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## Electrochemical behaviour in corrosion and galvanic corrosion of several alloys used in fixed partial dentures when immersed in a Fusayama saliva

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### ABSTRACT

The framework which supports ceramic in fixed partial denture generally includes several dental alloys in electrical contact (post-solder alloy in the joint between two pieces of a same parent alloy). In service, some parts of the framework can be exposed to saliva and then to individual or galvanic corrosion, with possible detrimental consequences for their mechanical resistance on long times. Eight parent alloys and four post-solder alloys were prepared as electrodes and tested in an aerated Fusayama-type artificial saliva. Electrochemical techniques were applied first to specify the individual behaviour of all alloys (follow-up of corrosion potential, determination of polarization resistance, Tafel experiment and calculations), and second to study the galvanic behaviour of each parent alloy coupled with its usual post-solder during four hours. Corrosion potentials were generally very high and corresponded to the immunity domains of the noblest elements or to the passive state for the other elements, excepted other elements fortunately present in low quantities in the alloys. Polarization resistance were high or very high. Tafel experiments allowed verifying that corrosion currents were also very low. Because of different individual behaviours a galvanic current may exist between coupled parent alloy and post-solder alloy.

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### KEYWORDS

Fixed partial denture;  
Parent alloy;  
Post-solder alloy;  
Electrochemical  
measurements;  
Galvanic coupling;  
Fusayama saliva.

### INTRODUCTION

In a fixed partial denture, the pre-soldered parts (joint thereafter entirely covered by cosmetic ceramic) are joined again by soldering their extremities (constituted by alloys called “parent alloys”) using

another alloy (called “post-solder alloy”) in order to obtain the final framework supporting the prosthesis (Figure 1). In some cases it is possible that a parent alloy is not wholly covered by ceramic, and then it can be in contact with saliva simultaneously with the post-solder joint. The two types of alloys can display different

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electrochemical behaviours, and then the {parent + solder} assemblage of alloys can endure galvanic corrosion. After long times of exposure to saliva, this can lead to a weakening of framework, especially when the surface of the alloy playing the anodic role is smaller than the surface of the other alloy.

The aim of this work is, for several usual couples of parent alloy and post-solder alloy, to more precisely characterize, in an artificial saliva, the separated behaviours of the two alloys by applying the Tafel method<sup>[1,2]</sup> in order to specify their corrosion potential  $E_{\text{corr}}$  (and also the corrosion current density  $I_{\text{corr}}$ ). This will also allow identifying which alloy may act as anode and which alloy may act as cathode in the assemblage, then how a possible galvanic corrosion can occur between a parent alloy coupled with its post-solder alloy, which will be thereafter directly studied.

### EXPERIMENTAL

#### Elaboration of the alloys and preparation of the electrodes

The alloys of this study are the same as the ones which were already studied in a previous work<sup>[3]</sup> concerning the metallographic characterization of soldered alloys for fixed partial dentures' frameworks. These alloys are firstly eight parent alloys: five alloys rich in noble elements Au+Pt+Pd > 60wt.% (called "High Noble"), one alloy with a not so high total content in noble elements (called "Noble") and two

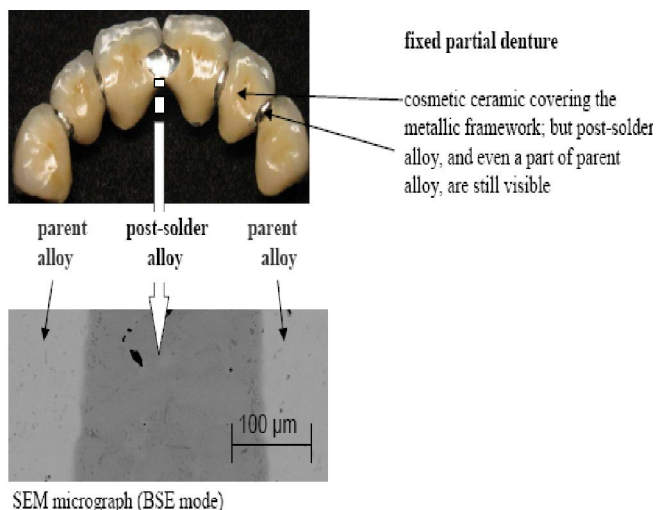
alloys based on both Ni and Cr (called "Predominantly Base"), all designations being conform with the Identallloy® norm. Four post-solder alloys were also studied. The chemical compositions of all alloys (data from the manufacturer: Ivoclar Vivadent®) are reminded<sup>[3]</sup> in TABLE 1 for the "High Noble", the "Noble" alloy and the two "Predominantly Base" alloys, and in TABLE 2 for the four post-solder alloys.

