned in acetone and rinsed in distilled water.

The test solution was artificial saliva in the present work. All chemical reagents uesd in the experiments were analyzed the pure , and the deionized waterwater was used in the experiment. The formulations are given: (0.33 g KH_2PO_4, 0.34 g Na_2HPO_4, 1.27 g KCl, 0.16 gNaSCN, 0.58 g NaCl, 0.17 g $CaCl_2$, 0.16 g NH_4Cl, 0.2 g urea, 0.03 glucose, 0.002 g ascorbic acid, 2.7 g mucin in 1000 ml deionized water/pH 7) [24]. According to previous published works, 0.1-0.5 (wt.%) sodium fluoride and sometimes even up to 2% contained in saliva solutions [14,15], Consequently, NaF solutions of 0.1, 0.3, 0.5, and 1wt.%) were prepared, and defined the experimental group (2,3,4 and 5) . Meanwhile, the artificial saliva without NaF is as a control group(group 1).

Based on preliminary data and a power analysis, 6 band specimens were determined to be adequate for each experimental condition (5 conditions×6 bands) to meet the constraints of α=0.05 and power1-β=0.80.

2.2. Electrochemical measurements

Electrochemical tests were performed with PAR multi-channel workstation. A three-electrode system was applied, with the orthodontic bands as working electrode, a saturated calomel electrode (SCE) as reference electrode and platinum gauge as counter electrode. Prior to the experiment, the sample was immersed in the test solution for about 1h to get a stable open circuit potential, and then the electrochemical measurements were carried out with 6 parallel samples.

The polarization curves were recorded potentiodynamically with a scanning rate of 0.5mV/s, starting from -0.25V vs.OCP to transpassive potential. Electrochemical impedance spectroscopy (EIS) tests were performed at OCP with a frequency range of 100kHz~10mHz at the amplitude of 10mV. The experimental results were interpreted based on an equivalent electrical circuit by using a suitable fitting procedure of ZsimpWin. The Mott-Schottky measurements were carried out at a fixed frequency of 1kHz, an amplitude of 10mV with a scanning potential range of -0.2 V~1.0 V, and the interval was 50mV. All measurements were carried out at ambient temperature approximately 37 ^{o}C.

2.3. Immersion test

Immersion test was performed on the as-received orthodontic bands with dimensions of 5mm×10×mm×0.2mm in different fluoride-containing (0 wt.%, 0.1 wt.%, 0.3 wt.%, 0.5 wt.% and 1 wt.%) artificial saliva at 37 ^{o}C for 30 days, After the test, the macro morphology was observed with scanning electronspectroscopy (SEM, FEI Quanta 250). Besides, the composition of corrosion products was analyzed with energy disperse spectroscopy (EDS).

3. STATISTICAL ANALYSIS

Statistical analysis was performed with statistical software (Version 13.0, SPSS, America), The electrochemical and EIS fitting parameter values of the orthodontic band with different concentrations of NaF in the artificial saliva ,each containing 6 replicate specimens (*n*=6) were compared by one-way analysis of variance (ANOVA) followed by the Tukey post-hoc test. One-way ANOVA and Dunnett's T3 post- hoc were used for carrier density. The purpose of these comparisons was to assess whether NaF concentration(0.1wt%, 0.3wt.%, 0.5wt.% and 1wt.%) for the bands caused significant changes in resistant corrosion properties compared with the control group. A value of P=0.05 was considered significant.

4. RESULTS

4.1. Potentiodynamic polarization studies

The potentiodynamic polarization curves are shown in Figure 1. As observed in Fig. 1, the band displays typical passive behavior in artificial saliva at different concentrations of NaF. With the increase of concentration of NaF, the pitting potential was significantly negative shifted. The value of these parameters can be obtained by using a curve-fitting approach, the fitting results are listed in Table 1.

The means and standard deviations for the electrochemical parameters for each group measured are shown in Table 1. E_{corr} represents the potential of zero current, E_{pit} stands for pitting potential, i_{corr} is the corrosion current density, b_a is the anode Tafel slope and b_c is the cathode Tafel slope. The E_{corr}, E_{pit} and i_{corr} of the control group was significantly different from all the experimental groups ($P<0.05$), there were significant statistical differences in experimental groups ($P<0.05$) except group2(0.1%) and group3(0.3%) for E_{corr} value. Significant differences existed between the control group and all the experimental groups for b_a, but no differences were found in experimental groups. There were no significant differences in all groups for b_c ($P >0.05$).

