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Breathing New Life into Historical Instruments.
How to Monitor Corrosion

Abstract ‘To play or to display’ is the dilemma that museums have to face, given the increasing trend towards historically informed performance. Brass instruments can suffer corrosion both during and after playing due to the high humidity inside them. To forestall or at least reduce corrosion, drying with a fan has been chosen as a preventive measure. The state of corrosion inside the tuning slides of the instruments was determined with a specially developed electrochemical sensor. The results of the project show that drying with a fan indeed reduces ongoing corrosion, when compared to a group of instruments played without preventive measures that showed an increasing corrosion rate over time.

Introduction Brass, a copper-zinc alloy, has been used to make wind instruments since the 16th century, mainly due to its very good acoustic properties, the ease of manufacturing instruments with it, and its good resistance to corrosion. Historical brass instruments are conserved in museums or collections such as the Burri collection in Bern,[1] which has more than a thousand instruments. The increasing importance of historically informed performance practice (HIP)[2] has resulted in these original instruments being played in concerts. Private and public instrument collections and museums are being forced to confront the issues that can be summarised as ‘to play or to display’ – should instruments be displayed only in museum cases, or should they be subjected to ‘normal’ concert use? The overall goal of the project “Brass instruments of the 19th and early 20th centuries between long-term conservation and use in historically informed performance

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The Swiss National Science Foundation (SNF) is acknowledged for financing the project “Brass instruments of the 19th and early 20th centuries between long-term conservation and use in historically informed performance practice” CR12I1_146330/1. The Italian Ministry of University and Research (MIUR) is thanked for financing the PRIN project prot. 2010329WPF_005 “Sustainability in cultural heritage: from diagnosis to the development of innovative systems for consolidation, cleaning and protection”. The Sardinian Regional Government is gratefully acknowledged for financial support (P.O.R. Sardegna F.S.E. Operational Program of the Regione Autonoma della Sardegna, European Social Fund 2007–2013 – Axis IV Human Resources, Objective 1.3, Line of Activity 1.3.1 “Avviso di chiamata per il finanziamento di Assegni di Ricerca”). Today part of the Klingendes Museum Bern, www.klingendes-museum-bern.ch (all links in this article last consulted 18 March 2020).

[2] See e. g. John Butt: *Playing with History. The Historical Approach to Musical Performance*, Cambridge 2002.

practice"[3] is to examine the corrosion phenomena in historical instruments currently being used, and to present an appropriate set of recommendations for their conservation and usage.

The main concern on the part of museums and conservators is corrosion in historical brass instruments due to the high humidity inside the instruments caused by playing.[4] Indeed, measurements made during this project have shown that after five minutes of playing, the relative humidity (RH) inside brass instruments exceeds 90%. It takes several days to reach ambient RH again afterwards.[5] s means that when instruments are played regularly, a thin film of water can be present inside them for quite a long time, meaning that long-term corrosion damage can occur. It is well known that in outdoor atmospheric corrosion, the 'time of wetness' (the time during which a water film is present on the metal surface) controls corrosion intensity. The working hypothesis for preventive conservation of these instruments was thus to reduce the time of wetness by drying them with a fan.

Any preventive conservation measure aiming to control the conditions inside historical brass instruments must, however, be tested and monitored for its efficiency. Conservators need a tool for obtaining the relevant information on the corrosion rate inside the instruments in a rapid, non-destructive way. As corrosion is an electrochemical process,[6] electrochemical techniques are our methods of choice. We shall here present an electrochemical in-situ sensor for corrosion rate measurement inside the tuning slides that is intended to help us monitor the effectiveness of preventive conservation. Parts of this summary paper have already been published in scientific journals.[7]

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[3] For details, see www.hkb-interpretation.ch/projekte/korrosion and the text by Adrian von Steiger in this volume, pp. 32–47.

[4] Einar Mattsson/Rolf Holm: Atmospheric Corrosion of Copper and Its Alloys, in: *Atmospheric Corrosion*, ed. by William H. Ailor, New York a. o. 1982, pp. 365–392.