The parent alloys were investment cast to obtain parallelepipedic ingots (10×10×1 mm<sup>3</sup>). These ones were cut into four parts (5×5×1 mm<sup>3</sup>). The post-solder alloys were obtained by melting, in a crucible, a solder rod heated using a gas-oxygen torch. The obtained ingots were half ball-like and their weights were about 1 gram). The parent alloy and the post-solder alloy each underwent heat-treatments (details already given in a previous article<sup>[3]</sup>) which reproduce what it is done in practice. The electrodes were realized by tin-soldering with an electrical wire, embedding in a cold resin, polishing until a mirror-like surface state is obtained. The area of emerging alloy, which was thereafter in contact with the electrolyte, was then a square of about 25 mm<sup>2</sup> in the case of a parent alloy and a disk of around 35 mm<sup>2</sup> for a post-solder alloy. The microstructure of the metallic part of all the electrodes was controlled using a Scanning Electron Microscope (SEM Philips XL30) in Back Scattered Electrons mode (BSE) with a 20kV acceleration voltage.

#### Electrochemical experiments

All electrochemical runs were performed using a potentiostat / galvanostat model 263A of Princeton Applied Research. This one was driven by the software M352 of EGG/Princeton. The electrolyte considered in this work was a Fusayama-type solution (composition given in TABLE 3), the pH of which was rated at 2.3 with lactic acid (concentration: 90%). This is an artificial saliva which was often used for the characterization, by electrochemical techniques, of various dental metals and alloys: Au-based<sup>[4-7]</sup>, Pt-based<sup>[7]</sup> and Pd-based<sup>[4, 6, 8]</sup>, and also based on less noble elements: Ag<sup>[4, 6, 9]</sup>, Hg<sup>[4]</sup>, Ni or Co<sup>[4, 6, 9-11]</sup>. Its temperature was maintained at 37°C (human body) using a Julabo F32 device.

In the electrochemical cell the studied alloy played the role of Working Electrode. The Counter Electrode was a platinum disk-like electrode and the reference in



**Figure 1 :** A fixed partial denture (top), and a microscope view of its microstructure in the post-soldered region

TABLE 1 : Chemical compositions of the eight parent alloys (in wt.%; manufacturer's data)

elements	Au	Pt	Pd	Ag	Ga	In	Re	Ru	Sn	Zn	others
<b>"High Noble" alloys</b>											
IPS dSIGN98	85.9	12.1	-	-	-	<1.0	-	-	-	2.0	In<1 Ir<1 Fe<1 Mn<1 Ta<1
IPS dSIGN91	60.0	-	30.6	-	1.0	8.4	<1.0	<1.0	-	-	-
Aquarius Hard	86.1	8.5	2.6	-	-	1.4	-	<1.0	-	-	Fe<1 Li<1 Ta<1
Lodestar	51.5	-	38.5	-	1.5	8.5	<1.0	<1.0	-	-	-
W	54.0	-	26.4	15.5	-	1.5	<1.0	<1.0	2.5	-	Li<1
<b>"Noble" alloy</b>											
IPS dSIGN59	-	<1.0	59.2	27.9	-	2.7	<1.0	<1.0	8.2	1.3	Li<1
elements	Ni	Cr	Mo	Al	Si	W	others				
<b>"Predominantly Base" alloys (containing less than 25wt.% Au+Pt+Pd)*</b>											
Pisces Plus		61.5	22.0	-	2.3	2.6	11.2	Mischmetal<1			
4all		61.4	25.7	11.0	<1	1.5		Mn<1			

TABLE 2 : Compositions of the four studied post-solder alloys (in wt.%; manufacturer's data)

elements	Au	Ag	Cu	Ga	In	Sn	Zn	to use with
.585 Fine solder	58.5	16.0	18.0	7.2	-	-	<1.0	dSIGN 98
.615 Fine Solder	61.3	13.1	17.4	-	7.6	-	<1.0	dSIGN91, Lodestar, dSIGN59,
.650 Gold Solder	65.0	13	19.6	2.0	-	-	<1.0	Aquarius Hard
LFWG	56.1	27.4	-	-	<1.0	<1.0	15.8	W, 4all, Pisces Plus

TABLE 3 : Composition of the Fusayama saliva

Product	Concentration (g/L)
KCl	0.4
NaCl	0.4
CaCl <sub>2</sub> , 2H <sub>2</sub> O	0.906
NaH <sub>2</sub> PO <sub>4</sub> , 2H <sub>2</sub> O	0.690
Na <sub>2</sub> S, 9H <sub>2</sub> O	0.005
urea	1

potential was given by a Saturated Calomel Electrode (241.5mV/Normal Hydrogen Electrode).

