UDK 544.23: 621.792.4
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 58(1)3(2024)

A NON-DESTRUCTIVE METHOD FOR EVALUATING THE ADHESION BETWEEN THE LAYERS OF METAL-POLYMER-METAL COMPOSITE MATERIALS

NEPORUŠNE METODE ZA OVREDNOTENJE ADHEZIJE MED PLASTMI KOMPOZITNIH MATERIALOV V SESTAVI KOVINA-POLIMER-KOVINA

Vladimir A. Glushchenkov[1], Heinz Palkowski[2,1,3], Rinat Y. Yusupov[1], Denis A. Matveev[1], Mikhail V. Khardin[1,*]

[1]Samara National Research University, Metal Forming Department, Samara, Russia
[2]Clausthal University of Technology, Institute of Metallurgy, Clausthal-Zellerfeld, Germany
[3]Clausthal Center of Materials Technology, Clausthal-Zellerfeld, Germany

Prejem rokopisa – received: 2023-02-07; sprejem za objavo – accepted for publication: 2023-07-31

doi:10.17222/mit.2023.727

Development of a non-destructive measuring technique for evaluating the bonding quality of metal-polymer-metal composite sheet materials is introduced. Sufficient adhesion is necessary to ensure further shaping steps or bearing loads when used without failure due to delamination. Much attention is paid to determining the quality of gluing. According to the patent-literature review, no non-destructive measuring or quality control of layers' bonding quality over the entire surface has been found. Thus, a new technical solution is proposed for defining the quality of bonding between the layers by means of electrodynamic repulsion forces arising from the pulse current passing over the metal layers of a composite in the counter direction. The feasibility of the method is tested in laboratory conditions. For its implementation, the necessary values of the test parameters are obtained, i.e., the values of the current transmitted through metal layers. In addition, the possibility of detecting local defects of different shapes and sizes with the proposed method is estimated.

Keywords: sandwich composite sheet, adhesion strength, magnetic pulse, non-destructive measurement

V članku avtorji predstavljajo razvoj tehnik neporušnega testiranja in ovrednotenja kvalitete vezave med plastmi kompozitne "sendvič" pločevine vrste kovina-polimer-kovina. Dobra vezava (adhezija) med plastmi te vrste kompozita je potrebna zato, da pločevina prenese nadaljnje stopnje preoblikovanja ali obremenitve, ne da bi pri tem prišlo do delaminacije oziroma razslojevanja. Veliko pozornosti je namenjeno določevanju kvalitete lepljenja. Med pregledom razpoložljive patentne literature za te vrste metod, metod neporušnega merjenja ali kontrolnih metod kvalitete vezave med posameznimi plastmi po celotni površini so avtorji ugotovili, da le-te v pisni obliki ali predpisih ne obstajajo. Avtorji v članku predstavljajo novo tehnično rešitev za določitev kvalitete vezave med plastmi polimera in kovine na osnovi nastajajočih elektrodinamičnih odbojnih sil pri prehodu preko kovinskih plasti kompozita v nasprotni smeri. Izvedljivost metode so avtorji testirali v laboratorijskih pogojih. Za izvedbo te metode so ugotovili potrebne testne parametre, kot so na primer vrednosti jakosti toka, ki se prenaša s kovinskih plasti. Dodatno so ocenili tudi kakšne so možnosti, da s predlagano metodo določijo lokacijo prisotnih napak, njihovo obliko in velikost.

