‘Listening to corrosion’

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ABSTRACT

Using condition monitoring techniques to achieve predictive maintenance is a prominent topic for military systems. Some of the main challenges related to this topic will be introduced, and after that a specific application will be used to demonstrate the successful development of a corrosion monitoring technique. One of the effective ways to cope with corrosion as a failure mechanism is to use dedicated sensors. Preferably, these sensors do not interfere with the prevalent corrosion process, i.e. they ‘listen to corrosion’ as it occurs spontaneously. A potentially interesting monitoring technique is based on electrochemical noise (EN), which is the spontaneous charge transfer generated by the corrosion process. A unique property of this technique is the possibility to identify corrosion processes based on their EN signature. This work describes the analysis of EN signals, based on which corrosion identification can be performed. Metastable pitting of AISI304 stainless steel serves as an example of the analysis procedure. The effectiveness of the procedure is then demonstrated by means of the identification of microbiologically influenced corrosion (MIC), which is generally regarded as one of the most difficult to predict corrosion mechanisms.

1.0 INTRODUCTION AND BACKGROUND

The operational availability of military vehicles is typically ensured by executing a considerable amount of preventive maintenance. Traditionally, the intervals at which this preventive maintenance is performed are predetermined during the design of the vehicle, and are not adapted to the specific use or operational context of a vehicle. This often results in severe over-maintenance, leading to high costs, but in some cases also still yields unexpected failures. In recent years, a transition to smarter maintenance concepts is seen, which are typically denoted as predictive maintenance.

The basic idea of this type of maintenance is to predict when failures are expected, taking into account the specific operational profile of an individual system, and perform just-in-time maintenance. This means that parts are not prematurely replaced at fixed intervals, but only when needed. It also means that unexpected failures due to severe usage of certain vehicles can be prevented, as these failures can be predicted. This leads to a maintenance concept that is both efficient (in terms of costs) and effective (in terms of down-time reduction).

To enable the practical application of predictive maintenance, proper assessment of the present condition (i.e. diagnosis) as well as accurate prediction of the remaining useful life of system or component (i.e. prognosis) are required. A lot of development has taken place in this field lately, partly facilitated by the emergence of data analytics, internet of things, availability of many different sensors and the ease of storage and processing of large amounts of data. However, diagnosing a system, and especially predicting its time to failure are not a trivial task yet. Some of the (technical) challenges will be discussed next in a general sense, and then a (diagnostic) solution for a specific (corrosion) problem will be elaborated.

Challenges

Although it is clear that monitoring is an essential part of preventive maintenance, the first challenge in this field is to decide what parameter should be monitored to achieve the required diagnosis or prognosis. Basically there are three options for monitoring [1]:

(i) Condition monitoring – a parameter that is representative for the component / system condition, e.g. a crack length, wear depth or vibration level, is *measured*. As soon as a predefined threshold level is exceeded, maintenance must be performed;

(ii) Load monitoring – if it is not possible to directly monitor the condition of the system (for technical or economic reasons), measuring the load on a component, e.g. the temperature or strain, can assist in *calculating* the degradation of the system. However, the use of a model to relate the load to the life time (e.g. fatigue or wear model) is required in this case;

(iii) Usage monitoring – if both previous approaches are not feasible, monitoring the usage of the system, i.e. operating hours, number of starts, power settings, can be registered. This info can also be used to *calculate* the degradation of the system, but again a model is required and the accuracy of the life prediction is lower than for load and condition monitoring;

Selecting the type of monitoring depends on the application as well as on the availability of (commercial) monitoring techniques. Typically condition monitoring provides the highest accuracy and the least technical knowledge on the monitored system (as it is the most direct approach). Load and usage monitoring typically require more simple and cheaper sensors, but come with more uncertainty and require knowledge on the system and its failure mechanisms to select or develop a model.

The second challenge is the analysis and interpretation of the raw monitoring data. Many OEMs nowadays install sensors on the assets that they deliver, providing large amounts of measurement data. However, to translate this raw data into useful information is not trivial. Typically it requires detailed knowledge on both the system and the associated failure mechanisms to retrieve a diagnosis or even prognosis from the data. Recent advances in data analytics have shown that machine learning and other artificial intelligence (AI) methods can partly replace that knowledge, as training these methods with large datasets can also results in useful models. However, for critical systems, as is typical for military vehicles, only a limited number of failures are present in the datasets, which significantly complicates the application of AI methods.

The third challenge in this field is the step from diagnosis to prognosis. As many commercial condition monitoring techniques already exist for years or even decades, they are extensively used to diagnose systems. However, the maintenance activities are then in most cases triggered by a condition measurement exceeding a predefined threshold value. This means that immediate action is required, which from a planning perspective is not the most attractive option. Therefore, adding a prognostic capability to the diagnosis would enable a much more efficient maintenance policy, as future tasks can be properly scheduled and planned at a convenient moment in time. Prognostic methods are typically based on either data-driven methods or physical model based approaches [2,3], but all have the drawback that proper validation of the models can only be done when the system actually fails. As mentioned before, failures are scarce for critical systems, so unless (accelerated) testing in a laboratory environment is possible, the validation is rather problematic. This could, however, be improved when these methods are combined with condition monitoring methods, that precisely assess the actual condition of the system continuously or periodically. Therefore, a promising way to make the step from diagnosis to prognosis would be to integrate condition monitoring techniques with prognostic methods.