[5] See the article by Martin Ledergerber, Emilie Cornet and Erwin Hildbrand in this volume (pp. 48–60).

[6] See R. Winston Revie/Herbert Henry Uhlig: *Corrosion and Corrosion Control. An Introduction to Corrosion Science and Engineering*, New York a. o. 2008, pp. 9–19.

[7] Bernhard Elsener/Federica Cocco/Marzia Fantauzzi/Silvia Palomba/Antonella Rossi: Determination of Corrosion Rate Inside Historical Brass Wind Instruments. Proof of Concept, in: *Materials and Corrosion* 67 (2016), pp. 1336–1343, https://doi.org/10.1002/maco.201608996; Bernhard Elsener/Marion Alter/Tiziana Lombardo/Martin Ledergerber/Marie Wörle/Federica Cocco/Marzia Fantauzzi/Silvia Palomba/Antonella Rossi: A Non-Destructive In-Situ Approach to Monitor Corrosion Inside Historical Brass Wind Instruments, in: *Microchemical Journal* 124 (2016), pp. 757–764, https://doi.org/10.1016/j.microc.2015.10.027; Federica Cocco/Marzia Fantauzzi/Bernhard Elsener/Antonella Rossi: Dissolution of Brass Alloys Naturally Aged in Neutral Solutions. An Electrochemical and Surface Analytical Study, in: *RSC Advances* 6 (2016), pp. 90654–90665, https://doi.org/10.1039/c6ra18200c.

Corrosion basics In general, copper and brass alloys are quite resistant to atmospheric corrosion due to the formation of protective layers of corrosion products, which greatly reduce the rate of attack.[8] If we wish to monitor the efficiency of a preventive action to reduce corrosion, electrochemical measurements are best suited to determining the corrosion state and the instantaneous corrosion rate of an alloy in a given environment.

Rather like a battery, corrosion reactions are redox reactions composed of oxidation (anode) and a reduction reaction (cathode):

Oxidation: $2\,Zn \rightarrow 2\,Zn^{2+} + 4\,e^-$ (1a)
Reduction: $O_2 + 2\,H_2O + 4\,e^- \rightarrow 4\,OH^-$ (1b)

The electrons released at the anode have to be consumed immediately by the cathodic reaction to maintain electro-neutrality, thus the anodic dissolution current and the cathodic current must be equal (Figure 1) and correspond to the corrosion current.

Of the information contained in Figure 1, only the corrosion potential E_{corr} can be measured experimentally. The corresponding corrosion current density i_{corr} flowing in the system is short-circuited, thus the current measured from external I = 0. However, i_{corr} can be indirectly determined by measuring the polarisation resistance Rp, the inverse of the current versus potential curve close to the corrosion potential ϵ_{corr} (insert in Figure 1).

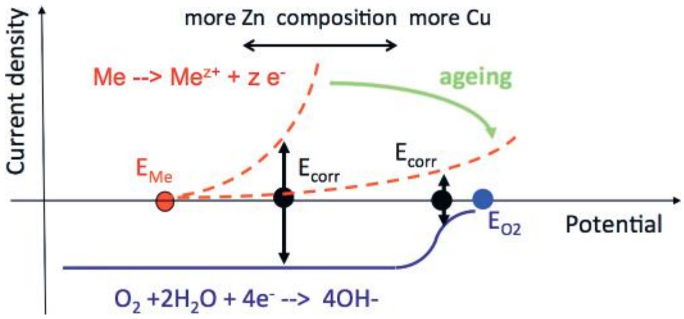

FIGURE 1 Schematic current density – potential curves for brass in neutral solutions. With time of immersion (ageing) the corrosion rate decreases and E_{corr} moves to more positive potentials. E_{corr} establishes at the point of electro-neutrality.

From the measured polarisation resistance Rp, the corrosion current density i_{corr} can be calculated with the Stern-Geary relation[9] that holds for uniform corrosion.