Ključne besede: kompozitne "sendvič" pločevine, adhezijska trdnost, magnetni pulzi, neporušne metode testiranja

1 INTRODUCTION

Among the composite materials used in mechanical engineering, a special one is a multilayer sheet material consisting of covering metal layers and a polymer core, thus having a "metal/non-metal/metal" combination.[1,2] In the automotive industry, steel-polymer-steel sheets or combinations of aluminum and polymers are already in use.[3,4]

A new 3-layered material of aluminum cover sheets and a PP/PE polymeric core, abbreviated as Al-P-Al, for aerospace and automotive applications is developed. Similar lightweight materials can also be used for medical purposes, e.g., for prosthetic applications.[5]

*Corresponding author's e-mail:
Khardin.mv@ssau.ru (Mikhail V. Khardin)

As shown in **Figure 1**, for the industrial use the manufacturing of such materials includes gluing of the metal and the polymer. To improve the quality of bonding, an epoxy is used in such cases as the promoter. However, in the case of biomedical use, epoxies have to be avoided. Here, other solutions have been developed.[5–7]

For testing the bonding quality, destructive test methods are used in general, such as shear tests or pull-off tests. They require the cutting of the samples for testing (witnesses), which helps us define, for example, the strength of gluing layers in case of their separation. For such pull-off tests, a glue with a bonding strength higher than that between the layers of a sandwich has to be used between the open surfaces of a metal and bracket. For metals, metal bonding glues with a pull-off strength of more than 25 MPa are available, making sure that failure by rupture will occur between the layers of a sandwich

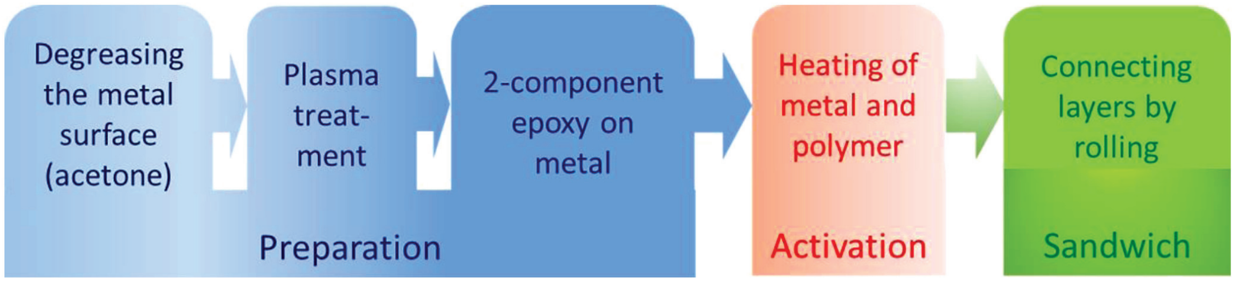

Figure 1: Basic technological operations used for manufacturing multilayer composite materials Al-P-Al

with their lower bonding strength. For its testing, a sandwich is glued to the brackets of a testing machine and this set-up is loaded with tension until failure of the sandwich or the brackets. At the same time, mechanical adhesion is estimated, i.e., the fastening strength of the glued surfaces.[8] Up to now, no non-destructive method for determining the quality of the adhesive strength has been known.

A number of non-destructive methods for detecting local defects (glueline/separation defect) with acoustic, radioscopic, optical and other physical methods are known and available on the market.[9–11] However, they do not allow estimating the adhesive strength quantitatively in the local area of a defect.[12]

The absence of a reliable quantitative non-destructive method of controlling the quality of glued layers in a sheet composite material makes its use more difficult in the industry manufacturing high-quality parts. On the other hand, highly controlled quality of bonding opens up the possibility of a wider use of sheet stamping operations in manufacturing parts from a multi-layer composite material.[13] Consequently, the development of a non-destructive quality control method for the analysis of the bonding quality between the glued metal and polymer is of specific interest.

2 EXPERIMENTAL PART

2.1 Material and methods

Currents induce magnetic fields. When a pulse current is fed in the oncoming direction along two closely located conductors, a pulsed magnetic field is formed around each of them. The interaction of these fields leads to the creation of electrodynamic repulsion forces.[14]

Using this physical phenomenon, a non-destructive testing method for determining the quality of bonding between layers of Al-P-Al composite materials is proposed as sketched in **Figure 2**.