Having addressed three of the main challenges in the field of predictive maintenance, the remainder of this paper will focus on a specific failure mechanism, i.e. corrosion. This case will illustrate the challenges discussed, as the choice for a proper quantity to measure, as well as the analysis and interpretation of the measurement data also appear here. Note that the challenges discussed here are not the only challenges, as achieving an acceptable Probability of Detection (PoD) and getting monitoring systems certified for practical use are two other notable challenges in the field.

2.0 CORROSION MONITORING

In many cases, corrosion is a predominant failure mechanism, where a reduced wall thickness leads to leakage, crack initiation of even fracture of a structural part. Corrosion of critical (sub-)systems often occurs unexpectedly and typically remains undetected until failure occurs. Therefore, mitigation of corrosion can be complicated and expensive. In order to reduce corrosion cost and to increase operational availability and system safety, a proper understanding of this phenomenon is required, as well as a robust monitoring solution to provide the required information.

Corrosion is in most cases an electrochemical process, involving charge transfer between anodic and cathodic sites. This charge transfer results in fluctuations in current and potential, determined as electrochemical noise (EN). Many different corrosion mechanisms exist and each one can be associated with specific characteristics in current and potential.

The present research focuses on a passive monitoring technique, called EN measurements, that detects and characterizes essentially those current and potential fluctuations associated with corrosion. This enables the maintainer not only to detect corrosion occurrence, but also its intensity and, most importantly, it facilitates distinguishing between different types of corrosion.

The detection and analysis of EN was first described by Hagyard and Williams in 1961 [4]. After a few years, in 1968, the next report on EN was published by Iverson [5]. Over the years, many more reports on the application of the technique and interpretation of EN signals appeared [6]. It is therefore valid to state that the principle of the detection and analysis of these kinds of signals in itself is not quite innovative. Nowadays however, with the rapid emergence of new materials and the gradual increase of computer power, EN can be measured and treated in intriguing novel ways that were not possible before. The detection of EN in itself has remained rather straightforward, which can also be regarded as an important asset of the technology, in terms of its simplicity. One of the challenges in this respect would be the reliability of the measured signal. Even under controlled laboratory conditions, it is e.g. essential to verify the level of the instrumental noise [7], which will inevitably be present, and to diminish the influence of external noise. In the case of external noise, the fact that corrosion processes typically exist in the frequency range between 10^{-3} and 1 Hz [8] can be regarded as advantageous for the detection of a clear corrosion signal. Most electromagnetic signals are many orders of magnitude higher in frequency.

The real asset of EN compared to other corrosion detection techniques is to acquire detailed information about corrosion intensity, its location in time and corrosion type. This information is captured in the EN signal that is spontaneously generated by the corrosion process, which is simple and straightforward to measure. Usually, the ability to characterize localized corrosion processes can only be provided by complicated techniques that are applicable only under limited (laboratory) conditions. Examples are Scanning Kelvin Probe (SKP), Scanning Reference Electrode Technique (SRET), Scanning Vibrating reference Electrode Technique (SVET), Local Electrochemical Impedance Spectroscopy (LEIS) and Scanning Electrochemical Microscopy (SECM) [9–12].

This work provides an explanation of the principle to extract corrosion ‘fingerprints’ from EN, based on the pitting characteristics of AISI304 stainless steel. Each localized corrosion event, such as pitting, generates a so-called transient in the EN signal [13]. Using a transient detection and analysis procedure, such a transient can be used to identify the type of corrosion, for example in the case of pitting corrosion of stainless steel [14]. However, the pitting behaviour of stainless steel is generally well documented. On the contrary, a localized corrosion problem such as microbiologically influenced corrosion (MIC) of carbon steel can potentially occur quite unexpectedly. Therefore, to further illustrate the potential of the proposed technique, this work also shows the application of EN transient analysis on the detection and characterization of MIC of carbon steel [15].

3.0 DETECTION PRINCIPLE

The principle of an EN measurement is shown in Figure 3-1. Two working electrodes generate a current, called electrochemical current noise (ECN) and a potential with respect to a separate reference electrode, called electrochemical potential noise (EPN). The ECN is measured by a zero resistance ammeter (ZRA) and the EPN is detected by a potentiometer (E).