Two types of experiments were performed with this apparatus:

- ❖ immersion during about 2 hours with recording of the free potential and two measurements of the polarization resistance (Rp) after each hour,
- ❖ Tafel experiment: increase in potential from  $E_{\text{corr}} - 250\text{mV}$  up to  $E_{\text{corr}} + 250\text{mV}$  (rate of  $10\text{mV min}^{-1}$ ) with determination of  $E_{\text{corr}}$ ,  $I_{\text{corr}}$ ,  $\beta_a$  and  $\beta_c$  (i.e. the anodic and cathodic Tafel coefficients).

Galvanic corrosion experiments for each parent alloy coupled with the post-solder alloy recommended by the manufacturer, were performed with recording both the common potential of the two alloys and the exchange current between them (duration: 4 hours).

The results were thereafter interpreted using the Pourbaix's diagrams corresponding to the major elements belonging to the chemical compositions of the alloys, for pH=2.3.

## RESULTS AND DISCUSSION

### Follow-up of the corrosion potential after immersion

During the {2 hours}-period before Tafel experiment, the corrosion potential ( $E_{\text{corr}}$ ) is generally variable, as illustrated in Figure 2 in the cases of an alloy and its usual post-solder (Aquarius Hard and .650). As shown in TABLE 4, during the first hour it increases (Aquarius Hard, dSIGN91, Pisces Plus, .650) or decreases (W, 4all, LFWG) or is already almost constant (dSIGN98, Lodestar, dSIGN59, .585, .615). During the second hour, the corrosion potential seems generally more or less stabilized, in all cases. In the Pourbaix diagrams drawn for the condition 1Mol/L of dissolved species (about the frontiers between immunity, passivation or corrosion domains), and considered for each element for pH=2.3, it appears that these corrosion potentials correspond, for the whole {2 hours}-period,

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to the immunity domains of all the noblest elements (Au, Pt, Pd and Ag), to the passive states of most of the other elements (Cr, Ga, Sn, Mo, W), but also to corrosion of Ni ( $\text{Ni}^{++}$ ), Zn ( $\text{Zn}^{++}$ ) and In ( $\text{In}^{+++}$ ). In the case of copper, the corrosion potential of the alloys which contain this element (all are post-solder alloys: .585, .615 and .650) seems stabilized at the  $\text{Cu}/\text{Cu}^{++}$  equilibrium. The values of the polarization resistances which were measured one time per hour, are of a high level since they are all higher than  $1\text{ k}\Omega \times \text{cm}^2$ . The ones measured for the noblest alloys are especially high, which confirms their high nobility.

### Tafel experiments

All the Tafel curves are displayed in Figure 3 and in Figure 4, with association in a same graph of the curve of a post-solder alloy and the curve (or curves) of the parent alloy (or the parent alloys) to which this solder alloy is destined according to the manufacturer. On these curves it can be firstly observed that the corrosion potential is more or less close to the one measured just before Tafel experiment: for most of them the difference is lower than 50mV. In contrast, the differences are

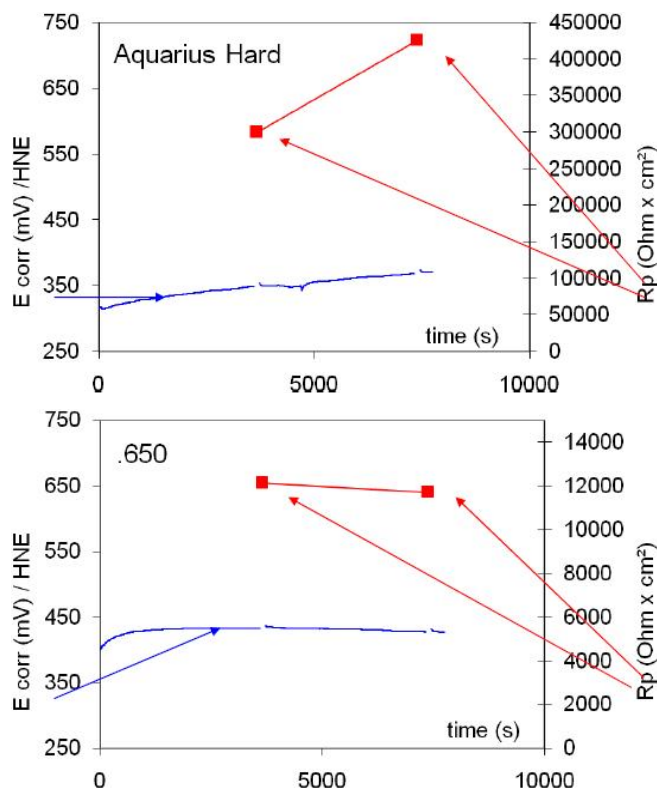


Figure 2 : Examples of variation of  $E_{\text{corr}}$  and  $R_p$  during the 2 hours of immersion before Tafel experiment (here: the parent alloy “Aquarius Hard” and its usual post-solder “.650”)

