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**Investigation on creation and properties of laser assisted
metal-plastic joining**

Ph. D. Thesis

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Budapest, 2015

1. Motivation

The growing importance of environmental protection and energy saving ambitions caused significant changes in the materials of our devices, especially in the case of vehicles [1]. Due to weight reduction efforts the proportion of different plastics in our vehicles is increasing, making the partially substitution of traditional materials, like steel possible [2]. However, because of their good heat resistance, stiffness and strength metals often can't be substituted completely by plastic materials. Therefore the joining of metals and plastics is needed. This kind of joining is used more and more often in the practice as well. The hybrid parts made with this joining method consists of metals and plastics as well, thus the expected stiffness of the part can be ensured with significant weight reduction. Therefore hybrid parts can take the advantages of both different materials simultaneously [3, 4].

There are several widespread technologies used for joining of metals and plastics. However, most of these technologies don't enable neither an extensive, universal application nor traditional bond geometries. All of the methods have disadvantages restricting the application and strength of created joining or making their automation difficult [5]. Therefore the aim of present research is the development of technologies which are cost effective, easy automatable, flexible and fast, and which can be applied in mass and vehicle production easily [1, 6].

In the last years, the extreme evolution of laser technologies can be seen. The advantages of the laser beam are the concentrated heat input and so the small heat affected zone and the minimized undesired changes in the base material. Laser beam can be moved along curves by using robots, the output power is programmable and can be controlled easily thus the laser beam is able to flexible processing of 3D geometries as well. Laser beam is well focusable, extraordinary fast and post processing of the part can be avoided, which enables further reduce of process time [7, 8, 9, 10, 11, 12]. The above described advantages make possible to fulfill technological requirements by using laser assisted processes.

One of recent innovations is the application of laser for metal-plastic joining. Although laser are used for plastic welding a long while, and the principle making the laser assisted metal plastic joining feasible is known for decades, this new technology is inspired and brought to life only by recent years trends. The method is intensively researched at several places in the world

because of his large potential to create a reliable, strong and direct bond between metals and plastics. The new technology can meet expectations of mass production and can cause a radical change in today's design and production principles. The joining method described in this dissertation can be able to eliminate the disadvantages of currently used technologies, which can ensure a serious benefit in its future applications.

The aim of the dissertation is to enrich the accumulated knowledge in this topic, to describe the accompanying phenomena in the materials during joining, to define the most important technological parameters influencing the joint quality, and thereby to support the technology's future application.

2. Literature review

To prepare a metal-polymer joining basically a laser transparent plastic and a laser absorbent metal is needed. The joining geometry is mostly overlapped sheet to sheet joining: the laser transparent material is placed at the top of the absorbent material facing directly the laser beam. The beam mainly goes through the upper transparent plastic and is partly absorbed on the surface of the lower metal sheet, where heat is generated. The heated metal gives a part of its heat back to the plastic, which starts to soften and melt. Clamping force is applied between the joined parts during the process to ensure a proper contact. As a result of the clamping pressure the molten plastic establishes a close contact to the steel surface, and fills the microscopic holes and structure of the surface. Finally the adhesion responsible for bond strength is created [13].

The laser assisted metal-plastic joining (LAMP joining) is an actively researched area at several places of the world. The aim of this research is often to create a packaging of micro-electro-mechanical systems (MEMS) implanted in human body. In this cases the joining of polyimide, polyethylene terephthalate and titanium is typical by means Yb:YAG laser source. Researchers detected the presence of Ti-O and Ti-C covalent bonds with XPS technique and showed the bond strengthening effect of increasing surface roughness [14, 15, 16]. The group led by Seiji Katayama is a pioneer of LAMP joining technique, they are owners of many valuable contributions. In the course of their work, they created a bond between austenitic stainless steel and polyethylene terephthalate as well as polyamide in overlapped geometry using a diode laser source. The authors report in each case about

the formation of small bubbles in the plastic material, as a result of thermal material degradation [17]. However the bond strengthening effect of bubbles is supposed due to increased pressure caused by the arising bubbles itself: the high pressure can result in a better contact between joined materials and molten plastic can fill the microscopic holes and structure of the metal surface [18]. Based on XPS results the authors suppose a covalent bond between functional groups of polymer molecule and metal oxides on the surface.