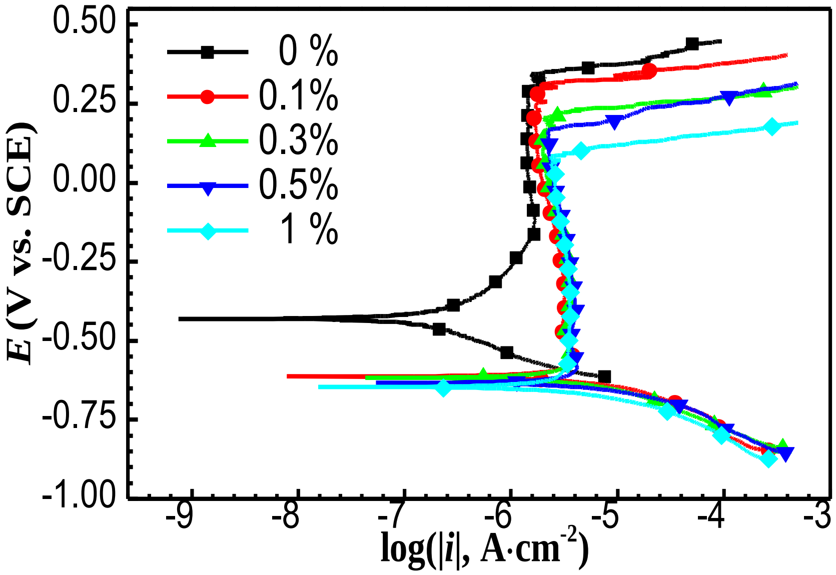


Figure 1. Potentiodynamic polarization curves of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°C.

Table 1. The electrochemical parameters of the orthodontic band in the artificial saliva with different concentrations of NaF at 37℃

Concentration Wt.%	E_{corr} mV	E_{pit} mV	b_a mV/dec	b_c mV/dec	i_{corr} uA·cm^{-2}
0.0	-430±6	343±7	212±3	-106±3	0.145±0.008
0.1	-617±5	287±7	63±5	-105±6	1.314±0.057
0.3	-620±3	196±5	61±2	-109±5	2.825±0.073
0.5	-635±4	156±7	62±4	-107±2	3.133±0.121
1.0	-647±4	83±3	63±4	-108±3	3.523±0.139

4.2 EIS

EIS of the bands were perfomed in solution with different NaF solution immersed for 1h. The plots of Nyquist and Bode immersed with different concentrations of NaF in the artificial saliva at 37℃ are given in Fig.2. For the Nyquist plots (Fig. 2(a)), with the increase of the concentration of NaF in the artificial saliva, the semi-circular arc radius decreases siginicantly, indicating that the corrosion resistance of bands is weaken. As shown in Fig. 2(b), with the increase of concentrations of NaF, the phase angle gradually moves to the higher frequency, the impedance modulus decreases.

The EIS best fitting parameters are shown in Table 2. R_s is the solution resistance; R_p is the polarization resistance; Q_{dl} is on behalf of double layer membrance capacitance and n is the deviation parameter of fitting results compared with the ideal state of capacitance. There were significant differences in all groups for R_s and R_p ($P<0.05$). Significant differences were found between Group 5 and the other groups for Q_{dl} and n.