significantly greater for .615 and for .650. Tafel calculations were often difficult to perform, even impossible in several cases, since the cathodic part and the anodic part were not straight on a length great enough. Nevertheless these calculations were attempted and brought the values of  $E_{\text{corr}}$  which were already commented, but also of  $I_{\text{corr}}$ ,  $\beta_a$  and  $\beta_c$  which must be considered with caution (TABLE 5). The values of the corrosion current density  $I_{\text{corr}}$  is generally extremely low, since most of the determined values are lower than  $1\mu\text{A}/\text{cm}^2$ , and the highest  $I_{\text{corr}}$  values which were obtained

Table 4: Corrosion potentials at the beginning ( $t=0$ ), in the middle ( $t=1\text{h}$ ) and at the end ( $t=2\text{h}$ ) of the 2-hours period before Tafel experiment and comparison with the theoretical states (at  $\text{pH}=2.3$  and for a corrosion criterion being  $1\text{Mol/L}$  of corresponding metallic ion) of all elements belonging to the alloys according to their Pourbaix diagrams; values of the polarization resistances at  $t=1\text{h}$  and  $t=2\text{h}$

Parent alloys	$E_{\text{corr}}/\text{NHE}$ (mV)			$R_p$ ( $\text{k}\Omega \times \text{cm}^2$ )	
	0h	1h	2h	$R_p$ (1h)	$R_p$ (2h)
dSIGN98	+240	+242	+242	85	91
	Au <sup>0</sup> Pt <sup>0</sup> Zn <sup>++</sup>				
Aquarius Hard	+317	+349	+369	300	426
	Au <sup>0</sup> Pt <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup>				
dSIGN91	+406	+455	+448	89	107
	Au <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup> Ga <sub>2</sub> O <sub>3</sub>				
Lodestar	+394	+396	+395	207	232
	Au <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup> Ga <sub>2</sub> O <sub>3</sub>				
W	+299	+208	+173	80	79
	Au <sup>0</sup> Pd <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Sn(OH) <sub>4</sub>				
dSIGN59	+31	+18	+11	4.8	5.2
	Pd <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Sn(OH) <sub>2</sub> /Sn(OH) <sub>4</sub> Zn <sup>++</sup>				
4ALL	+31	+58	+67	62	136
	Ni <sup>++</sup> Cr(OH) <sub>3</sub> MoO <sub>2</sub>				
Pisces Plus	+14	+215	+217	120	148
	Ni <sup>++</sup> Cr(OH) <sub>3</sub> WO <sub>3</sub>				
Postsolder alloys	$E_{\text{corr}}/\text{NHE}$ (mV)			$R_p$ ( $\text{k}\Omega \times \text{cm}^2$ )	
	0h	1h	2h	$R_p$ (1h)	$R_p$ (2h)
.585	+333	+321	+321	0.9	0.9
	Au <sup>0</sup> Ag <sup>0</sup> Ga <sub>2</sub> O <sub>3</sub> Cu <sup>0</sup> /Cu <sup>++</sup>				
.615	+305	+296	+289	4.8	5.0
	Au <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Cu <sup>0</sup> /Cu <sup>++</sup>				
.650	+401	+433	+427	12.1	11.7
	Au <sup>0</sup> Ag <sup>0</sup> Ga <sub>2</sub> O <sub>3</sub> Cu <sup>0</sup> /Cu <sup>++</sup>				
LFWG	+267	+101	+109	9.1	10.1
	Au <sup>0</sup> Ag <sup>0</sup> Zn <sup>++</sup>				



(essentially for the post-solder alloys), are lower than  $0.01\text{mA}/\text{cm}^2$ . The Tafel coefficients are not easy to comment in most cases, especially the anodic one. The cathodic coefficient can give some indications about the oxidant species implied in the cathodic reactions. These ones can be dissolved oxygen for the lowest values of  $\beta_c$  (near  $60\text{mV}/\text{decade}$ ), and  $\text{H}^+$  for its highest values

(near  $120\text{mV}/\text{decade}$ ) notably since the Fusayama solution of this study is a little acid ( $\text{pH}=2.3$ ).