W. Tillmann and her colleagues report about the decomposition of plastic next to the joining boundary as well resulting in a bubble formation in the molten plastic. According to their results the joint strength has an optimum as a function of laser beam moving speed and laser power, which can be explained by the alteration of bubble area [19]. C. Lamberti and colleagues state that the source of joining force is the hydrogen-bond between polar groups of polyamide and oxides of the aluminum surface [20]. The strength of overlapped joints could be increased efficiently with structuring of steel surface: grooves or small holes were created on the steel surface by laser structuring or cutting as a part of several researches [21, 22, 23, 24]. A. Roesner and her colleagues joined polycarbonate and poly(methyl methacrylate) sheets to stainless steel pins by means of diode laser source. The created strong bond could be improved by roughing of steel surface and by using shape locking pin geometry [1].

Summary of literature

According to the contributions laser assisted metal plastic joining can be divided in joints with or without penetration. In the case of penetration joint the heated metal part penetrates into the plastic due to applied clamping pressure, in the case of joint without penetration the joined metal part do not penetrates into the plastic material. In both cases we can distinguish between line and spot-like joining geometry.

Based on reviewed scientific contributions it can be stated, that almost in all cases continuous mode solid state laser sources are applied to create the LAMP joining. Therefore there isn't any information about the behavior of polymeric materials in the case of pulse mode laser irradiation, as well as about the changes of transparency as an effect of short time but large energy laser pulses. Commonly laser transparency of polymers is measured at very low laser powers and by using continuous mode laser beam, so the effect of

irradiation time and the laser material interaction using large laser power is unknown. Because of rare application of pulsed mode laser beam the effect of pulse settings on laser material interaction is unknown as well. Also the thermal conditions during joining are almost unclear. The contributions do not deal with the joining of basic metallic structural materials like carbon steels. Except of one case, all groups are researching overlapped joint geometry. Therefore there isn't any information about spot and penetration joining. However, such joints are in demand in industrial practice. There are several research groups in the world dealing with LAMP joining, nevertheless the accompanying processes of the joining are unknown or their description is insufficient. Many authors mention the bubble formation phenomena, but the exact process of the formation and its effect on bond strength is unclear. There is very little information about the effect of clamping pressure applied during joining, however the pressure at the boundary surface of metal and plastic is stated to be a very important condition of joining mechanism. Since the preparation of spot joints is a barely studied area, there is very little information about the effect of micro- and macrogeometry of metal on the bond strength as well.

3. Research aims

Based on literature review of metal polymer joining the aim of my research was the investigation of a laser transparent, spot, penetration joint created with pulse mode Nd:YAG laser, to establish the basics of a new industrially applied method. Accordingly detailed research aims are following:

- at the field of laser-material interaction:
 - to elaborate a method suitable for measurement of polymer transparency as a function of time under conditions of laser joining;
 - to investigate the effect of laser power, pulse settings and laser spot diameter on plastic transparency used for joining by applying the newly developed method;
 - to define temperature and temperature distribution of metal pin evolving in the case of used laser heating;
- at the field of joint creation:

- to specify the process of bond formation in case of structural steel and plastic most suitable for bond preparation: description of characteristic penetration and bubble formation;
- to explore the correlation between technological settings, like laser average power, pulse parameters, heating time as well as clamping force and penetration depth just as bubble formation
- to describe the correlation between joining force and joint strength influencing parameters like laser average power, pulse parameters, heating time, clamping force, surface roughness, sheet thickness and pin geometry
- to compare LAMP joining with same geometry adhesive joint.

4. Materials, devices and methods used for experiment

In the case of joining experiments primarily poly(methyl methacrylate), while at transparency measurements poly(methyl methacrylate) and polypropylene were used. During joint creation the used sheet thickness was 2 to 5 mm, while in the case of transparency measurements 2, 5 and 10 mm thick sheets were applied. The joints were created by applying 5 mm diameter S235 material steel pins. By comparing LAMP joining with gluing Loctite 454 and Loctite 496 type cyanoacrylate based adhesives were used. In all transparency measurement and joint creation experiments a LASAG SLS 200 type pulse mode Nd:YAG laser source was applied. The experimental setup of transparency measurement is showed in figure 1.