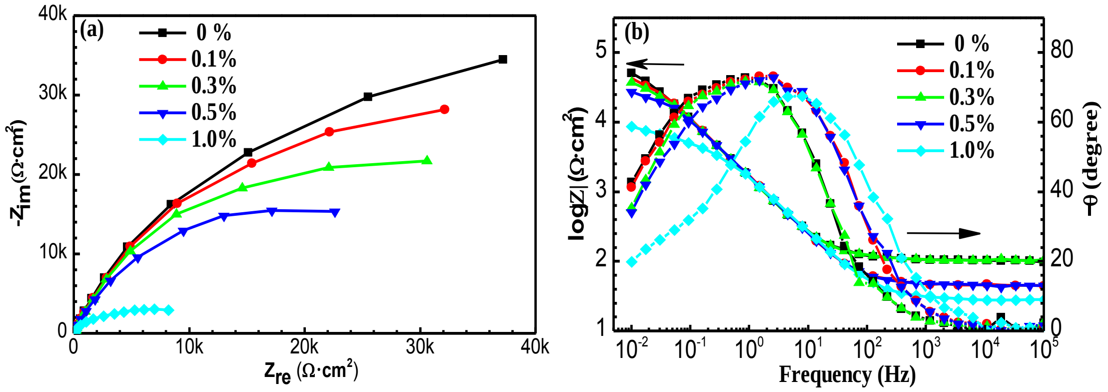


Figure 2. Nyquist plots (a) and Bode diagrams (b) of the orthodontic band in the artificial saliva with different concentrations of NaF at 37℃.

Table 2. EIS best fitting parameters results of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°C.

NaF wt.%	R_s, Ω·cm^2	Q_{dl}, uF·cm^{-2}·S^{n-1}	n	R_p, kΩ·cm^2
0	105.30±0.26	128.2±3.9	0.85±0.01	84.85±3.50
0.1	98.73±0.34	131.2±6.2	0.84±0.02	69.79±5.30
0.3	75.41±0.57	132.1±3.8	0.85±0.01	58.04±5.53
0.5	45.53±0.39	133.8±5.7	0.83±0.02	40.36±4.94
1.0	27.83±0.45	145.5±7.3	0.79±0.03	7.95±3.43

4.3 Capacitance results

Fig.3 displays the Mott-Schottky results for the paaive films formed on the orthodontic band at different NaF concentrations within the passive regions. The bands show only n-type behavior. Moreover, the values of N_d increase with the increasing concentrations of NaF.

Table 3 shows the calculated carrier density (N_d) values of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°C. N is charge carrier density, and n represents the type of semiconductor. There were significant differences in all groups for carrier density($P<0.05$).

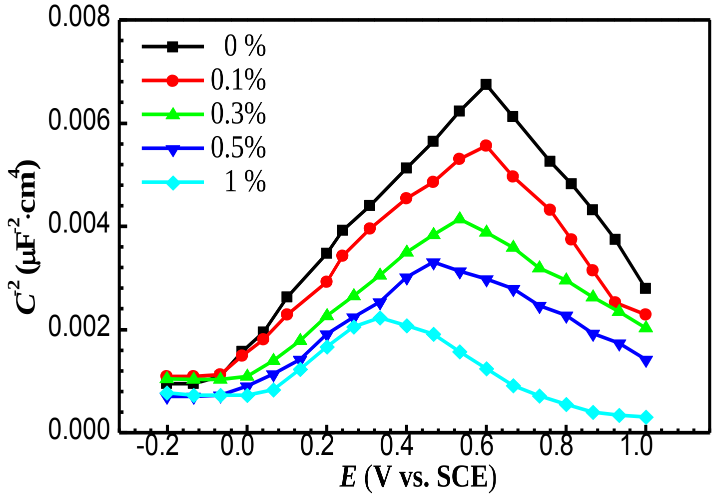


Figure 3. Mott-Schottky plots of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°C.

Table 3. Semiconductor type and carrier density of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°C.

Concentration wt.%	Semiconductor type	Carrier density, (Nd)/10^{21}cm^{-3}
0	n	1.17±0.05
0.1	n	1.44±0.10
0.3	n	1.58±0.07
0.5	n	1.82±0.13
1.0	n	2.03±0.05

4.4 Corrosion mophology

The images of the band specimens immersed in the artificial saliva with different concentrations of NaF under static conditions at 37℃ for 30 days are shown in Fig. 4. At the absence of NaF, there is no obviously corrosion pits on the orthodontic band specimen, As seen in Fig.4a. At the low concentration of NaF of 0.1wt.% (Fig.4(b)), the orthodontic band mainly experiences a uniform corrosion, with slight pit corrsion. As the concentration of NaF continues to increase, corrosion becomes more localized (Fig. 4(c)-(e)). Since the passive film of orthodontic band is further destroyed and corrosion pits are generated locally.