$i_{corr} = B/Rp$ (2)

The constant B in equation (2) depends on the specific system under test; a value of 26 mV for B has been assumed in this work.

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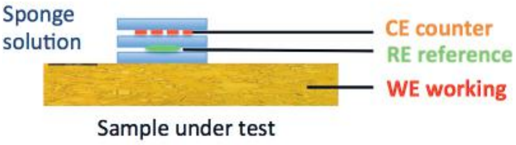
[8] Mattsson/Holm: Atmospheric Corrosion of Copper and Its Alloys.

[9] Revie/Uhlig: *Corrosion and Corrosion Control*, pp. 53–82.

Finally, the loss in thickness or the corrosion rate v_{corr} (in µm/year) can be calculated using Faraday's law. The resulting conversion factor for pure copper is 12, thus 1 µA/cm^2 corresponds to 12 µm/year. This procedure is valid for general corrosion as it occurs on brass alloys in neutral electrolytes.

The electrochemical sensor A dedicated electrochemical sensor was developed to perform the electrochemical measurements inside the historical brass instruments, open circuit potential and polarisation resistance. The sensor combined an Ag/AgCl solid-state reference electrode and a small platinum grid as counter electrode, both embedded in a thin sponge (Figure 2a). The first prototype of the sensor had a surface of about 2 cm^2 (1.5 × 1.5 cm) and was used in the proof of concept for flat samples only (Figure 2b).[10] A further development consisted of a tubular sensor mounted on a thin plastic tube to insert the sensor in the brass instruments.[11]



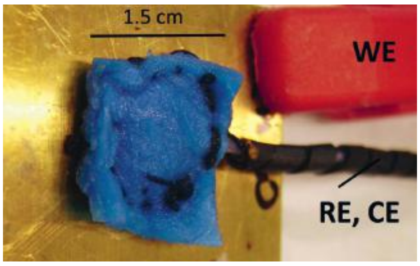


FIGURE 2 a) Schematic drawing of the sensor for electrochemical measurements on flat samples. The sponge is circa 1.5 × 1.5 cm (area 2 cm^2). Thickness circa 2 mm. b) Photograph of the sensor used on flat brass samples

This tube allowed us to slightly inflate a small balloon mounted at its end and to press the sensor gently against the inside of the tuning slide. When the measurement at one point was completed, the air pressure was released and the sensor was moved to the next measuring position without scratching the inside of the instrument part.

The flat sensor was tested on two model brass alloys (CuZn18 and CuZn37) in different surface states. In order not to alter the surface being tested, a non-aggressive phosphate buffer solution (pH 7) with a small amount of NaCl was used. The results of corrosion potential E_{corr} and polarisation resistance Rp measured with the sensor were comparable to the results obtained in the traditional electrochemical cell as is shown in Figure 3. Freshly polished samples show the lowest Rp values (10–20 kΩcm^2) corres-

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[10] Elsener/Cocco/Fantauzzi/Palomba/Rossi: Determination of Corrosion Rate.

[11] Elsener/Alter/Lombardo/Ledergerber/Wörle/Cocco/Palomba/Fantauzzi/Rossi: A Non-Destructive In-Situ Approach to Monitor Corrosion.

ponding to the highest corrosion rate of about 30 μm/year; the 'as received' Cu18Zn brass alloy showed Rp values of around 80 kΩcm², corresponding to a corrosion rate of 4 μm/year. On an old mouthpiece, the corrosion rate was found to be 0.4 μm/year. Note that these are the initial corrosion rates. After longer contact with the solution, the values decrease by about a factor of 10.[12] Overall, despite some scatter in the individual results, a linear correlation with a slope of about 0.95 was found between the corrosion rate measured with the sensor and in the electrochemical cell, indicating that the sensor was working properly and reproduced the exposure situation of a traditional electrochemical cell. The accuracy is good and can be estimated to be about ±20% over two orders of magnitude.[13]