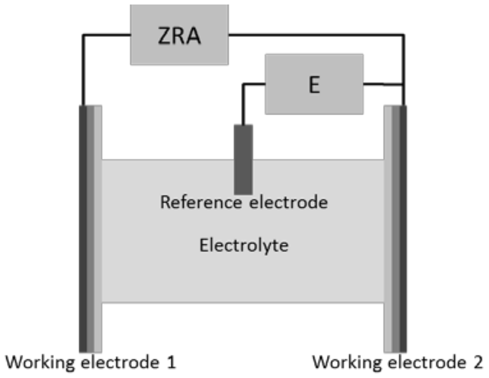

Figure 3-1: Practical layout of an EN measurement cell (from [16]).

In practice however, an EN measurement can be performed with more practical auxiliary electrodes, e.g. embedded under a protective coating, as is illustrated in Figure 3-2.

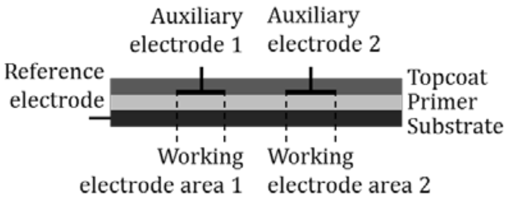

Figure 3-2: Practical EN setup using embedded electrodes (based on [17–20]).

In this setup, the auxiliary electrodes are inert and the actual working electrode area is the substrate area underneath the auxiliary electrodes. By connecting the auxiliary electrodes 1 and 2 with a ZRA, an ECN is obtained. The substrate is electrically connected to a potentiometer in order to obtain an EPN signal. The resulting data after the EN measurement are the ECN and EPN signals. Processing this data in the correct way can result in the identification of corrosion processes. This, together with the passive nature of the measurement, is the asset of EN compared to other corrosion detection techniques.

The basis of the ability to identify corrosion processes based on their EN signature is the decomposition of those signals in their intrinsic oscillations. This operation is performed by the empirical mode decomposition (EMD). After EMD, the intrinsic oscillations are in turn transformed into instantaneous frequencies using the Hilbert-Huang transform (HHT). The result of the HHT is visualized in a so-called Hilbert spectrum. In the present work, the principle of EMD and HHT will be explained based on the pitting process of AISI304 stainless steel. Figure 3-3 shows an example ECN (top) and EPN signal (bottom) of a pitting process of AISI304 stainless steel exposed in an aqueous HCl solution at pH4.

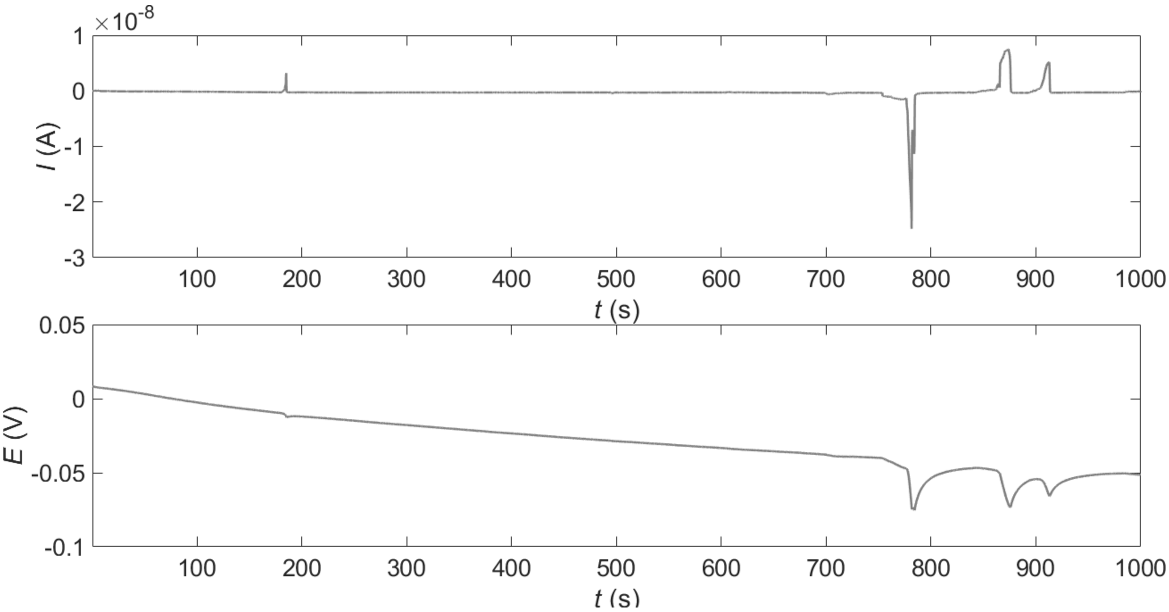

Figure 3-3: Example of an ECN (top) and EPN signal (bottom) of AISI304 during exposure to HCl at pH4 for a duration of 1000s.