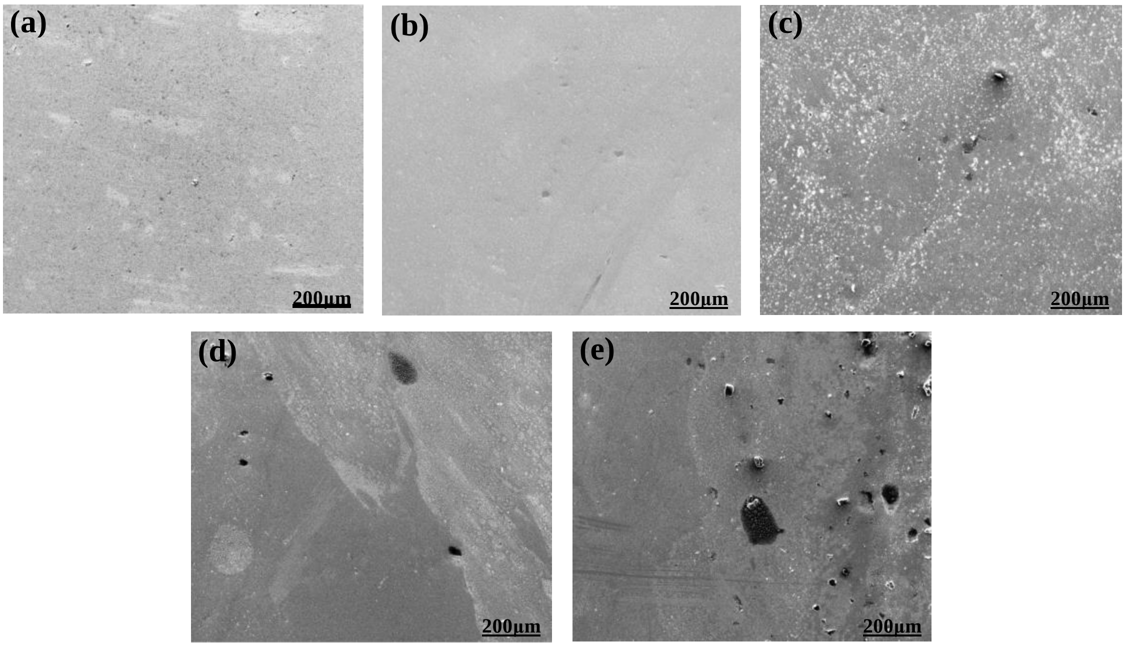
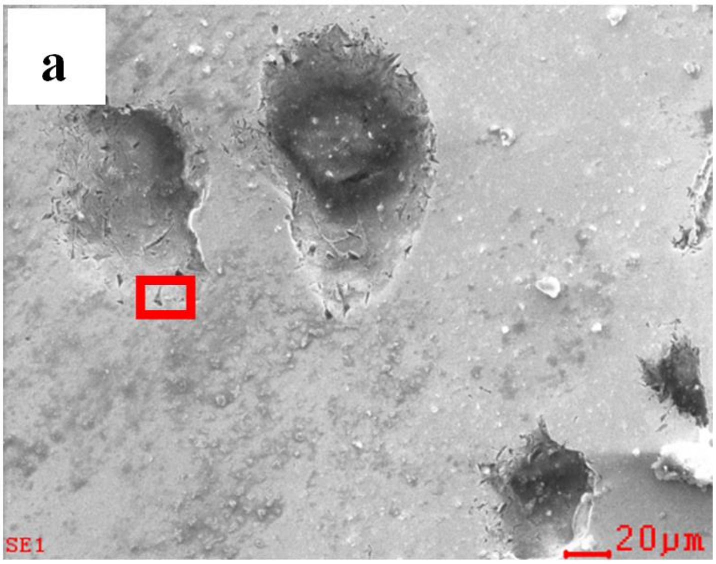


Figure 4. Corrosion morphology of the orthodontic band in the artificial saliva with different concentrations of NaF at 37°Cfor 30 days. (a) 0 wt.%NaF, (b) 0.1 wt.% NaF, (c) 0.3 wt.% NaF, (d) 0.5 wt.% NaF, (e) 1 wt.% NaF.



