Regularities and features of acoustic emission under plasma electrolytic oxidation of wrought Al-Mg alloy

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Abstract. The paper highlights the characteristic features of acoustic emission (AE) in the process of plasma electrolytic oxidation (PEO) of aluminum alloy, and also reveals the correlation of 4 main stages of PEO process with AE signals. The fundamental possibility to establish and detail the features of oxidation stages and to compare different PEO modes by AE signals was demonstrated. The results obtained substantiate the high potential of AE method as an instrument of in situ research, production monitoring and control, and evaluation of quality of PEO process on aluminum alloys.

1. Introduction
Plasma electrolytic oxidation (PEO) is one of the main and most promising methods of electrochemical thermal treatment of non-ferrous alloys (Al, Mg, Ti, Zr, Nd, etc.) allowing to produce "coatings" (oxide layers) with high mechanical, anticorrosive, thermal insulation and electrical insulation properties on the working surfaces of parts [1, 2]. However, the modern approach to the development of PEO technology and its adaptation to specific technical tasks normally consists of the empirical selection of process current parameters and electrolyte component compositions in order to achieve the best properties of the forming oxide layers that meet the product requirements. In this regard, most of the studies were carried out in different (incomparable and non-reproducible) conditions – different electrolytes, electrical processing modes, different treated alloys. Therefore, deriving common regularities that allow predicting the effect of a given variable factor on the oxide layer properties, or determining the optimal parameters to achieve the best oxide layer properties cannot be accomplished.

Many authors noted [2–6] that in order to obtain high-quality PEO oxide layers on non-ferrous alloys it is necessary to monitor the oxidation process in situ at the structural level. Concurrently, the problem is most vital for PEO of casing-type products with a complex shape (cylinder blocks, compressor housings, etc.), for which several working surfaces are treated in one process stage. However, methods and technical solutions for in situ monitoring and adjustment of the PEO process are still far from real practical applicability and require research and improvements at the level of devices and systems.

One of the promising methods for PEO mode monitoring may be the acoustic emission (AE) method [7]. Yet a very limited number of papers are devoted directly to AE diagnostics of PEO [3, 6] and were successful in proving experimentally that there is a primary correlation of AE nature with PEO electrical mode. Presently, there is almost no understanding of the nature and features of AE at
the main stages of oxide layer formation in PEO on various alloys. Therefore, the purpose of this work was to study the patterns and features of AE during PEO of wrought Al-Mg alloy to assess the possibility of creating in situ monitoring and diagnostic control tools. This will make it possible in the future to create highly sensitive precision control systems for PEO modes for the formation of oxide layers on products of complex shape.

2. Experimental materials and methods

It is known that the recorded AE signal has minimal distortion only near its source [7], therefore, AE sensor should be placed directly on the specimen, and as close as possible to the region of actual process flow. However, the fulfillment of this condition during PEO is difficult, because of the high voltage and electrical current flowing through the electrolyte between the specimen and the electrode. Thus, without a special auxiliary circuit, such measurements will inevitably result in damage to expensive AE equipment. Therefore, before performing this work, a special measurement circuit for registering AE from a specimen with three protective barriers was developed, its essential follows below. Protective barriers should ensure the safety of analog-to-digital converter board (ADC) of the information processing unit (IPU) as the most expensive element of the AE equipment. The dielectric layer between AE sensor (AES) and the specimen was used as the first protective barrier. It was also the AES holder and the acoustic couplant. The second barrier was an interface device (ID) installed immediately at the AES output. The task of using the ID also includes minimizing the losses when transmitting a signal to the external preamplifier (PA) located in a safe zone. An external PA with adjustable gain is the third barrier. Thus, the dielectric acoustic couplant was the protection cornerstone. Should its breakdown occur, inexpensive ID will burn out. However, since it is located almost directly at AES within the electrode bath area, the connecting cable is likely to fail. In this case, and also if the breakdown passes through ID, the external PA burns out. The power supply of intermediate devices (ID and PA) is low-voltage and independent (from external rechargeable batteries). No-filler epoxy glue was used as a dielectric acoustic couplant; AES is band piezoelectric sensor П111-0.02-0.3 (Russia); PA is 2-cascade broadband filter amplifier MSAE-FA010 (Russia); ADC is 4-channel 12-bit Advantech PCI-1714 board (Taiwan); ID is 2-cascade broadband preamplifier with zero gain (own make). The results of approbation demonstrated the operability of this measurement circuit with the possibility of amplifying acoustic signals without increasing the level of electrical crosstalk that unavoidably accompany measurements during PEO and that prevailed over the level of AE signals in other measurement circuits reviewed (AE registration through a bath [3]; through the air with a microphone [6]; through a waveguide welded to the specimen; immersionally, by immersion of the insulated AES in a bath). The operability of the proposed measurement circuit was confirmed using a calibrated acoustic pulse generator MSAE-UCA01 (Russia) according to technique [8].