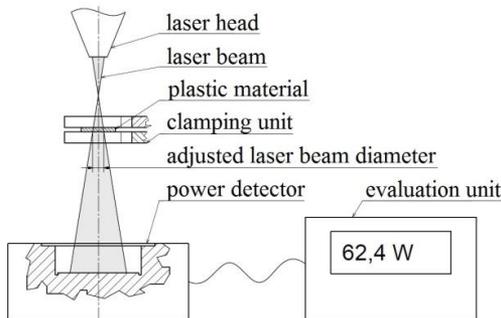


Fig. 1. Experimental setup of transparency measurement

During transparency measurement the examined material was pushed into the way of laser beam after 10 seconds of switching it on, and thereafter the material was irradiated for further 10 seconds. During the process the power of laser beam was measured with a special laser power measuring device. The 10 s long waiting time before pushing the sheet into the way of beam is needed for the measuring device to take the power of incident laser radiation on and to display a constant power value which is characteristic for the laser beam. As a quotient of the measured power after pushing in the sheet and the maximum power value during measurement the transparency can be calculated and its change can be illustrated as a function of irradiation time. The given spot diameter is the diameter of laser beam on the lower, leaving side of the sheet without scattering.

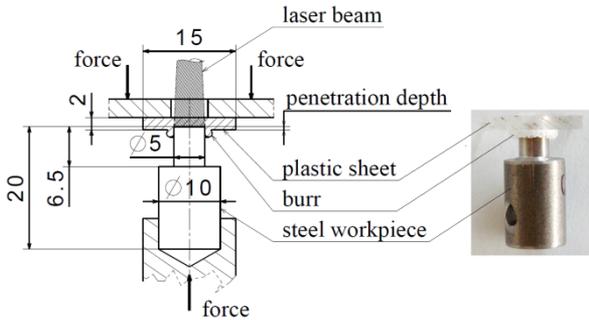


Fig. 2. Setup of joining experiments, geometry of samples and the created joining

The schematic view of joining experiments and the created joining is showed in Figure 2. During the experiment the laser beam mainly goes through the plastic and heats directly the head surface of the metal pin, the beam is coincident with the head surface. During the process the heated pin gives a part of its heat back to the plastic, which starts to soften and melt. Due to applied clamping force the pin penetrates into the sheet. Finally the joining is created in some seconds.

After joining the penetration depth of the pin into the plastic was measured. The joints were qualified with tensile testing.

The pin temperature was measured with a thermocouple welded on the lateral pin surface next to the edge, the temperature distribution was recorded with thermovision camera, according to figure 3.

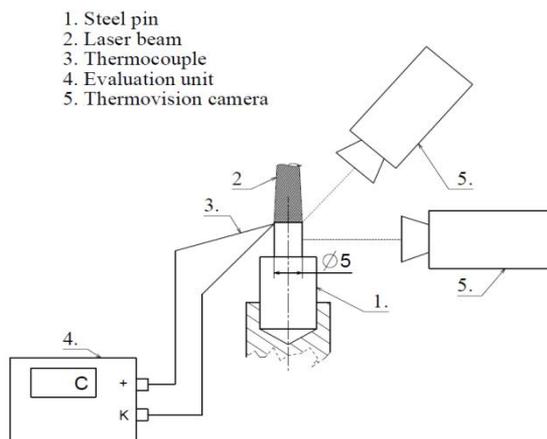


Fig. 3. Experimental setup of temperature measurement

The effect of pin micro- and macrogeometry on joining strength was investigated as well. By investigating the effect of surface roughness the face and lateral surface roughness was increased. Towards further enhancement of joining force joints were prepared with different shape locking geometries (conical, threaded, grooved, flanged geometries).

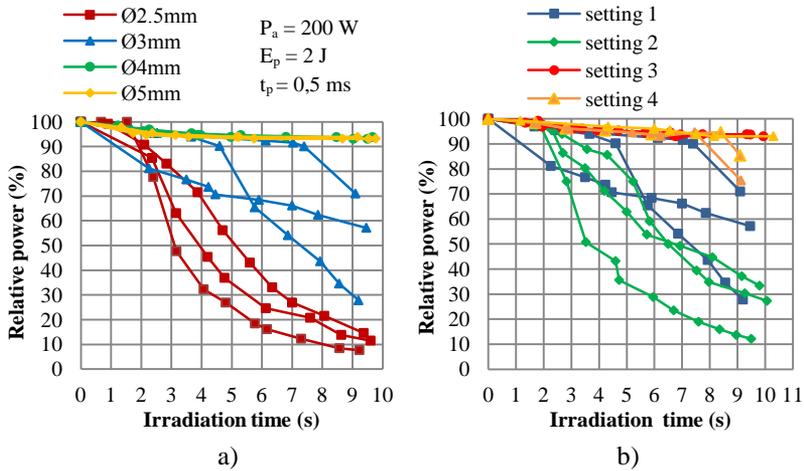
During gluing experiments the samples were created under same conditions as in the case of LAMP joining, by using the same clamping unit as well. In some cases flat bottom holes were drilled into the PMMA sheet and pins were glued into the prepared holes.