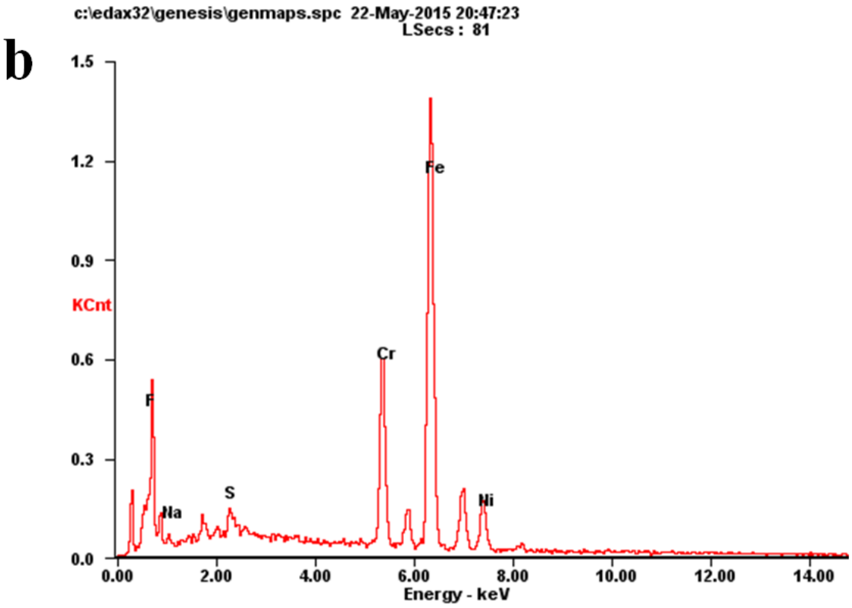


Figure 5. Corrosion morphology and EDS of the orthodontic band in the artificial saliva with 1wt.%NaF at 37℃ for 30 days. (a) corrosion morphology, (b) Energy spectrum.

Fig. 5a shows the magnifying localized corrosion SEM image of orthodontic band immersing in the artificial saliva with 1wt.% NaF under static conditions at 37℃ for 30 days. It can be seen that the oval pits formation on the orthodontic band. After energy spectrum analysis of the pitting corrosion site shown in Fig.5b, it can be seen that a large number of fluoride elements on the pitting site. The composition of elements of pitting corrosion site is shown in Table 4, where fluorine content is 7.21wt.%.

Table 4. EDS of the orthodontic band in the artificial saliva with 1wt.% NaF at 37°C for 30 days.

Element. %	wt.%	At.%
F	07.21	18.07
Na	01.33	02.74
S	01.00	01.48
Cr	17.14	15.69
Fe	62.24	53.04
Ni	11.07	08.98

5. DISCUSSION

5.1. Potentiodynamic polarization studies

The polarization curves offer some significant features concerning the corrosion behaviour of orthodontic band in artificial saliva. Several studies have represented the corrosion resistence of orthodontic mental materials by means of potentiodynamic curves. Bakhtari et al. [25] discussed the potentiodynamic curves of different wires and brackets with regard to corrosion current. Rondelli et al

[26] evaluated the susceptibility to pitting of stainless steel wire and Ni-Ti wire, and found generally no pitting occurred up to high potentials. As observed in Fig.1 , the band displays typical passive behavior in artificial saliva at different concentrations of NaF. This behaviour typically indicates that a passive film is formed on the specimen surface. The E_{corr} is the potential where the total of all cathodic reaction is equal to the sum of all anodic reactions on the specimen surface. With the increase of concentration NaF, the corrosion potential decreased gradually.The relationship between net current density i and polarisation potential E should follow the B-V equation, that is，

$$i = i_{corr}\left[\exp\left(2.303\frac{\Delta E}{b_a}\right) - \exp\left(-2.303\frac{\Delta E}{b_c}\right)\right] \qquad (1)$$

$$\Delta E = E - E_{corr} \qquad (2)$$

They (i_{corr}, b_{a} and b_{c}), which show the corrosion behaviours and kinetics [27]. Tafel extrapolation method were used to fitting the electrochemical parameters of the orthodontic band in the artificial saliva with different concentrations of NaF at 37℃.

Table 1 shows that with a fresh surface, the anodic Tafel slope (b_{a}) of the the orthodontic bands is positive than absolute value of cathodic Tafel slope (b_{c}) without adding NaF, thus indicating anode is the controlling step of the reaction. With the addition of NaF, the anodic Tafel slope(b_{a}) is negative than absolute value of cathodic Tafel slope (b_{c}) that means cathode is the controlling step of the reaction. The mechanism of the reaction has been changed. With the increase of concentration of NaF, the corrosion current density increases, the corrosion rate of orthodontic band increases. When the electrode potential is raised to a certain value, the current density increases rapidly, which has been difined as pitting potential. With the increase of concentration of NaF, the pitting potential was significantly negative shifted. The O on the passive film could be squeezed out by F^- and the pitting corrsion were induced [14].