In the process of PEO, AE was recorded continuously using a standard ADC recording program in the range of 0.02-1.0 MHz with a sampling frequency of 2 MHz. However, given the gradual changing of PEO process, AE recording was carried out in frames lasting 40 ms (20 full cycles of exposure) every 2 minutes, which is quite enough to trace any fast (the result of each exposure cycle) and slow (the result of several exposure cycles) changes in the process. Frames of voltage and current changes in the same PEO cycles were recorded synchronously with AE. Comparison and processing of research data were performed using the Octave mathematical package.

Specimens for PEO were made in the form of 200×20×6 mm strips of wrought Al-Mg alloy 1560 (chemical composition, wt%: 6.2 Mg; 0.65 Mn; 0.5 Ti; 0.4 Si; 0.3 Fe; 0.18 Zn; 0.087 Cu; Al – bal.), which is one of the main structural alloys in carriage-, ship-, machine-building industries, etc. PEO of specimens was carried out at alternating current (AC) at the actual surface area 34±1 cm², in an electrolyte based on distilled water aqueous solution with addition of KOH (3 g/l), sodium pyrophosphate Na₃P₂O₇·10H₂O (8 g/l), and sodium metasilicate Na₂SiO₃·5H₂O (12 g/l). All reagents were of chemical purity. Constant temperature of the electrolyte was maintained at 285±3 K with the use of the cooling system during PEO.
The research included investigations of AE under several modes of PEO differing in: oxidation process duration (60, 90, 120, 150 and 180 minutes); current density \( J = 6, 9, 12, 15 \) and 18 A/dm\(^2\), and negative-to-positive pulse ratio (C/A ratio, \( C/A = 0.7, 0.85, 1.0, 1.15 \) and 1.3). A total of 125 variants of PEO modes have been studied. For all PEO modes, current frequency (500 Hz), process current duty cycle factor (65%), cathodic and anodic forming pulse duration ratio (55%), and interpulse time between cathodic and anodic impulses (50%) were fixed.

3. Results and discussion
In the framework of this work, the results of experiments with the maximum PEO duration (180 min) are provided as most complete examples including all stages of the PEO modes studied. Notably, the following derivations, regularities and features of AE signals in PEO are fully valid for the shorter PEO durations.

The main stages of oxide layer formation during PEO of aluminum and magnesium alloys are known to be distinguished visually, and using photo and video recording [5] (figure 1), or photoelectric effect registration [5, 8], or by changing the peak voltage (anodic forming A. Gunterschulze curves), or by the voltage curve changing shape during the exposure cycle [2, 4, 5]. The main PEO stages are the following (figure 1):

I - anodizing and the beginning of anodic "sparking" (AS), which is visually accompanied by a general glow of the specimen;

II - PEO with predominantly anodic microarc discharges (AMAD), in which uniform and merging micro-flashes of white or blue-white color are visually observed on the overall dark background of the specimen;

III - transition (mixed) mode of PEO with the occurrence of microarc discharges in the anodic and cathodic half-cycles (ACMAD), in which a visual pattern similar to stage II is observed, but micro-flashes are more rare and powerful (bright), and with the beginning shift of their color to a white-yellow tint;

IV - PEO mainly with cathodic polarity including the formation of local arc discharges (AD) visually representing a "soft glow" from inside the specimen, mainly of yellow or yellowish-red tint.

The established optical and electrical signs of PEO stages were also observed in our experiments and were used to determine timing marks of the beginning and the end of the defined oxidation stages I-IV. Based on the timing marks, it was revealed that at each PEO stage, a characteristic AE type corresponds to cycles of electrical voltage, which is illustrated by the example of a mode with a current density of 6 A/dm\(^2\) and a current ratio \( C/A=1.15 \) (figure 1) as including all four characteristic stages of the PEO process.