In Figure 3-3 the ECN and EPN signals contain transients that are typical for the metastable pitting process of AISI304 in this environment. Those transients contain instantaneous frequency information that should be extracted and analyzed in order to define the type of corrosion. This operation is performed using the HHT. As mentioned before, the HHT is based on the EMD, which assumes that any signal consists of multiple intrinsic modes of oscillation, each one representing part of the underlying physico-chemical process. The principle of EMD and HHT was first introduced by Huang et al. [21]. Super positioning the intrinsic oscillation modes would result in the original signal again. This paper will focus on the analysis of the ECN as an indicator of corrosion type. Figure 3-4 shows the decomposition of the ECN signal of Figure 3-3 into their intrinsic oscillations, from high frequency (d1), which stands for ‘detail component 1’, to low frequency (d10) and finally the DC drift (r10), which stands for ‘residual component’. The residual component is not considered for further analysis.

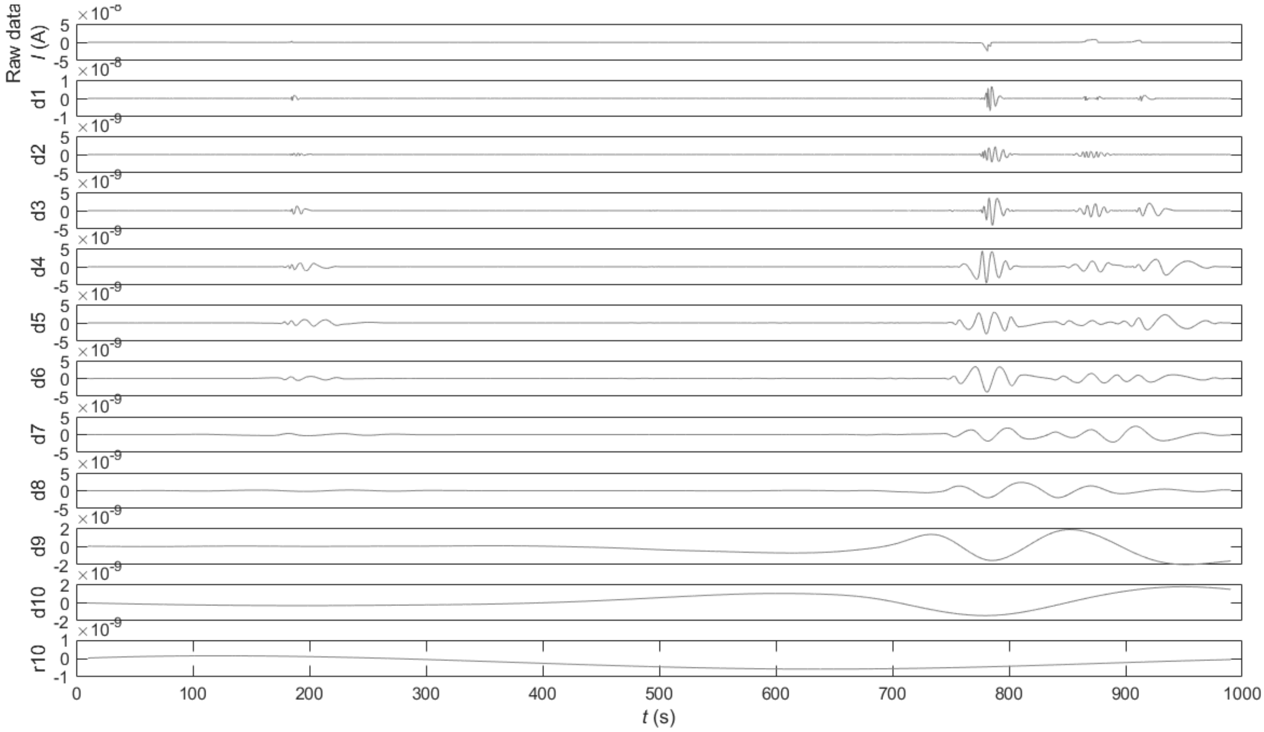

Figure 3-4: Decomposition of the ECN signal of Figure 3-3 into their intrinsic oscillations, from high frequency (d1) to low frequency (d10) and finally the DC drift (r10).

The decomposition of the original ECN signal into its intrinsic oscillations is most apparent when considering the single large transient at approximately t=780s in Figure 3-3. In Figure 3-4, this transient clearly results in high frequency oscillations in d1, with the oscillations decreasing in frequency towards d10. Each transient contributes to this decomposition, however each contribution is relative to the original energy present in each transient. In other words: the larger the transient in the original signal, the more it will dominate the decomposition into intrinsic oscillations, and hence the Hilbert spectrum that results from this decomposition. Producing the Hilbert spectrum is the next step: after EMD, the HHT extracts instantaneous frequencies from each intrinsic oscillation. The result is a decomposition of the original EN signal into its instantaneous frequencies, at each moment in time. Instead of a constant frequency and amplitude, such as obtained by classical Fourier transform, here the result is an instantaneous frequency with variable amplitude over time [21]. This makes the HHT suitable for the analysis of nonstationary data. The instantaneous frequencies and their variable amplitudes over time are presented in a Hilbert spectrum. Figure 3-5 shows the Hilbert spectrum of the ECN signal of Figure 3-3, where the original signal is depicted at the back of the figure, the instantaneous frequencies are depicted along the y-axis and the time along the x-axis.