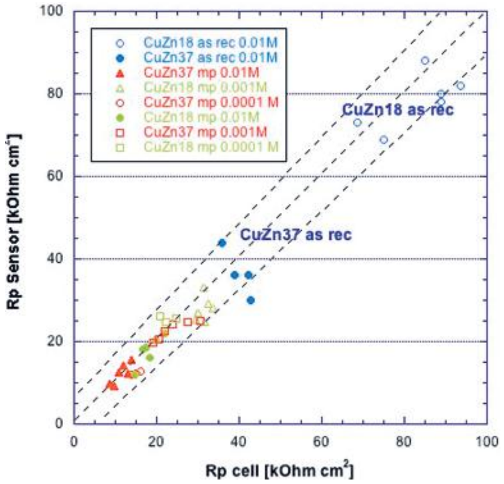

FIGURE 3 **Correlation of the specific polarisation resistance, Rp, measured by the sensor, versus the specific Rp measured using the electrochemical cell. Phosphate buffer solution pH 7 with chloride concentrations from 10^{-4} to 10^{-2} M NaCl. Materials CuZn37 and CuZn18, surface condition 'as received' and mechanically polished[14]**

Results from historical instruments In order to assess the preventive conservation (using a fan to dry the instruments after playing), measurements with the sensor were performed before, halfway through and at the end of the testing period of 14 months, in all the 34 tuning slides of the 16 instruments. In total, this came to 102 individual measurement positions. We present here the results of four selected horns (Table 1) from two different manufacturers, Raoux-Millereau, Paris (F) and Couesnon Monopole, Paris (F).[15]

Description of the brass instruments (horns) These instruments were manufactured between 1900 and 1932. In a previous project it was found that manufacturers used

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[12] Cocco/Fantauzzi/Elsener/Rossi: Dissolution of Brass Alloys, p. 90662.

[13] Elsener/Cocco/Fantauzzi/Palomba/Rossi: Determination of Corrosion Rate, p. 1341.

[14] Ibid, p. 1342.

[15] See also Elsener / Alter / Lombardo / Ledergerber / Wörle / Cocco / Palomba / Fantauzzi / Rossi: A Non-Destructive In-Situ Approach to Monitor Corrosion, p. 758.

different alloys for different parts of the instruments, for reasons that might be related to the different quality of the sound and also according to alloy availability.[16] So the overall variation of the composition (Table 1) is quite large. All these instruments were bought between 2011 and 2013 and have been stored at the Burri collection since then. With the exception of 5024, they were cleaned at a workshop shortly after purchase by immersing the instruments in a citric acid bath (Cuproten) accompanied by fine brushing with a nylon brush, followed by neutralisation in a sodium carbonate (Na_2CO_3) solution. The inside of instrument 5024 has not been cleaned in the past 30–50 years. Only exterior stains were removed, and the pistons were cleaned mechanically.

TABLE 1 The four instruments (horns) used in the investigation. The range of compositions (wt%) determined by XRF is provided.[17]

Instrument	Manufacturer	Year	Condition	XRF composition (wt%)
HKB 5004	Couesnon Monopole, Paris	1922	Cleaned in 2011/2012	Cu: 60–76 Zn: 22–37
HKB 5009	Raoux-Millereau, Paris	1900	Cleaned in Dec 2012	Cu: 64–69 Zn: 30–35
HKB 5024	Couesnon Monopole, Paris	1932	no cleaning	Cu: 61–74 Zn: 22–38
HKB 5025	Couesnon Monopole, Paris	1910	Cleaned in 2013/2014	Cu: 62–73 Zn: 26–38

Condition assessment with the sensor before playing Measurements of corrosion potential were made in-situ with the tubular sensor (Figure 4), and readings were taken every 1 cm from the entrance on both sides of the tuning slide. After the sensor was pressed against the tube with the small balloon and electrolytic contact had been established, readings of the corrosion potential were taken after 0.5, 1 and 2 minutes at each point. Examples of the results obtained on three different tuning slides are shown in Figure 5. No systematic variations of the corrosion potential were found except the clearly lower ϵ_{corr} values at points of soldering.