To investigate the created samples stereo- and light microscopes as well as scanning electron microscopy were used. The thermal properties of plastic were measured with DSC and TGA method, the decomposition was studied with special gas analyzer device.

5. Results

Using the introduced new method I measured the transparency of various plastics in the case of pulse mode Nd:YAG laser irradiation. The method enables the measurement of transparency separate from polymer thickness and composition, and the change of transparency as a function of irradiation time can be determined as well. Using the described method the investigated polymers can be divided in free groups dependent on their behavior during

laser irradiation. In the first group transparency of the material does not change during the irradiation time under applied conditions. The transparency of second group materials is constant in the first part of irradiation, while it starts to decrease in the second part. The transparency of the materials belonging to the third group is decreasing in the whole irradiation. The reason for decreasing transparency is the thermal degradation of polymer: during degradation there are bubbles forming in the material. The bubbles scatter and reflect laser radiation and thus deteriorate transparency.



Setting Nr.	P_a (W)	f_p (Hz)	E_p (J)	t_p (ms)	P_p (kW)	ρ_a (W/mm ²)	ρ_p (W/mm ²)
1.	200	5	40	10	4	28,3	566
2.	200	100	2	0,5	4	28,3	566
3.	200	10	20	10	2	28,3	307
4.	200	26	7,5	10	0,75	28,3	100

Fig. 4. Effect of laser spot diameter (a) and laser pulse settings (b) on changes of PMMA transparency

In the case of pulse mode laser beam the transparency can be influenced by laser pulse power, laser beam spot diameter, power density and pulse time as well. If the average power density of a pulse mode laser beam exceeds a critical value, the polymer transparency starts to decrease at a moment of irradiation time. However, transparency is not only a function of average power density, but a function of pulse power density as well. If the pulse power density exceeds a certain value, the transparency decreases even if average power density itself does not reach its critical value. The introduced method allows determining the mentioned critical values as well. Exceeding

the critical laser pulse parameters the transparency starts to decrease during irradiation time. The change in transparency of PMMA as a function of laser spot diameter, laser pulse settings and irradiation time is showed in figure 4.

I measured the characteristic temperature distribution of steel pins during laser heating process and the effect of plastic sheet on the surface temperature of steel during joining as well. I showed that the heating speed and maximal temperature of samples are influenced by laser pulse parameters. I also demonstrated that the maximum temperature of steel and plastic during joining is decreased due to 93% transparency of PMMA sheet and due to heat drain of plastic sheet. I defined the characteristic temperature distribution of pin surface, which is of crucial importance from the viewpoint of plastic behavior during joining. The evolving temperature distribution is a simultaneous effect of TEM₀₀ laser power distribution and pin geometry.

I described the process of laser transparent spot penetration joining process and I selected the materials able for LAMP joining. Finally PMMA material was chosen for joining because of its favorable joint strength and good optical properties. Based on the greater or less plastic parts remaining on steel surface after tearing of the samples a strong adhesive bond forms at the boundary of steel and plastic: plastic “islands” also visible with naked eye or thin plastic layer identifiable only with microscope remains on the steel surface (figure 5.), namely in the case of optimal joining the tearing occurs in the base plastic material.

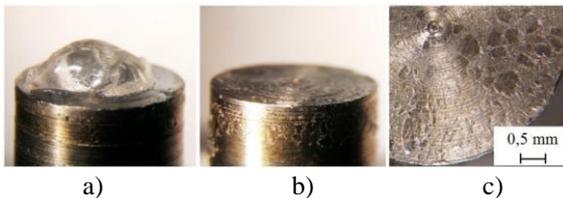


Fig. 5. Remained PMMA on steel surface in the case of 4s (a) and 6s (b, c) heating time

In all joining experimental cases gas bubbles form, which are the product of polymer decomposition; the material temperature material exceeds decomposition temperature of PMMA during joining. There is a correlation between bubble distribution, shape and size and surface temperature distribution of steel, heating time and flow of molten plastic during joining. The size of bubbles increases with increasing heating time, while the amount of bubbles has a maximum as a function of heating time.

According to the measured results, the joint strength is influenced by the laser settings as well. Increasing average power increases penetration too, caused by higher temperature and lower plastic viscosity. The power increase also increases joining force however force is limited by more intensive bubble formation. In the investigated range laser pulse settings have an influence on joint quality only by using high pulse energies. In this case penetration decreases slightly, while strength decreases strongly, caused by the lower arising temperature during joining. The effect of average power and pulse energy is showed on figure 6.