Figure 1 shows that stage I is characterized by continuous AE (a large number of burst signals of comparable amplitude) in the anodic part of the exposure cycle and almost no AE in its cathodic part, which becomes clear from the chronological recording frames of voltage and AE amplitude changes in one full exposure cycle. At stage II, amid a continuous AE, higher amplitude distinguishable AE burst signals appear in the anodic part, and AE signals also appear in the cathodic part of the cycle. At stage III, AE signals are also present in the anodic and cathodic parts of the cycle, but at the anodic half-cycle it is mainly formed by burst acoustic signals. At stage IV, AE signals virtually disappear in the anodic exposure half-cycle (they become low-amplitude) and are observed after powerful AE signals appear in the cathodic half-cycle. However, over time, the repetition frequency of AE signals noticeably decreases, i.e. AE does not occur in every cycle of exposure. From the same recording frames, the shape of voltage curve is seen to be changing at each PEO stage, which is consistent with the data [2, 4]. Furthermore, by peak voltage values, the result of determining the stages also corresponds to the inflection points of the A. Gunterschulze anodic forming curves, built with account for the high voltage region [2, 4].

Thus, by comparing AE to exposure cycles, the stages of PEO can be recognized and the times of their beginning and end can be determined. Further, using this AE information, an general comparison
(evaluation) of various PEO modes can be performed by the stages achieved in each mode, and also by the duration of processing at each stage. The latter is illustrated by the example shown in figure 2, in which AE records (similar to the one provided in figure 1) obtained when accompanying different PEO modes with the same process duration (180 min) are summarized.

**Figure 1.** General pattern of AE amplitude change staging in cycles, where: I - IV are PEO stages; photographs at the top are visual signs of PEO stages per [5]; inserts at the bottom are chronological record frames of AE (red line) and voltage (blue line) in the exposure cycle; amplitude and time are presented in ADC samples (smpl).

| $J, A/dm^2$ | 0.7 C/A | 0.85 C/A | 1.0 C/A | 1.15 C/A | 1.3 C/A |
|-------------|---------|---------|---------|---------|---------|
| 6           | I       | I       | II      | II      | II      |
| 9           | II      | II      | III     | III     | III     |
| 12          | III     | III     | IV      | IV      | IV      |
| 15          | III     | III     |         |         |         |
| 18          | III     | III     |         |         |         |

**Figure 2.** The general pattern of AE amplitude change depending on the PEO mode with oxidation duration of 180 minutes (designation of AE stages as in figure 1; designations of the control parameters are transcribed in the text).
In figure 2, the boundaries of PEO stages are plotted as per AE signs in accordance with the foregoing data and which are also confirmed in two ways: firstly, consistency with the visual signs of PEO stages; secondly, consistency with the change in PEO voltage. In addition, the indicated AE behavior is in line with modern concepts of PEO sequence [1, 2] according to which the process begins (stage I) with primary passivation and anodizing of the surface across the entire area of the specimen immersed in the electrolyte as a result of electrochemical reactions. In this case, there are many surface AE sources of low power, signals from which overlap with each other. Therefore, it is logical that their resulting signal is of a noise-like (continuous) type. Then, at stage II, an electrical breakdown of the forming thin passive (barrier) layer begins, which is accompanied by high power acoustic pulses in the anodic half-cycle. As the oxide layer grows and the voltage increases, the occurrence of oxide layer micro-arc breakdown begins in the both the anodic and the cathodic half-cycles (stage III). At the same time, many authors note that cathodic breakdown is lower in power and occurs at the metal substrate (in the coating depth), which is unfavorable as regards its capturing by optical methods [5, 8]. However, this is beneficial for the AE method, since the signal is actually generated on an acoustic waveguide (specimen), on which the AE sensor is directly installed. At stage IV, the process almost completely goes “inside the coating” that has a high resistance to propagation of AE signals (due to scattering and attenuation of waves inside the ceramic oxide layer and reflection at the substrate interface). Therefore, it is accompanied by a decrease in the amplitude of AE signals in the anodic half-cycle, but, for the reason indicated above, the AE signal remains in the exposure cathodic half-cycle.

Therefore, the AE method makes it possible to detail the stage-by-stage course of the PEO process on various alloys surfaces and, by this way, to monitor in situ the evolution of the oxidation layer. It should also be emphasized that the information already available is sufficient to adjust the operating mode of the power supply or to temporarily turn off its individual circuits/processed objects.

4. Conclusions
The applicability of AE method for in-situ monitoring and optimization of PEO modes was demonstrated by the found exact correlation of acoustic emission modes with the stage-by-stage course of the PEO process established by other research methods. Identification and correction of PEO mode deviations from the standard (reference) mode, detailing the parameters of PEO process on each of the treated surfaces during group processing, and also the timely stop of PEO before the process begins to move over to the next (undesirable) stage, like to the area of destructive arc discharges - all this only makes an incomplete list of tasks that can be solved using AE-monitoring of PEO process today. This calls for establishing the reference values of AE signals in the amplitude and spectral regions that are most sensitive to the PEO stages, which is the subject of further research.

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