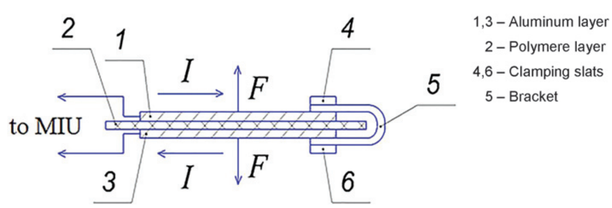

Figure 2: Scheme of the non-destructive testing set-up for magnetic pulse installation (MIU) to determine the bonding quality in Al-P-Al sandwich composites

Metal sheets (1 and 3) with a polymer foil (2) between them are connected to the output of the magnetic pulse installation (MIU).[15] Their electrical closure is carried out using the bracket (5) and clamping slats (4 and 6).

When the battery is discharged by MIU capacitors to the metal sheets, the pulsed current I flows in the counter direction. In this case, electrodynamic forces arise, affecting the separation. If the value of force F, generated by the field, is greater than the predetermined adhesive strength between the layers, delamination will occur; if it is lower (with a given stock ratio), the integrity of the material is not violated and the material is recognized as suitable for further processing.

2.2 Samples for experimental verification of the proposed technical solution

For our experiments, a layered composite (Al-P-Al) was used with a total thickness of 0.9 mm, consisting of two aluminum sheets (AlMn) – each 0.3 mm thick – separated by a 0.3 mm thick polymer foil. The mechanical properties of the AlMn alloy in the annealed state are listed in **Table 1**;[16] so are the ones for the polymer, a thermoplastic polyolefin (polypropylene, polyethylene and talc, PP/PE).[17]

Table 1: Mechanical properties of the AlMn alloy and polymer, PP/PE

Material state	Young's modulus, GPa	Yield strength, MPa	Ultimate tensile strength, MPa	Elongation to rupture, %
AlMn annealed	69	127.4	49	20
PP-PE	11	–	33 ± 11	500

2.3 Breakdown voltage of the polymer layer

In the process of a working cycle, when connecting to the MIU, a high-voltage potential is generated between the plates, which can lead to an electrical breakdown of the polymer. That is why it is necessary to determine its breakdown voltage. The samples are connected to the high-voltage output of the AID-70M testing machine allowing the testing of electrical insulating materials (dielectrics) up to 70 kV. To prevent sur-

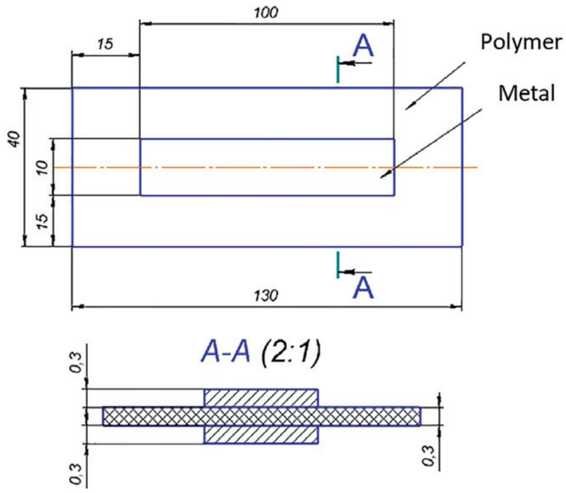

Figure 3: Sample drawing for experimental studies of the adhesion strength

face breakdowns (over the ends), the polymer foil protrudes the metal sheets by about 15 mm at all sides for the tests.

For the experimental investigation, samples sketched in **Figure 3** were used. As a result, the breakdown voltage of the polymer is ~17 kV. This makes it possible to carry out experiments to assess the quality of gluing in a wide range of MIU modes, with different values of the current transmitted through the layers, and determine the geometric sample dimensions to verify the proposed technical solution for the gluing quality control.

2.4 Determination of the adhesion strength between the composite layers

At this stage of processing MPM materials, their adhesive strength is appraised using experimental results of the pull-off or shear tests, therewith defining a tolerable process window and keeping the processing parameters within this field. Up to now, there has been no indicator for evaluating the adhesive strength to be used in-line and as a non-destructive test. The method introduced below could be a solution to this issue.