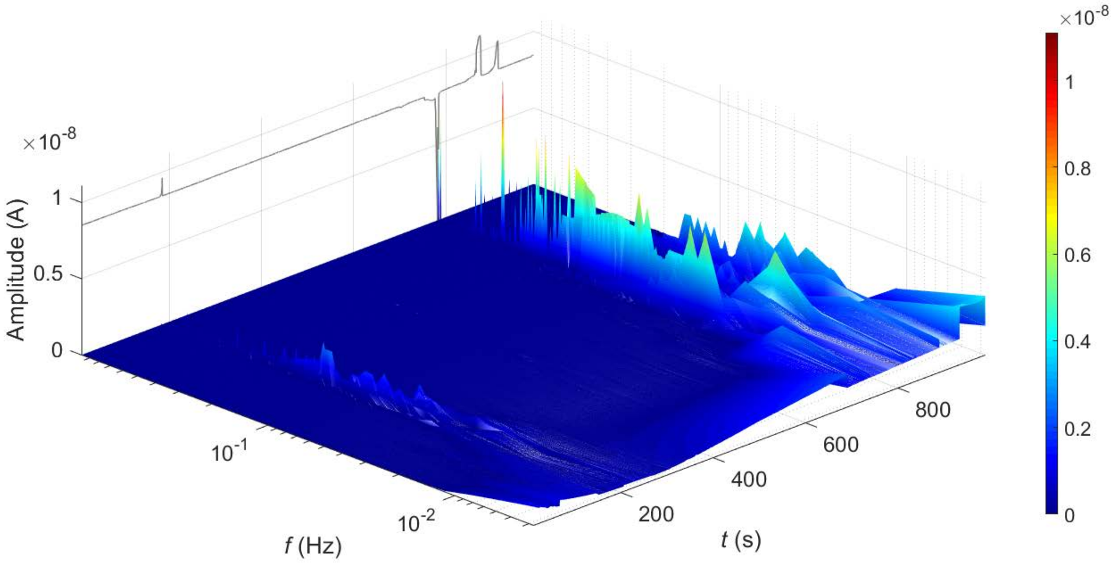

Figure 3-5: Hilbert spectrum of the ECN signal of Figure 3-3, showing the variable amplitude of the instantaneous frequencies over time. The original signal is depicted at the back of the figure.

The Hilbert spectrum shows the distribution of the energy of the EN signal in the time-frequency domain. For characterization of corrosion processes it is interesting to verify which instantaneous frequencies are dominating the spectrum at which instant of time. In the Hilbert spectrum of Figure 3-5 the single large transient occurring at approximately t=780s is associated with the largest amplitudes of instantaneous frequencies. Although there is a single large peak at approximately $5*10^{-1}$ Hz, most of the energy is located around 10^{-1} Hz. This is also the case for the first transient occurring at approximately t=180s, however, although its instantaneous frequency distribution is comparable to the large transient at t=780s, its contribution to the Hilbert spectrum is much smaller. This means that the pits that are related to these two transients have shown comparable corrosion kinetics, i.e. both are comparable from a mechanistic point of view, however the pit growing and repassivating around t=180s would have generated a smaller charge transfer and hence would have been smaller.

After the production of the Hilbert spectrum, the transients, generated by the pitting processes, should be analyzed separately for a proper understanding of the corrosion process. This results in specific regions of interest that provide insight about the prevailing corrosion mechanism. Each region provides the full decomposition into instantaneous frequencies of a transient for the duration of that specific transient. The transient locations are detected automatically, thus generating a set of regions of interest for each Hilbert spectrum. Figure 3-6 shows the result of the automatic transient detection procedure for the EN signals of Figure 3-3. The transient locations are indicated in blue.