The points for polarisation resistance (Rp) measurements were selected on the basis of the corrosion potential recorded previously. Rp measurements were performed in areas with quite homogeneous corrosion potential over several centimetres and were begun 2–5 minutes after contact with the sensor.

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[16] See Marianne Senn/Hans J. Leber/Martin Tuchschmid/Naila Rizvic: Blechblasinstrumentenbau in Frankreich im 19. Jahrhundert. Analysen von Legierung und Struktur des Messings zugunsten eines historisch informierten Instrumentenbaus, in: *Romantic Brass. Französische Hornpraxis und historisch informierter Blechblasinstrumentenbau. Symposium* 2, ed. by Daniel Allenbach, Adrian von Steiger and Martin Skamletz, Schliengen 2016, pp. 398–419.

[17] See also Elsener / Alter / Lombardo / Ledergerber / Wörle / Cocco / Palomba / Fantauzzi / Rossi: A Non-Destructive In-Situ Approach to Monitor Corrosion, p. 760.

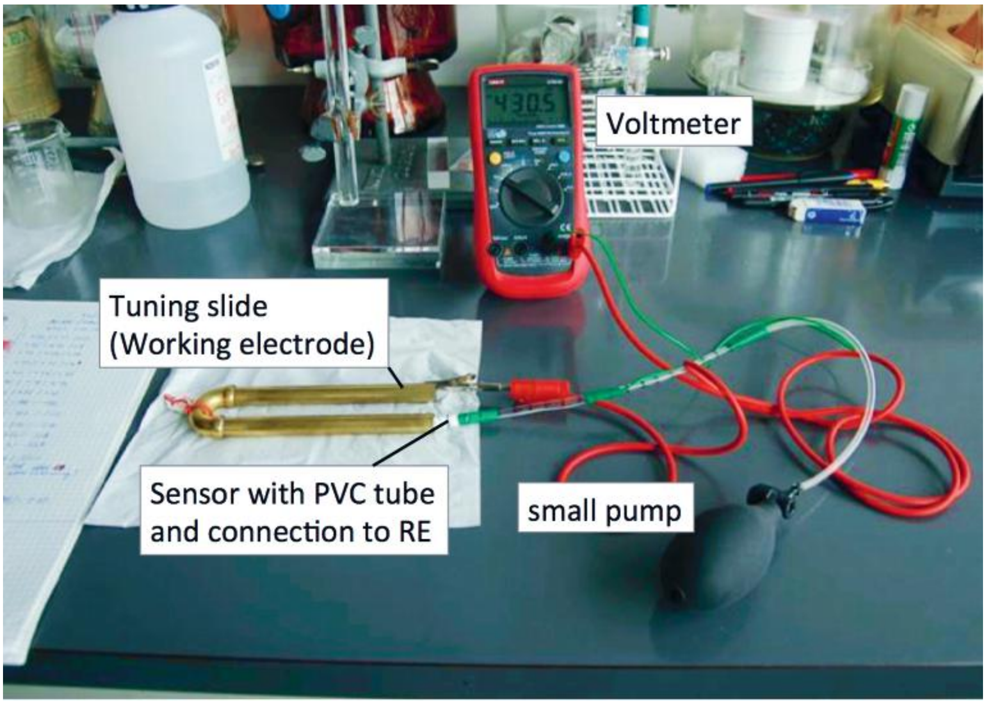

Figure 4 Experimental arrangement for the in-situ measurements of corrosion potential and polarisation resistance, using the tubular sensor

Figure 5 Results of corrosion potential measurements performed with the sensor inside the tuning slides. The numbers here indicate the potential measured at a given position after 30 seconds, 1 and 2 minutes. Measurements were taken at a distance of 1 cm in the left and right arm of the tuning slide. Potential of the sensor vs Ag/AgCl sat = -170 ±10 mV.
a) HKB 5004.2 – b) HKB 5009.2 – c) HKB 5025.1

The results of the Rp measurements of the nine tuning slides from the four different instruments show great variations, both within individual tuning slides, and even more among different tuning slides.[18] Overall, no influence by the composition or cleaning procedure can be observed. The polarisation resistance thus allows us to determine the instantaneous corrosion rate at the point of measurement. However, no precise indication of the corrosion state (the reason why Rp is higher or lower) is possible.