5.2 EIS

The EIS diagrams of bands immersed for different fluoride ions solution are displayed in Fig. 2; the left side is the Nyquist diagram, while the right is the corresponding Bode diagram. The radius of capacitive loop is diminished gradually as increase of concentration of NaF. Therefore, the corrosion resistance of the passive film is reduced. The equivalent circuit in Fig. 6 is established to fit the parameters of EIS in Fig. 2 [28,29]. R_{s} is the solution resistance from the reference electrode to the working electrode, R_{p} consists of passive film resistance R_{f} and charge transfer resistance R_{ct}, Q_{dl} is the constant phase angle element of the electric double-layer capacitor. The impedance values of the equivalent circuit in Fig.6 can be written as:

$$Z = R_s + \frac{1}{Z_{Q_{dl}} + \frac{1}{R_p}} \qquad (3)$$

In general, the capacitor of the electric double layer cannot be considered as the ideal capacitive element because of the roughness of the electrode surface, the impedance of which is always concerned with the angular frequency of the pumping signals and conveyed in the following form [28-30]:

$$Z_{Q_{dl}}^{-1} = Q_{dl}(j\omega)^{n} \tag{4}$$

where ω is the angular frequency, j is the symbol for imaginaries, and n represents the index of the constant phase angle element ranging from 0.5 to 1. The element is equivalent to a capacitor when $n = 1$ [31].

Tables 2 shows the best fitting value parameters in the EIS diagram of bands through the ZsimpWin software. The orthodontic band without F^- has the highest R_p value. In addition, the capacitance values of the double-layer capacitor decrease with the increasing concentration t of F^-. The result indicates that F^- is harmful for corrosion resistance. The electric double layer ,C, can be calculated by the following formula [32]:

$$C = \frac{(QR)^{1/n}}{R} \tag{5}$$

where the Q is a constant phase angle of the electric double layer, and n is the deviation parameter of fitting results compared with the ideal state of capacitance, R is the passive film resistance. Then the passive film thickness can be estimated [32-34]:

$$d_{film} = \frac{\varepsilon\varepsilon_0 A}{C} \tag{6}$$

where ε_0 represents for permittivity of vacuum, A for effective area, ε for dielectric constant, and D_{film} for the thickness of passive film which can be qualitatively described with Q_{dl}. From the result of Table 2, with the increasing of the concentration of NaF in the artificial saliva, the value of Q_{dl} increase, which means the thickness of the passive film decreasing, the corrosion resistance of the passive film.

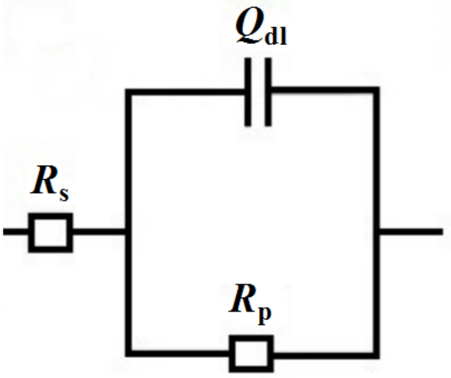


Figure 6. Equivalent circuit applied to analyze the measured EIS results.

5.3 Capacitance results

Semiconductor properties of passive film can be characterized with the following Mott-Schottky theory [34,35]:

$$\frac{1}{C^2} = \pm\frac{2}{e\cdot N\cdot\varepsilon\cdot\varepsilon_0}(E - E_{fb} - \frac{kT}{e}) \tag{7}$$

wherein: *C*—space charge capacitance;
N— charge carrier density;
ε—semiconductor dielectric constant valued as relative dielectric constant;
ε_0—permittivity vacuum and $\varepsilon_0=8.85\times10^{-12}F/m$;
E—electrode potential;
E_{fb}—flat band potential;
K—Boltzmann's constant and $k=1.38\times10^{-23}J/K$;
e—electron charge and $e=1.602\times10^{-19}C$;
T—Kelvin Temperature, *K*.