The evaluation of the adhesion strength is carried out on Al-P-Al sheet samples of 10 × 10 mm^2. Extension cords are attached to the metal parts by spot laser welding to fix them in the taps of a Tinius Olsen H5KT test machine. **Figure 4** shows the typical force-elongation diagram.

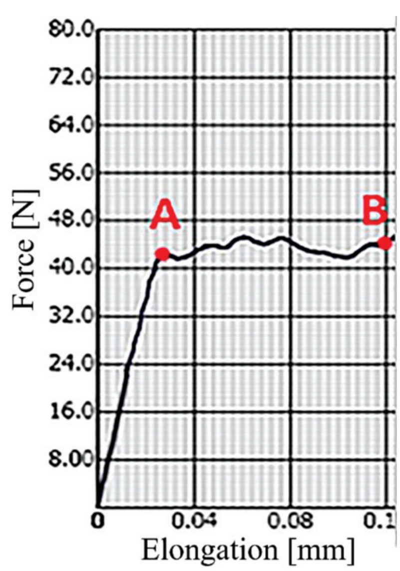

Figure 4: Indicator chart of testing for the separation of Al-P-Al sandwich

Up to point A a pure elastic deformation of the test sample is given, while seperation between the Al-P layers occurs within section A–B, see **Figure 4**.

The experiments performed allow determining the adhesion strength indicator to be č(40–50) N. This indicator has to be compared with the experimental values of the electrodynamic forces obtained with the proposed test method.

2.5 Determination of electrodynamic efforts

The magnitude of the electrodynamic forces between two infinitely thin conductors of the same length is defined with equation (1)[18]

$$F = \frac{\mu_0}{4} \cdot I_1 \cdot I_2 \cdot k \cdot k_F \tag{1}$$

where the magnetic air permeability

μ_0 equals $4\pi \times 10^{-7}$ H/m,

I_1, I_2 – currents flowing in the parallel conductors,

k – geometric factor or circuit coefficient of the electrodynamic forces,

k_F – coefficient taking into account the shape and size of conductors.

The geometric factor depends on the length of the conductors, i.e., their range distances:

$$k = \frac{2l}{a} \cdot \left[\sqrt{1 + \left(\frac{a}{l}\right)^2} - \frac{a}{l} \right] \tag{2}$$

l – length of the conductors,

a – axial distance between them.

Given that the distance between the conductors is significantly lower than their length, we get:

$$k = \frac{2l}{a}$$

k_F takes into account the shape of the cross-section of the conductors, that is, the form of the power lines of the magnetic field. This calculation is carried out with the help of Dwight's diagrams.[19]

The discharge current of the capacitor battery is variable. The value of I_{max} is tens of kA. For the calculation, the following current equation is used:

$$I_D = \frac{I_{max}}{\sqrt{2}}$$

Given the same current value for both conductors (in our case, for both sheets of the composite) we obtain an expression to calculate the maximum electrodynamic force:

$$F_{max} = 10^{-7} \cdot I_D^2 \cdot \frac{l}{a} \cdot k_F \quad (3)$$

For the sample dimensions used in the experiments (a = 0.6 mm, b = 0.3 mm, H = 10 mm, L = 100 mm) it is not possible to find the proper k_F coefficient in Dwight's diagram, as with $(a–b)/(h+b) = 0.03$ and $h/l = 30$; a curve with such values is missing there.

To find the fitting values of the electrodynamic forces affecting the separation of the composite layers, the method of their determination through the kinematics of the strip moving under the action of these forces is proposed. On two non-glued plates, the current also skips in the counter direction. The emerging electrodynamic force causes acceleration on one of the sheets ($F = m \cdot a$). High-speed recording of this process allows us to photograph the kinematics of the central free-moving strip in the initial time stage and calculate the value of F.