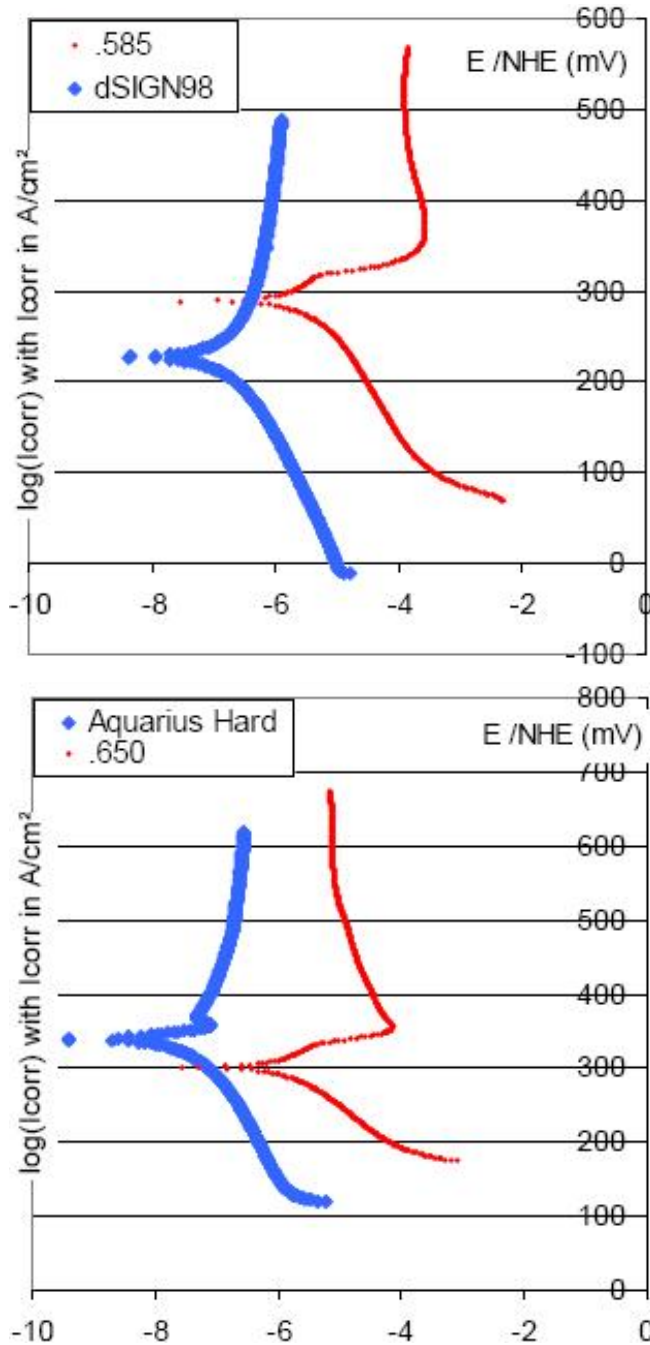


Figure 3 : Tafel curves obtained for the parent alloy dSIGN98 (respectively Aquarius Hard) and for its usual post-solder .585 (resp. .650) presented together in the top (resp. bottom) graph