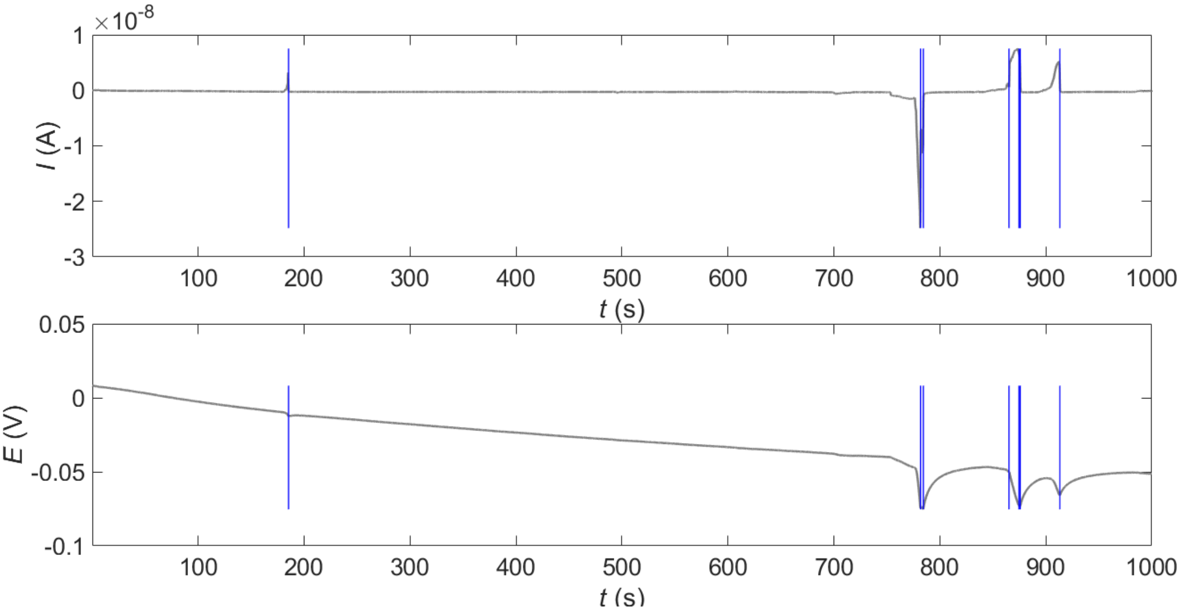

Figure 3-6: Transient locations of the EN signals of Figure 3-3, indicated in blue.

In case of continuous corrosion monitoring over a long span of time, those regions of interest (or multiple Hilbert spectra containing regions of interest) can indicate a change in corrosion characteristics over time (e.g. from general to a specific form of localized attack). In addition, Hilbert spectra allow a more profound investigation of individual transients by zooming in to their location in the spectrum and analyzing their instantaneous frequency decomposition.

4.0 DISCUSSION ON THE DETECTION OF MIC

MIC can be interpreted as the alteration or acceleration of corrosion processes resulting from the presence and activity of microorganisms [22]. During this process, it generates a biofilm at the substrate [23,24]. One of the main issues with MIC is that it may lead to unexpected failure of systems. It occurs in occasions where otherwise no corrosion would be expected. Often, MIC induces localized corrosion, e.g. pitting [22,25]. In this sense, MIC should therefore perhaps not be considered as a separate corrosion process, but more as a distinct way of enabling specific (localized) corrosion processes, involving microorganisms.

Because MIC often generates pitting corrosion, the detection of the damage induced by this process is not straightforward. Since MIC often occurs at locations that are hard to reach and corrosion pits typically grow inwards while retaining only a small pit opening, pits often remain undetected. Once pits become visually detectable, the corrosion damage is already too severe from a functional point of view. This may result in unplanned maintenance and loss of operational availability. An EN sensor however should in principle be able to detect even the smallest charge transfer induced by localized corrosion. This would result in the detection of MIC already at an early stage.

MIC was induced by the SRB (sulphate reducing bacteria) Desulfovibrio Indonesiensis, contained in a tailor-made electrochemical cell filled with nutrients. This electrochemical cell is schematically depicted in Figure 4-1.

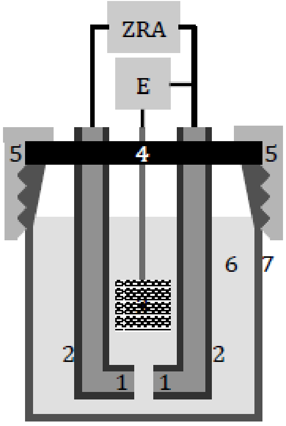

Figure 4-1: Electrochemical cell for the MIC experiments (from [15]).

This figure shows the working electrodes (1), coated with a suitable non-reactive coating (2) and electrically connected to a ZRA and potentiometer (E). Furthermore, it shows the platinum mesh serving as reference electrode (3), the rubber and plastic seal to keep the cell interior sterile (4, 5) and finally the electrolyte containing the nutrients (6) and the glass container (7). For more elaborate experimental details and results, please refer to the authors’ scientific paper on this subject [15]. This work provides an overview of the most important findings regarding the detection and identification of MIC using EN.

After exposure to the SRBs for a duration of 23 days, the EN signals indicated strong pitting corrosion activity. This would otherwise be hard to detect, since the size of the pits was still only in the µm range. Moreover, those pits were located underneath patches of biofilm with diameters of several hundreds of µm. Figure 4-2 shows a close-up image of the two working electrodes after exposure.

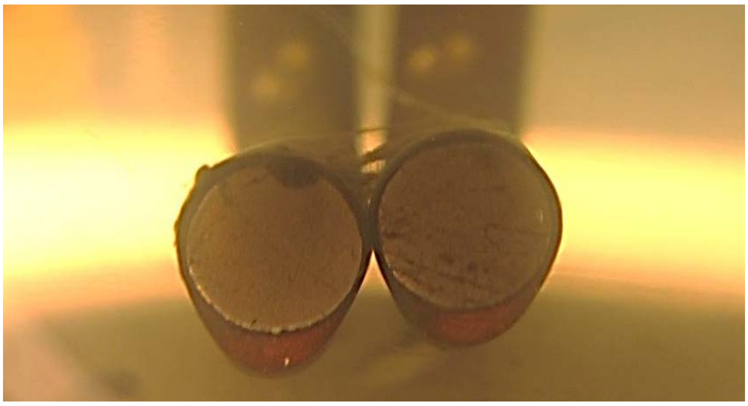

Figure 4-2: Working electrodes after 23 days of exposure.