Figure 5 present the principal sketch of the measuring arrangement created for this purpose and the devices used.[20]

The principle of the operation is as follows: launching of the camera (6) and charging of the battery to the desired level of the magnetic-pulse installation capacitors take place simultaneously (1). Both the light source and the synchronization unit (3) are activated (7) due to the signals from the computer (4) and the camera (6). The synchronization unit with a defined delay gives off a triggering pulse to the discharge of the magnetic-pulse installation and the oscilloscope sweep (2).

The delay time (delay) of the discharge current is less than 1 µs. At the same time, the signal of the discharge current from sensor 9 is recorded with the oscilloscope. A diagram of the operation of the measuring set-up is shown in **Figure 6**.

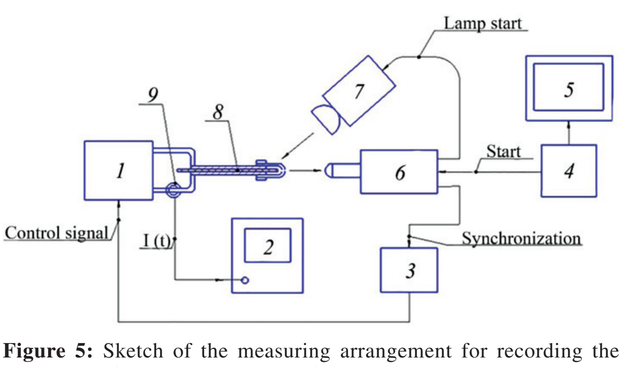

Figure 5: Sketch of the measuring arrangement for recording the movement of the strip under the influence of electrodynamic load
1 – magnetic-pulse installation, 2 – oscilloscope, 3 – synchronization unit, 4 – computer, 5 – display, 6 – high-speed film camera, 7 – pulse backlight, 8 – sample, 9 – pulse current sensor

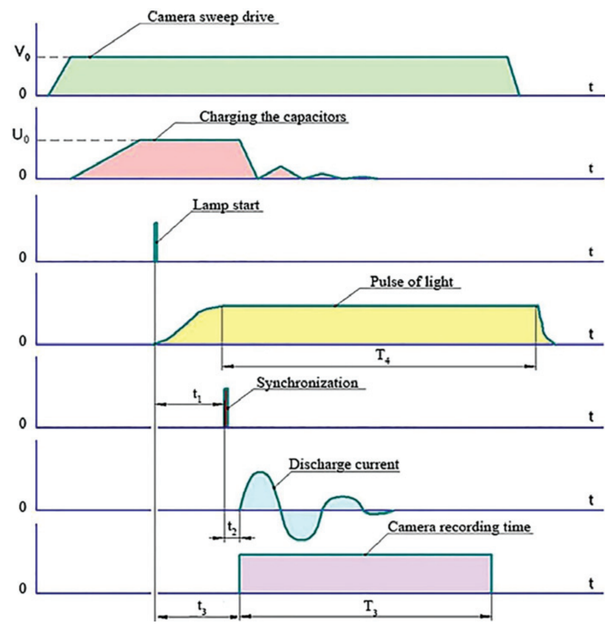

Figure 6: Diagram of the operation of the measuring set-up

The first graph shows the longest cycle of the camera sweep drive. Below is the charging cycle of the capacitors that provide the subsequent pulsed discharge. Next, the backlight turns on to ensure filming. When the lamp is in the operating mode during time t_1, synchronization occurs; after interval t_2 the magnetic-pulse installation is turned on and the capacitor bank is discharged. Simultaneously with the beginning of the discharge, the camera is turned on, carrying out frame-by-frame shooting.