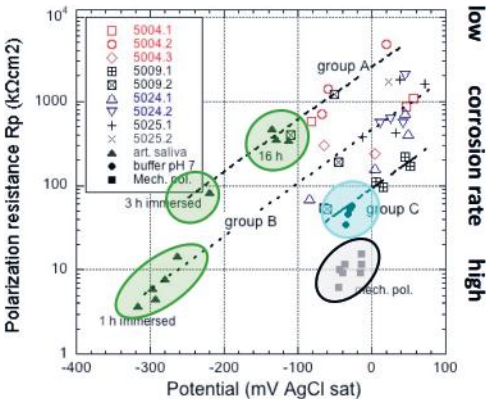

FIGURE 6 **Diagnostic plot of the specific polarisation resistance Rp versus corrosion potential ϵ_{corr} of nine different tuning slides (25 measurement points) of the four horns measured before playing the instruments. Included in the plot are data obtained on the Cu37Zn model alloy immersed for 1, 3 and 16 hours in artificial saliva (◆),[19] data from all the model alloys in a mechanically polished surface state (■), and data from all the model alloys immersed for one hour in the buffer solution pH 7 without chlorides (●).[20]**

Diagnostic plot log Rp versus ϵ_{corr} [illegible]ll the individual pairs ϵ_{corr}/Rp measured at different positions in the tuning slides of the four horns (Table 1) are plotted in a log Rp versus ϵ_{corr} diagram (Figure 6); combining the information from the corrosion potential and the corrosion rate allows us to rationalise the data. Indeed, three different groups of data (indicated by a diagonal line) can be found: *group* A with high to very high Rp values over the whole potential range are found for tuning slide HKB 5004.2, but also single points of HKB 5009.2 and HKB 5004.1 fall into this group. This group of data reflects situations with very high Rp values/very low corrosion rates inside the tuning slides. A much larger group (indicated as *group* B) shows Rp values about a factor of 10 lower than group A (with a corrosion rate a factor of 10 higher). The tuning slides HKB 5024.1 and 5024.2 and HKB 5025.1 belong to this group, as do two points of HKB 5004.1 and one of HKB 5004.3. A third group (*group* C) can be found in the range of 50–200 kΩcm^2, thus at quite low values of Rp. The tuning slides of HKB 5009.1 belong to this group.

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[18] See ibid, p. 761.

[19] Federica Cocco: *Comportamento elettrochimico e analisi* XPS *e* XAES *di leghe di ottone per la produzione di strumenti musicali del* XIX *secolo* [*Electrochemical and* XPS/XAES *Surface Analytical Investigation of Brass Alloys of Musical Instruments of the 19th Century*], Master Thesis, University of Cagliari, 2013.

[20] Silvia Palomba: *Indagine elettroanalitica del rame, dello zinco e di leghe di Ottone* [*Electrochemical Investigation of Copper, Zinc and Brass Alloys*], Master Thesis, University of Cagliari, 2015.

The three groups A, B and C can be described each with a diagonal line with slope circa 160 mV per decade Rp. For a given (constant) environment characterised by pH value and oxygen content (in the sensor sponge), the surface state and the alloy composition give rise to systematic and correlated variations of both ϵ_{corr} and Rp, showing that Rp increases (corrosion rate i_{corr} decreases) with increasing potential. However, the data from inside the tuning slides lack any information on the surface composition.

Correlation to the surface state he comparison with data from well-controlled laboratory experiments where the surface state is known substantiates the E_{corr}/Rp results obtained inside the tuning slides. Three sets of data have been included in Figure 6.