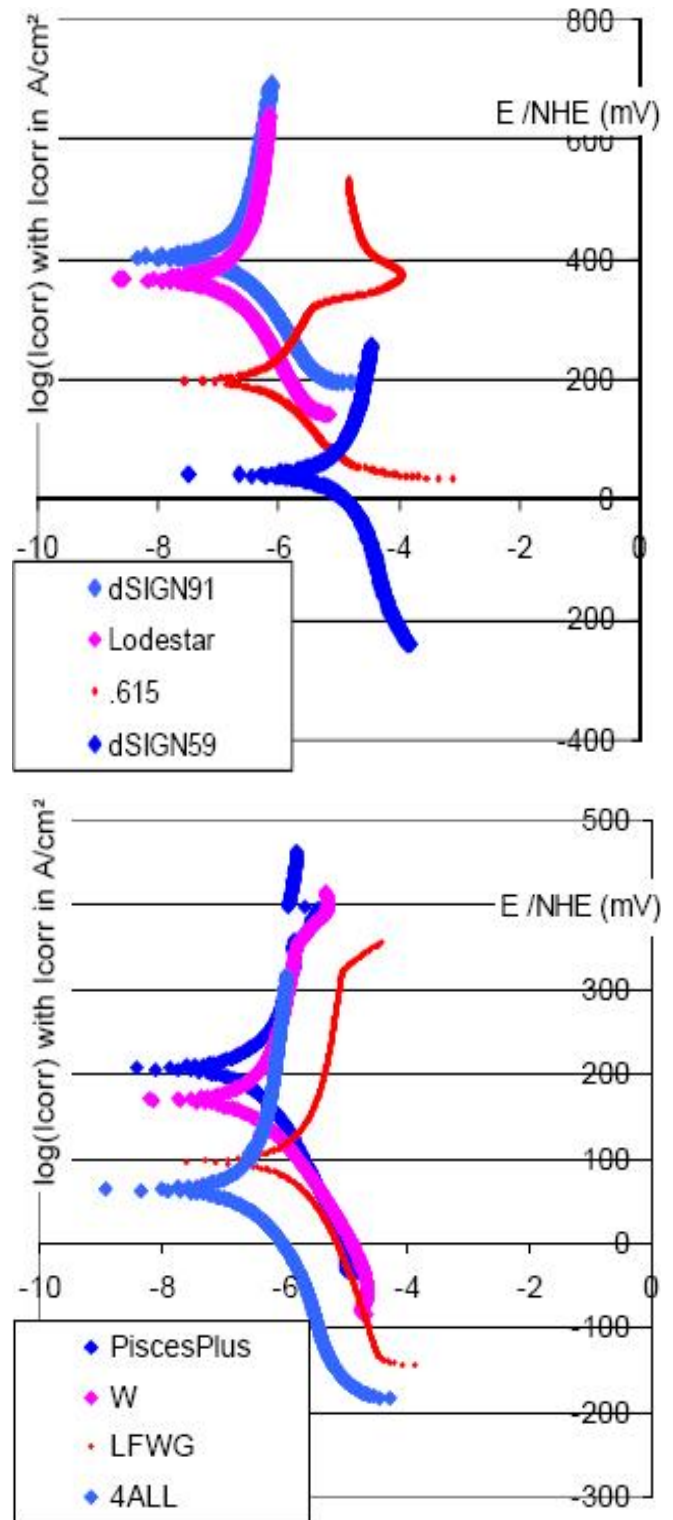


Figure 4 : Tafel curves obtained for the parent alloys dSIGN91, Lodestar and dSIGN59 (respectively W, 4all and Pisces Plus) and for their usual post-solder .615 (resp. .LFWG) presented together in the top (resp. bottom) graph

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Table 5: Corrosion potentials ( $E_{corr}$ ) and corrosion current densities ( $I_{corr}$ ) obtained after Tafel experiments and calculations, and comparison with the theoretical states (at pH=2.3 and for a corrosion criterion being 1Mol/L of corresponding metallic ion) of all elements belonging to the alloys according to their Pourbaix diagrams; values of the Tafel coefficients (anodic and cathodic)

Parent alloys	$E_{corr}$ / NHE (mV)	$I_{corr}$ / NHE (nA/cm <sup>2</sup> )	Beta (mV/decade)	
			$\beta_a$	$\beta_c$
dSIGN98	+226	342	373	155
	Au <sup>0</sup> Pt <sup>0</sup> Zn <sup>++</sup>			
Aquarius Hard	+334	97	279	183
	Au <sup>0</sup> Pt <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup>			
dSIGN91	+405	416	Too high	180
	Au <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup> Ga <sub>2</sub> O <sub>3</sub>			
Lodestar	+368	391	638	262
	Au <sup>0</sup> Pd <sup>0</sup> In <sup>+++</sup> Ga <sub>2</sub> O <sub>3</sub>			
W	+170	562	386	123
	Au <sup>0</sup> Pd <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Sn(OH) <sub>4</sub>			
dSIGN59	+42	28029	Too high	273
	Pd <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Sn(OH) <sub>2</sub> /Sn(OH) <sub>4</sub> Zn <sup>++</sup>			
4ALL	+64	273	232	99
	Ni <sup>++</sup> Cr(OH) <sub>3</sub> MoO <sub>2</sub>			
Pisces Plus	+207	411	164	148
	Ni <sup>++</sup> Cr(OH) <sub>3</sub> WO <sub>3</sub>			
Postsolder alloys	$E_{corr}$ / NHE (mV)	$I_{corr}$ / NHE (nA/cm <sup>2</sup> )	Beta (mV/decade)	
			? a	? c
.585	+289	7959	493	131
	Au <sup>0</sup> Ag <sup>0</sup> Ga <sub>2</sub> O <sub>3</sub> Cu <sup>0</sup> /Cu <sup>++</sup>			
.615	+198	1169	252	146
	Au <sup>0</sup> Ag <sup>0</sup> In <sup>+++</sup> Cu <sup>0</sup>			
.650	+301	1789	87	67
	Au <sup>0</sup> Ag <sup>0</sup> Ga <sub>2</sub> O <sub>3</sub> Cu <sup>0</sup> /Cu <sup>++</sup>			
LFWG	+97	2252	237	163
	Au <sup>0</sup> Ag <sup>0</sup> Zn <sup>++</sup>			