The frame frequency (FPS) of shooting in the experiments is 29740 frames/s. Consequently, the time between the frames is 33.6 µs. **Figure 7** shows four fragments of the sample movement. Starting with the third

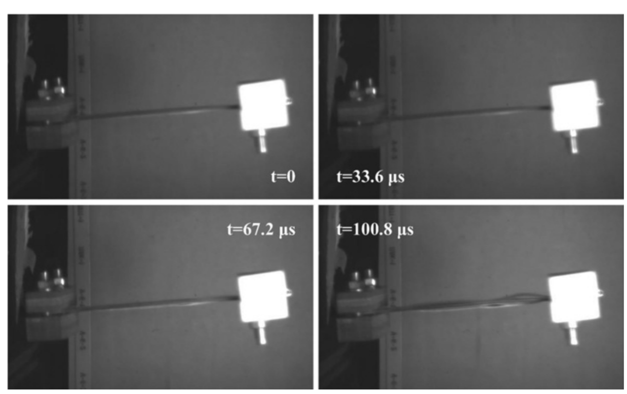

Figure 7: Fragments of recording the layer repulsion process

Table 2: Dependence of the sample movement on time

Sample movement, mm	Time t, s·10^{-6}
0	0
0	33.6
$0.4 \cdot 10^{-3}$	67.2
$1.2 \cdot 10^{-3}$	100.8

frame (67.2 μs), the sample is observed, as indicated by the arrows.

Numerical results of processing the records are given in **Table 2.**

The obtained values allow calculating the acceleration (a = 531462.49 m/s^2) and, knowing the mass of the moving section ($m = 0.25 \times 10^{-3}$ kg), the electrodynamic force (F = 132.87 N).

In addition, the data obtained allow us to get the value of the k_F and apply it in further calculations:

$$k_F = \frac{F_{max}}{2\cdot 10^{-7}\cdot I_D^2\cdot k} = \frac{132.87}{2\cdot 10^{-7}\cdot 295602\cdot 125} = 0.006$$

Thus, all the necessary values included in Equation (1) are obtained to determine the electrodynamic forces arising from the non-destructive quality control of the composite layers.

3 RESULTS

The purpose of the experimental studies is to find the dependence of the force of the metal strip – adhesive strength – on the value of the current transmitted by the metal layers, i.e., to find the minimum current value, at which a high-quality sandwich sample remains undamaged while delamination is observed in a sample with a weak bond.

After each discharge, a visual and instrumental control of the samples is carried out for the impairment of the adhesive strength of the layers: possible deformation of the metallic layers is recorded using a simple dial gauge.

In the absence of any defects, indicated by deformations, a sample is connected again to the MIU and a higher current is created. The current is measured using a RGA belt (a current sensor) and its value is presented on the oscilloscope screen. Results of testing defect-free specimens are listed in **Table 3**.

Table 3: Experimental results: height of sample delamination h at current flow I_d

Sample no.	Current I_d, kA	Height of delamination h, mm
1	12.83	0
	13.59	0
	14.16	0
	16.57	0
	17.50	0.19
2	15.86	0
	17.11	0.17
3	15.80	0
	17.19	0.15

Thus, with a current value of (17.11–17.50) kA, a delamination between metal an polymer is observed, having a bulge of around (0.15–0.19) mm. This current value corresponds to an electrodynamic force of (44–46) N, which corresponds to the experimentally obtained value of the force of (40–50) N (see **Figure 4**).

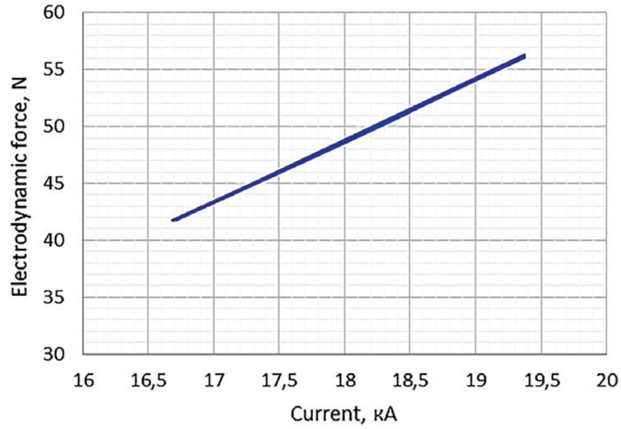

Figure 8: Dependence of the electrodynamic force on the current flow through the metal layers

The results obtained allow a construction based on the dependence of the electrodynamic force on the value of the pulse current passed through the metal layers of the three-layered Al-P-Al composite (**Figure 8**).