Fig. 3 shows the Mott–Schottky plots of the film formed on orthodontic bands at different NaF concentrations solutions. Obviously, the curves are divided into two regions in the light of membrane formation potential. For potential from -200mV to +300mV, the Mott–Schottky plots of orthodontic band show linear relationship with a positive slope, which means that the passive films behave as an n-type semiconductor. In addition, for the potential more postive than 600mV, the slope of the plots turn to negative. This is due to the enhancement of the conductivity of the oxides, resulting in the conversion of passive film from N type to the P type [36]. The concentration of the electron donor, N_d , can be calculated from the positive slope of the C^{-2} vs. *E* curves [34-36].

$$N=\frac{2}{m\cdot e\cdot\varepsilon\cdot\varepsilon_0} \qquad (8)$$

where m is the slope of the Mott–Schottky plots in the region of Linear; *e* is the electron charge, $e=1.602\times10^{-19}C$; ε_0, the vacuum permittivity, $\varepsilon_0=8.85\times10^{-12}F/m$; ε, the relative dielectric constant of the semiconductor. It can be seen in Table 3, the defects density values orders of magnitude are around $10^{21}cm^{-3}$. Moreover, the values of N_d increases with the increasing concentrations of NaF. Thus, passive films of orthodontic band formed at lower concentrations of NaF have better stability, which is consistent with the measurement results of EIS.

5.4. Corrosion morphology

The degree of corrosion can be directly reflected by the surface morphology. Huang [37] ever used SEM to analyze the surface characterizations of the NiTi wires in artificial saliva of various degrees of acidity and considered the differences in the corrosion resistance among NiTi dental orthodontic archwires did not correspond with the surface roughness and pre-existing defects. In our investigation, similarly，SEM was employed to observe the surface morphology of as-received bands after the immersing test. Qualitative surface topography of orthodontic bands is affected by exposure to high fluoride concentration agents, since the passive film of orthodontic band is further destroyed and corrosion pits are generated locally. However, the friction between the archwire and the tube of orthodontic band will increase after exposureing to sodium fluoride solution for a long time, because corrosion increases the surface roughness of the band. The EDS analysis of the pitting corrosion site shows that fluoride is involved in the formation of pitting corrosion and consistents with the results of previous electrochemical tests.

Over the years, several studies have evaluated the corrosion resistance of orthodontic metal materials in the fluorine-containing artificial savila in vitro, but our research substantially differs from those in terms of materials tested and systematic methodology. Lee et al. [18] considered that the fluoride-related degradation of unloading mechanical properties could contribute to prolonged orthodontic treatment.

Barros et al. [19] also found that commercial titanium orthodontic have high corrosion sensitivity in artificial saliva, especially in high concentration fluoride (0.5wt.% sodium fluoride), and the anodic current density, corrosion rate increased and breakdown potential decreased. Another study confirmed that more pits and cracks formed on Titanium alloy miniscrew implants surface after immersion in 0.2wt.% NaF solutions compare to in 0.1wt.% NaF in laboratory immersion test [22]. Although fluoride-containing dental products are effective for prevention of caries, the amount and the frequency of using fluorine-containing products should be considered so as not to cause the severe orthodontic metal materials corrosion.

It is a proven fact that corrosion of orthodontic appliances occurs, however, the disadvantage of corrosion on orthodontic treatment and on patient's health is still not fully stated. It would be necessary to conduct additional assays and test the effect of buccal cell on corrosion.

6. CONCLUSIONS

(1) The corrosion mechanism of orthodontic band in artificial saliva could be changed by fluoride, with the increase of concentration of fluoride, corrosion current density increased, corrosion potiential, pitting potiential and the impedance of passive film decreased.

(2) In artificial saliva with different concentrations of NaF, the passive film of orthodontic bands showed only n-type behavior. Passive films of the bands formed at high concentrations of NaF had more disordered structures and higher defect densities.

(3) The pitting corrosion could be observed on the orthodontic band in artificial saliva with different concentration of NaF for 30 days immersion, the higher content of NaF was, the severer pitting corrosion appeared. Fluorine ion participated in the development of corrosion.

(4) Considering variety of fluorine-containing products are being used in daily life, the amount and the frequency of using fluorine-containing products should be considered so as not to cause the severe orthodontic band corrosion.

CONFLICTS OF INTEREST
The authors have no conflicts of interest relevant to this article.

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