Figure 4-2 shows smooth working electrode areas, covered by populations of SRBs. At the surface, corrosion attack was visible. Figure 4-3 shows one of the two working electrodes where multiple locations of corrosion attack are visible.

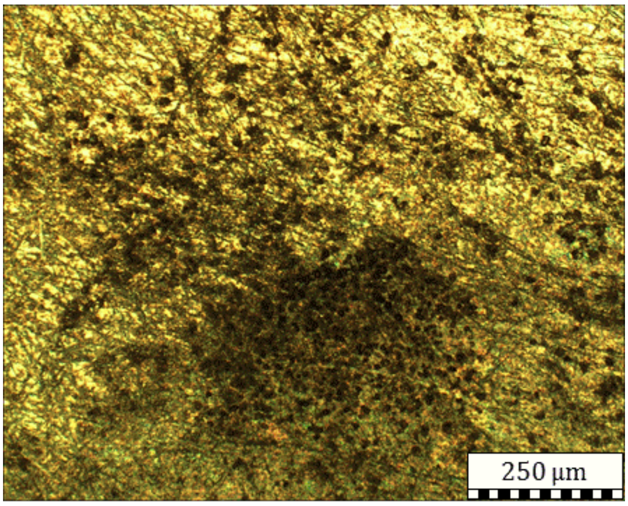

Figure 4-3: Micrograph of part of the area of a working electrode, showing locations of corrosion attack.

The ECN signals of the pitting process towards day 23 show increasingly distinct transients, which is a remarkable difference with the ECN signals obtained earlier. Figure 4-4 shows the evolution of the ECN transients over time at days 14 (a), 22 (b) and 23 (d).

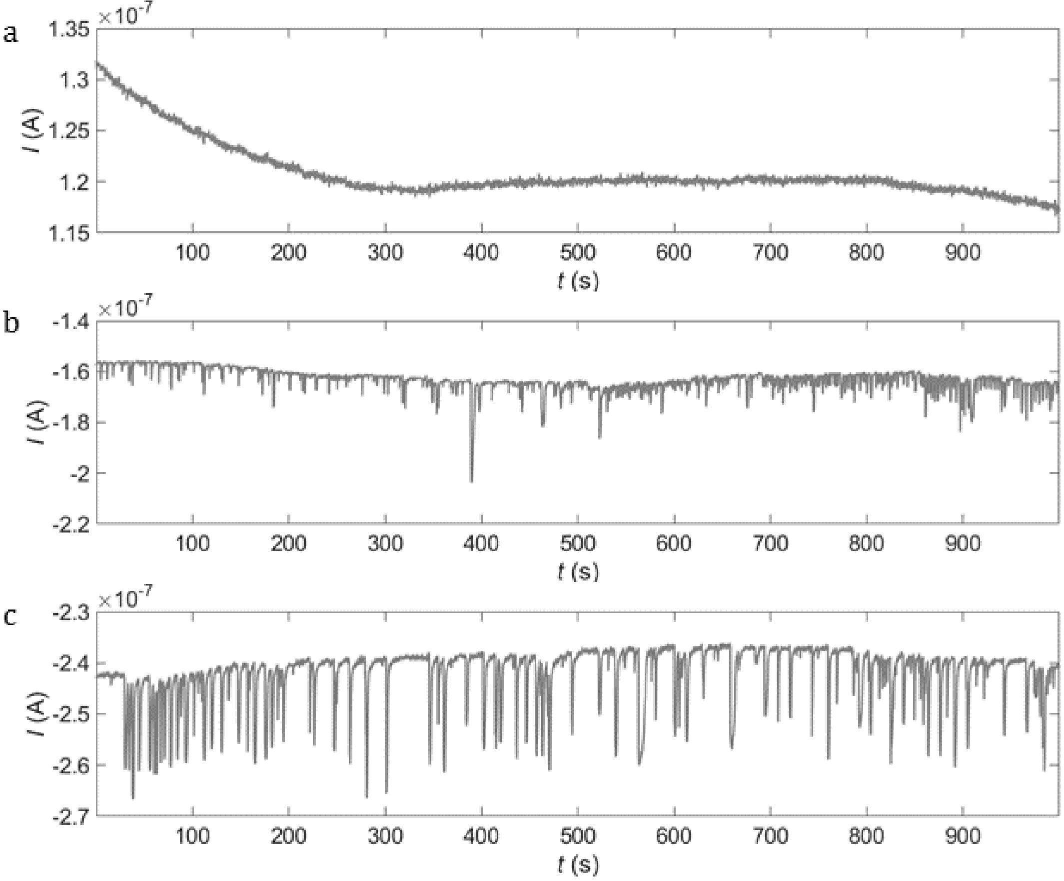

Figure 4-4: Evolution of the ECN transients over time at days 14 (a), 22 (b) and 23 (d).