Galvanic couplings

All couples {parent alloy; its usual post-solder alloy} were electrically coupled and immersed for 4 hours in the Fusayama saliva at 37°C. The common potential of the couple and the exchange current between the parent alloy and its post-solder alloy were recorded. Three selected results are displayed in Figure 5, in which the potentials of the parent alloy and of the post-solder alloy previously measured at the beginning of the 2 hours–

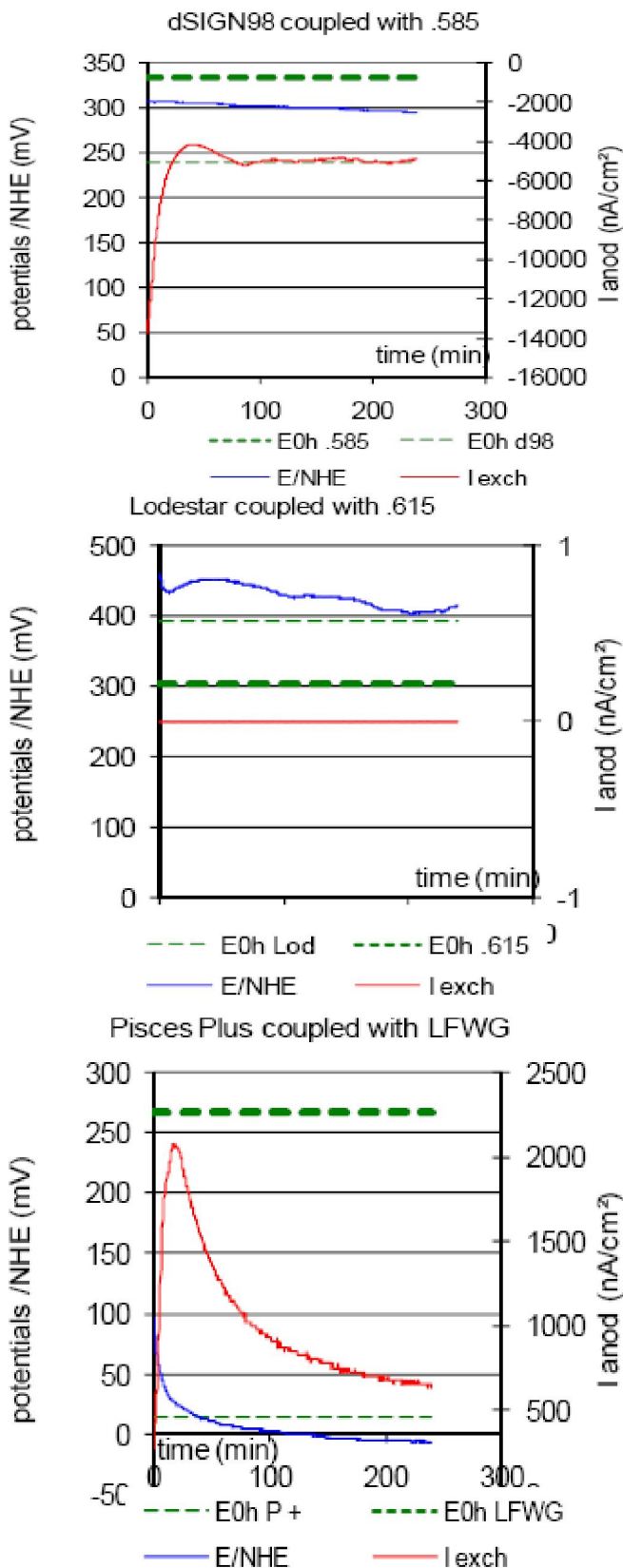


Figure 5 : Galvanic coupling of three parent alloys with their usual post-solder alloys

immersion before Tafel experiment are added for comparison.