The *first set of data* refers to the mechanically polished CuZn37 model alloy exposed to artificial saliva.[21] One can note that after 1 hour of immersion, the data points fall on the line of group B, after 3 and 16 hours respectively of immersion the E_{corr}/Rp data points instead fall on the line of group A. XPS surface analysis performed on the samples after 16 hours of immersion in artificial saliva documented the formation of a surface film composed of CuSCN and $Zn_3(PO4)_2$.[22] This surface film has protective properties and increases Rp, thus strongly lowers the corrosion rate.

The *second data set* refers to mechanically polished brass alloys exposed for 1 hour in the buffer solution pH 7 (no chloride ions).[23] One can note that these E_{corr}/Rp data fall on the line of group C; we may assume the formation of a non-protective surface film.

The *third set of data* refers to measurements with the sensor on mechanically polished brass alloys. The Rp values are very low (circa 10 $k\Omega cm^2$), indicating a very high corrosion rate of these freshly polished samples where practically the bare alloy is exposed to the solution.

Combining data from well-controlled laboratory experiments where XPS surface analysis was performed[24] with the E_{corr}/Rp results obtained inside the tuning slides (Figure 6) allows for the following tentative interpretation: The surface condition of samples lying on *line* A can be assumed to show a protective surface film similar (or much more protective) to that formed upon immersion in artificial saliva for 16 hours. The Rp values are generally high to very high and the corrosion rates correspondingly low to very low. The surface condition of samples lying on *line* B can be assumed to present a more or less thick oxy-hydroxide film, formed after prolonged air exposure or in the initial

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[21] Cocco: *Comportamento elettrochimico*.

[22] See Cocco/Fantauzzi/Elsener/Rossi: Dissolution of Brass Alloys; Cocco: *Comportamento elettrochimico*.

[23] Palomba: *Indagine elettroanalitica*.

[24] Cocco/Fantauzzi/Elsener/Rossi: Dissolution of Brass Alloys; Cocco: *Comportamento elettrochimico*; Palomba: *Indagine elettroanalitica*.

phases of immersion in artificial saliva.[25] Rp values compared to *line* A are clearly lower, thus the corrosion rates are higher. Samples lying on *line* C could be points where a nearly bare surface is present. A mechanistic interpretation based on the effect of surface composition on the rate of the oxygen reduction reaction has been proposed.[26]

Efficiency of preventive conservation The tuning slides of 16 brass instruments of different age, conservation conditions and state of cleaning[27] were characterised with corrosion potential and polarisation resistance (Rp) measurements before the playing period began, and after 7 and 14 months (for the procedure, see above). During the playing period, the instruments were played daily for at least 5 minutes (with some exceptions) in order to increase the humidity inside the instrument. One group of musicians used only conventional measures (emptying valves to drain condensed water), while the second group used a fan to ventilate the instrument with air in order to dry it out (preventive conservation).

The results of the measurements constitute a huge amount of data and are presented here as a cumulative probability plot, a statistical analysis for any distributed values. Note that a normal Gaussian distribution in a histogram results in a straight line in the cumulative probability plot. The following groups have been created:

- instruments before playing started
- instruments without preventive conservation, after 7 months and after 14 months
- instruments with preventive conservation, after 7 months and after 14 months

Instruments without preventive conservatio The statistical analysis in the cumulative probability plot of the instruments (tuning slides) without preventative conservation measures is presented in Figure 7a. It can be noted that the average of polarisation resistance after 14 months of playing is slightly lower (shifted to the left) compared to the average Rp before. This indicates that the *average* corrosion rate measured (which value can be read at the 50% level) was about a factor two higher after a 14-month playing period than could be expected by the high humidity inside the instruments. The lines at the three measuring times (0, 7, 14 months) are nearly parallel, indicating that the standard deviation remained the same.

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[25] Cocco: *Comportamento elettrochimico*.

[26] Elsener/Alter/Lombardo/Ledergerber/Wörle/Cocco/Palomba/Fantauzzi/Rossi: A Non-Destructive In-Situ Approach to Monitor Corrosion.

[27] See the article by Adrian von Steiger in this volume, pp. 32–47.