If the adhesion strength (F_{set}) is specified for a composite material in technical conditions, then, considering the safety coefficient – for example, 1.2 (F_{set}/1.2) – we find the adequate value of the current being tested (current I) with the help of the constructed graph (**Figure 8**), and through it, we determine the condenser discharge energy of the magnetic-pulse installation, which must be used for a non-destructive test. If, with this current value (discharge energy of the capacitor battery), no delamination is observed, the logical conclusion is that the bonding quality corresponds to the required one.

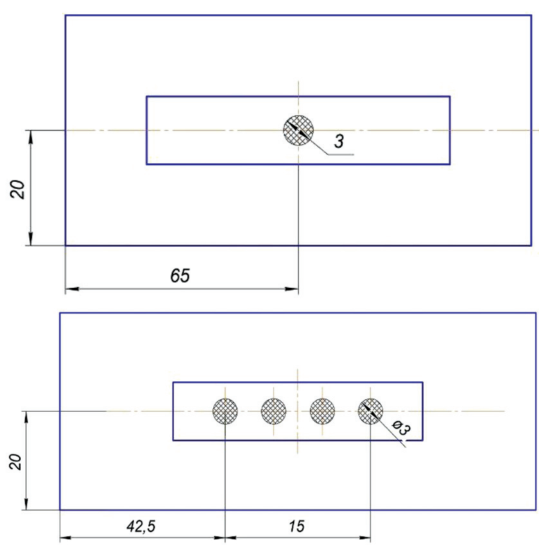

Figure 9: Samples with local defects

4 DISCUSSION

4.1 Possibility to use this method to detect local defects

This non-destructive method is suitable for determining the quality of bonding in general. The question to be answered now is how sensitive it is, e.g., when detecting local defects in a bonding structure. Therefore, three-layered Al-P-Al samples with the same dimensions as before were used, but with pre-established macroscopic local defects of different dimensions in the core material. Circles with diameters of 3, 6, 8 mm, located in the center of the composite as single circles or several ones set in a row were cut into the polymer, see **Figure 9**.

An increase in the diameter of the defects leads to a decrease in the current, at which delamination of a sample occurs. In all the cases, the place of the gap defect is clearly seen when the current is passing through.

The analysis of defective samples carried out during the test has shown that delamination in an Al-P-Al sandwich with a circular defect of 3 mm and height of 0.1–0.3 mm is observed at a current, I_d, of (16 ÷ 17) kA.

The variation in the values for one testing set is explained by the differences in the bonding quality of each sample (standard deviation) and placing the defects (geometrical deviations). However, according to the data obtained we can see some dependence of the current magnitude required for delamination on the size of the defect: the smaller the diameter of the defect, therewith the ratio of the defect, the higher is the force (current) required for delamination of the layers.

The observed local deformation of the strip above the defect proves the possibility of using the proposed method of control for detecting local defects as well.

5 CONCLUSIONS

A non-destructive method for testing the quality of bonding between the layers in Al-P-Al composite sheet materials, i.e., metal/dielectric intermediate layer/metal, is introduced. The first experimental tests show its basic applicability. Defects due to delamination can be detected and correlated with electrodynamic forces and currents. Moreover, implemented defects of a macroscopic size lead to a characteristic change in the currents needed for delamination, depending on the size of a defect. This method also has a potential for detecting smaller defects in such composites. Up to now it has not been possible to quantitatively assess the value of bonding, but we can determine whether the bonding is sufficient for an estimated load, based on the failure/non-failure of the part under defined current loads.

Further tests have been performed to develop an industrially fitting measuring device, a tool able to provide quantitative measures and values for on-line detection of the bonding quality in three- or even multilayer sandwich structures of the Al-P-Al type, consisting, in general, of metal/polymer/metal, or similar structures.