In four or five cases the common potential of the {parent alloy + post-solder alloy}-couple is comprised between the individual potentials of the two alloys before the Tafel experiments. It is the case of dSIGN98, Aquarius Hard and dSIGN59 (coupled with their post-solders). It is also almost the case of Lodestar coupled with .615 (potential slightly above the potential interval but decreasing) and of Pisces Plus coupled with LFWG (initially inside the potential interval but slowly decreasing below). In the other cases the common potential is obviously below the interval of the two initial individual values, probably after a fast decrease the end of which is sometimes visible at the beginning of the galvanic coupling experiment. The exchange current is negative only for dSIGN98 coupled with .585 (Figure 5, first graph), is equal to zero for Lodestar coupled with .615 (Figure 5, second graph), and is positive in all the other cases, e.g. Pisces Plus (Figure 5, third graph). This means that dSIGN98 is the single parent alloy in anodic situation when coupled with its post-solder (according to the order of the two initial individual potentials).

The other parent alloys are in cathodic situation. This is consistent with the order of the individual  $E_{\text{corr}}(t=0)$  values in the cases of dSIGN91 coupled with .615 and for W coupled with LFWG. One can consider that there is also almost a good correspondence between the individual  $E_{\text{corr}}(t=0)$  values and the exchange current for the {Lodestar + .615}-couple (exchange current equal to zero, i.e. lower than the limit detectable by the potentiostat), even for the {Aquarius Hard + .650}-couple (exchange current positive but very close to zero). In contrast, the cases of dSIGN59, 4ALL and Pisces Plus are more difficult to explain.

### General commentaries

The parent alloys, as well as the post-solder alloys, are very resistant against corrosion in saliva, here simulated by an aerated slightly acid Fusayama solution. Potentials are generally of a high level and polarization resistance can also reach very high values. This can be explained by the presence of very high quantities of noble elements such as Au or Pd, and by the passivation phenomenon involving most of the other elements.

Chromium probably plays an important role for the protection of the nickel-based alloys, by passivation into a  $\text{Cr}(\text{OH})_3 / \text{Cr}_2\text{O}_3$  layer isolating the alloy (and notably nickel which may be oxidized into dissolved  $\text{Ni}^{++}$  species in such conditions of potential and low pH), as for austenitic stainless steels for example. Since most of alloys contain low or very low contents of not noble elements (except the nickel-based alloys), if corrosion is possible it is extremely limited.

One can then think that these parent or post-solder alloys do not know really significant corrosion and that the different corrosion potentials may correspond to the extremely limited oxidation of the less noble elements (e.g.  $\text{Zn} \rightarrow \text{Zn}^{++} + 2 e$  in dSIGN98 or  $\text{Cu} \rightarrow \text{Cu}^{++} + 2 e$  in the copper-containing post-solder alloys). It is also often possible that these potentials may be related to the slow oxidation-reduction reactions in the solution itself (e.g. dissolved  $\text{O}_2 \leftrightarrow \text{H}_2\text{O}$  and  $\text{H}_2\text{O} / \text{H}^+ \leftrightarrow \text{H}_2$ ) as is to say is simply the potential of the solution as it can be measured using an inert electrode. In the latter case the differences of corrosion potential which can be seen between the tested noble alloys may result from an effect of the different substrates on the rates of these reactions in solvent. This can also influence the values of  $R_p$  before Tafel runs and of  $I_{\text{corr}}$  determined by this later experiment.

The differences of  $E_{\text{corr}}$  values between a parent alloy and its post-solder let think to a possible galvanic corrosion. The galvanic coupling runs led to results which were not all easy to explain, and which were sometimes not in accordance with the corrosion potential hierarchy between the two coupled alloys. It is possible that coupling, by imposing a new potential common to the two alloys, leads in some cases to the classical galvanic corrosion. Analysis of the species present in solution after test may give a better knowledge about the involved elements. But, in the case of the noblest alloys, coupling can involve some changes for the solvent reactions on surface of the alloys ( $\text{O}_2 \leftrightarrow \text{H}_2\text{O} / \text{H}^+ \leftrightarrow \text{H}_2$ ), with new localisations of the reactions between the two alloys, according to the different rates of the reduction system and oxidation system.

### CONCLUSIONS

The alloys for frameworks for fixed partial denture

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which were studied here are extremely corrosion-resistant. Even in an aerated and acid solution reproducing saliva, the electrochemical properties show that the presence of high quantities of noble element or of elements which lead to passivation, allow a good or very good behaviour. The difference of behaviour between assembled alloys may sometimes lead to galvanic current between the two but probably with no or very limited consequences for the mechanical properties neither for the post-solder nor for the parent alloy which is moreover often entirely covered by ceramic.

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