In Figure 4-4a, the ECN signal consisted of a white noise. Combined with the offset of the mean value of the ECN signal, this is indicative of an anaerobic general corrosion process. The amplitude of the noise pattern visible is in the order of 1nA. At day 22, transients are present in the ECN signal, visible in Figure 4-4b. Those transients already had an amplitude of up to 10nA, with one larger one in this particular ECN signal, around t=390s. Gradually, the amplitude of the transients increased up to 30nA at day 23, when the experiment was terminated and microscopic inspection of the working electrodes followed. It is then interesting to combine time-frequency information of the transients from Hilbert spectra with visual observations. The Hilbert spectra of Figure 4-4b and c are shown in Figure 4-5.

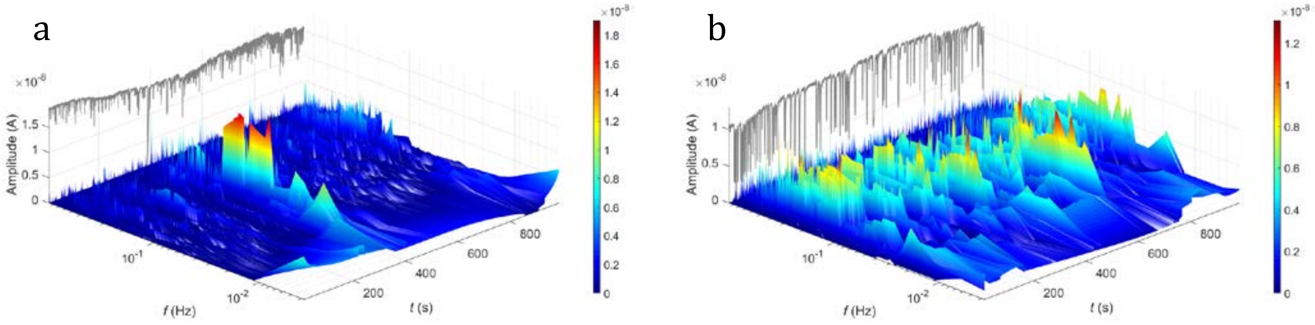

Figure 4-5: Hilbert spectrum of the ECN signal of Figure 10b (a) and c (b).

Analogous to the Hilbert spectrum from Figure 3-5, the original ECN signal is depicted against the back of the spectrum with its relative amplitudes. Already in Figure 4-5a, the instantaneous frequency decomposition of each transient is visible and can be distinguished from the areas in between the transients, where a white noise (i.e. frequency independent noise) pattern is visible. As mentioned before, this pattern is indicative of a slow, anaerobic general corrosion process in this case. In Figure 4-5b a shift of dominant instantaneous frequencies towards the lower frequency range is observed as compared to Figure 4-5a. The transients visible in Figure 4-5a were smaller and relatively short, and can be associated with an onset of pitting in this case. Charge transfer at this earlier stage of pitting only lasts for a very limited span of time, and many pitting events take place at the surface, resulting in many small transients. The shift towards lower instantaneous frequencies in Figure 4-5b indicates a pitting process that generates longer transients and hence a greater extent of pitting attack. As the bacteria develop the biofilm and show increased metabolistic activity, the resulting pitting attack underneath the biofilm is also likely to increase. In Figure 4-5b, a shift towards lower instantaneous frequencies is already visible in this single Hilbert spectrum; after approximately 400s the dominant instantaneous frequencies shift from around $2*10^{-1}$ Hz to around $8*10^{-2}$ Hz. Finally, analogous to the Hilbert spectrum of Figure 3-5 it is worth to mention that the single large transient occurring at around t=390s also dominates the Hilbert spectrum (since this transient is associated with the largest energy compared to other transients), with clear maxima in the instantaneous frequency area from $7*10^{-2}$ to $2*10^{-1}$ Hz.

5.0 CONCLUSIONS

With the aim to develop predictive maintenance concepts for military vehicles, a specific method for corrosion monitoring has been discussed. Compared to other common corrosion measurement techniques, the detection of electrochemical noise (EN) is simple and straightforward. This can be regarded as an important asset for its application in practical corrosion management solutions. The most important step in obtaining relevant information about the corrosion process is however the analysis of the EN signal. This is in most cases a relatively complicated procedure, but can be automated to a large extent. Over recent years, together with the co-authors of our scientific work, we published the development of a novel procedure to analyze transients in EN, with the aim to obtain ‘fingerprints’ of the corrosion processes. These allow the asset manager to acquire valuable information on the type of corrosion, which had not been possible under representative practical conditions before. Pitting corrosion of stainless steel and microbiologically influenced corrosion of carbon steel served as examples to illustrate the potential of the technique.

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