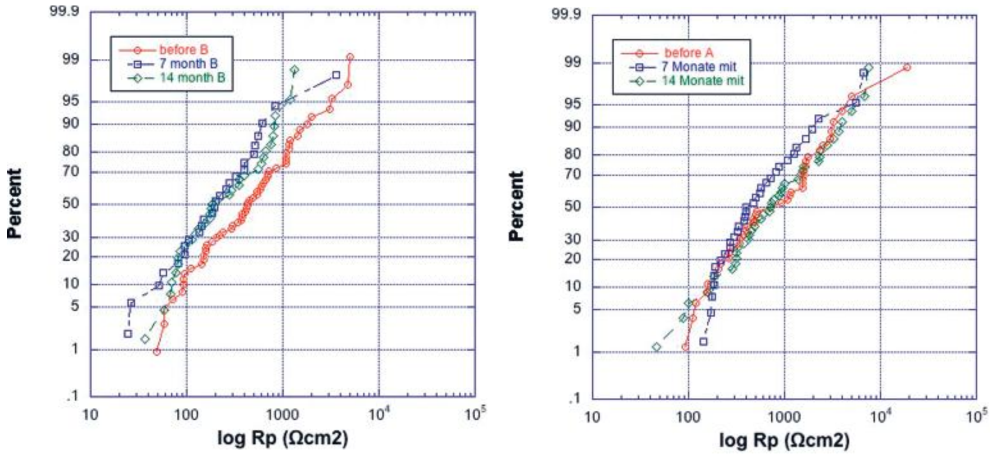

FIGURE 7 **Statistical analysis (cumulative probability plot) of the polarisation resistance Rp measured in the different tuning slides of the brass instruments before, after 7 months and after 14 months of playing. Left: without preventive conservation; right: with preventive conservation (drying with a fan)**

Instruments with preventive conservatio When we compare the Rp measurements in the tuning slides of instruments with prevention conservation before, after 7 months and after 14 months (Figure 7b), we see that the average of the Rp values remained approximately constant (the green curve, 14 months, is identical to the red curve, 0 months). This indicates that the average corrosion rate measured in the tuning slides did not change with time; drying with the fan as a preventive conservation measure was thus effective in preventing the onset of more rapid corrosion such as was observed in the tuning slides without preventive conservation (Figure 7a).

To evaluate a change in the polarisation resistance value at the *individual* measuring points in the tuning slides of the different instruments, a simpler analysis procedure was used (“traffic light”), similar to the evaluation of the endoscope measurements:[28]

- “green” – Rp values are constant (within ±20%)
- “orange” – no clear trend in the Rp values over time
- “red” – the Rp values decrease over playing time

Based on this analysis, instruments with preventive conservation showed 70% of the values “green” and 20% “orange”. Instruments without prevention instead showed 70% of the values “red” and 20% “orange”. Thus, also based on an analysis of the individual tuning slides, it can be clearly shown that the preventive measure of drying the instrument with a fan after playing is effective.

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[28] See the article on endoscopy by Martin Ledergerber in this volume, pp. ###–###.

Conclusions The non-destructive electrochemical sensor for in-situ measurements inside the tuning slides of historical brass instruments worked well; the same results were obtained on model alloys as with conventional electrochemical measurements.

The results of the measurements before the playing period showed a great variation in the rate and state of corrosion inside the tuning slides and among the instruments; for this reason, statistical analysis (cumulative probability plots) had to be used.

After 14 months of playing, the two groups of instruments 'without prevention' and 'with prevention' could clearly be distinguished: inside the tuning slides of instruments where the drying procedure had been applied, the corrosion rate was the same as before, or increased only slightly. On the contrary, inside the tuning slides of instruments without this preventative procedure, the corrosion rate increased over time, on average by a factor of two.

As a scientific result, the diagnostic plot 'log Rp versus E_{corr}' allows us to group the individual measurement points and to allocate them to very low, low and medium-high corrosion rates. The reason why individual measurement points belong to which group can be found in the thickness and composition of the surface film inside the brass instruments.

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