6 REFERENCES

[1] D. V. Aseeva, Assessment of mechanical and technological properties of multilayer materials "metal-non-metal", Master Thesis, Samara National Research University, Samara, 2017, 104

[2] H. Palkowski, A. Carradò, Metal-Polymer-Metal Laminates for Lightweight Application, Key Engineering Materials, 684 (**2016**), 323–334, 20163, ISSN: 1662-9795, doi:10.4028/www.scientific.net/KEM684.323

[3] M. Harhash, O. Sokolova, A. Carradò, H. Palkowski, Mechanical Properties and Forming Behaviour of Laminated Steel/Polymer Sandwich Systems with Local Inlays – Part 1, Composite Structures, 118 (**2014**) 1, 112–120

[4] H. Palkowski, G. Lange, Austenitic Sandwich Materials in the Focus of Research, In: 2nd International Conference on Deformation Processing and Structure of Materials, ISBN 86-85195-06-3, Belgrade, 2005, 19–28

[5] M. Harhash, A. Carradò, H. Palkowski, Lightweight titanium/polymer/titanium sandwich sheet for technical and biomedical application, Materialwissenschaft und Werkstofftechnik, 45 (**2014**) 12, 1084–1091

[6] M. Reggente, M. Natali, D. Passeri, M. Lucci, I. Davoli, G. Pourroy, P. Masson, H. Palkowski, U. Hangen, F. Carrado, M. Rossi, Multiscale mechanical characterisation of hybrid Ti/PMMA layered materials, Colloids and Surfaces A, 532 (**2017**), 244–252

[7] M. Reggente, S. Kriegel, P. Masson, G. Pourroy, F. Mura, J. Faerber, D. Passeri, M. Rossi, H. Palkowski, A. Carradò, How alkali-activated Ti surfaces affects the growth of tethered PMMA chains: a close-up study on the PMMA thickness and surface morphology, Pure Appl. Chem., 91 (**2019**), doi:10.1515/pac-2019-0223

[8] E. Kraus, Increasing the strength of adhesive joints of polymeric materials by laser and plasma surface treatment, Kazan Technological Research University, Kazan 2017, 175

[9] E. H. Schindel-Bidinelli, Konstruktives Kleben, Wiley-VCH Verlag, Weinheim 1988, 325

[10] M. Rasche, Handbuch Klebtechnik, Hanser Verlag, München 2012, 616

[11] A. V. Pocius, Adhesion and Adhesives Technology, Hanser Verlag, Munich 2012, 370

[12] C. J. Van Oss, M. K. Chaudhury, R. J. Good, Interfacial Lifschitz-van der Waals and Polar Interactions in Macroscopic Systems, Chemical Reviews, 88 (**1988**), 927–941

[13] W. S. Gutowski, S. Li, C. Filippou, L. Russel, Mechanisms of effective control of automotive coatings adhesion to polymers and composites, European Coatings Conference, Germany, Dusseldorf 2015, 153–159

[14] G. N. Aleksandrov, Theory of electrical machines, SPbGTU, St. Petersburg 2000, 540

[15] V. A. Glushchenkov, V. F. Karpuhin, Technology of magnetic-pulse processing of materials, Fedorov, Samara 2014, 208

[16] S. G. Alieva, Industrial aluminum alloys, Metallurgy, Moscow 1984, 528

[17] GOST R 57746-2017 (ISO 15107: 1998) – Polymer composites. Determination of peeling strength of adhesive joints, Publishing House of Standards, Moscow

[18] A. S. Grachev, Electric devices: Guide to solve the problems of designing electric devices, Mari State University, Yoshkar-Ola 2009, 111

[19] GOST R 30323-95 – Short circuits in electrical installations. Methods for calculating the electrodynamic and thermal effects of short-circuit current, Publishing House of Standards, Moscow 1994

[20] V. A. Glushchenkov, Methods of studying the kinematic parameters of rapid-flowing processes of magnetic-pulse processing of materials, Samara State Aerospace University, Samara 2011, 52