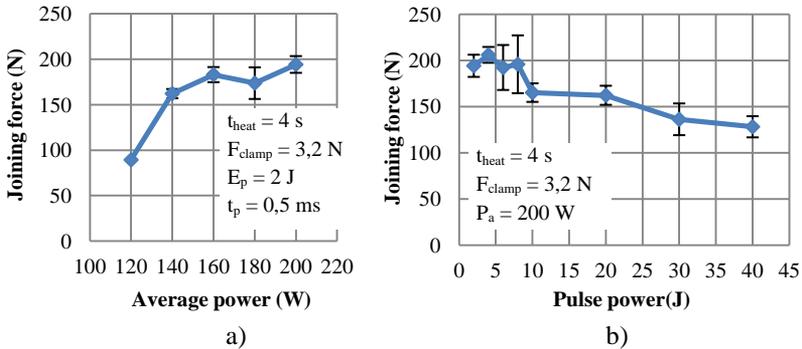
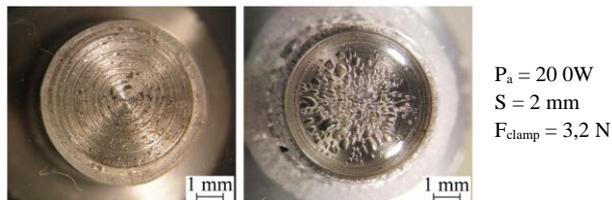


Fig. 6. Effect of laser average power (a) and pulse power (b) on joining force

Most important joint strength influencing parameters are the heating time and penetration depth. By increasing the heating time penetration grows as well due to higher steel temperature and lower plastic viscosity. If penetration depth increases, contact area between joined materials increases too, and bond strength can be improved. However, increased heating time results in a growing amount and size of gas bubbles, which has a weakening effect on the bond, because they decrease load carrying cross section of plastic material. The joining couldn't be created without mentioned bubbles because of characteristic temperature distribution on head surface: at the midpoint of the head surface plastic starts to decompose when simultaneously plastic next to the edge is not soften enough to allow penetration of the pin. Bubbles formed during joining process are demonstrated in figure 7.



a) b)

Fig. 7. Bubble formation at the face surface in case of 3 s (a) and 7 s (b) heating time

Bubbles form primarily next to the face surface of the pin during joining, therefore the role of face surface played in strength of bond decreases with longer heating times. In contrast, the significance of lateral surface in the bond strength increases with increasing heating time because of deeper penetration. The two described phenomena are responsible for typical shape of heating time-joining force diagram, which shows a maximum value. Force increases till bubble formation do not decreases the role of face surface to such a pitch that it can't be compensated by growing role of lateral surface. The role of head and lateral surface is detectable in the shape of tearing diagrams as well. The shrinkage of the plastic material due to different heat expansion coefficient of PMMA and steel raises a normal stress in the plastic which contributes to the strength of joint as well: a shrink-fit like effect is evincible on the lateral surface. The effect of heating time on penetration and strength is shown in figure 8 in the case of joints created with 5 mm thick PMMA sheet.

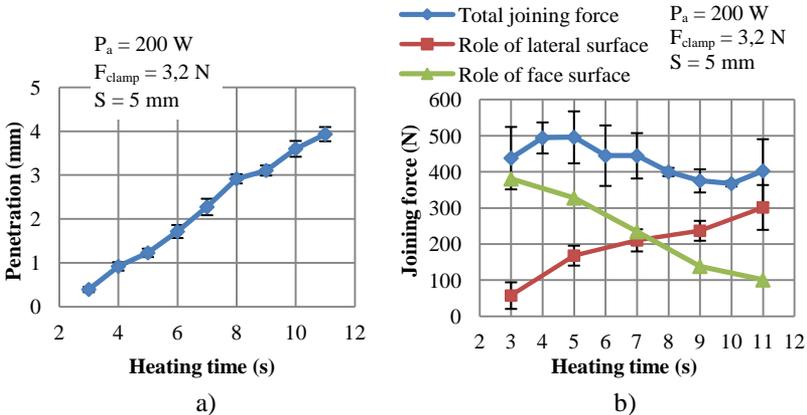


Figure 8. Effect of heating time on penetration (a) and joining force (b) as well as the role of face and lateral pin surface played in joining force formation

Thus the formed bubbles during joining decrease the strength of joint, however by choosing the appropriate clamping force bubble formation can be controlled. Increasing clamping force reduces the speed of bubble formation, therefore the area of bubbles decreases and bond strength can be improved, according to Figure 9.

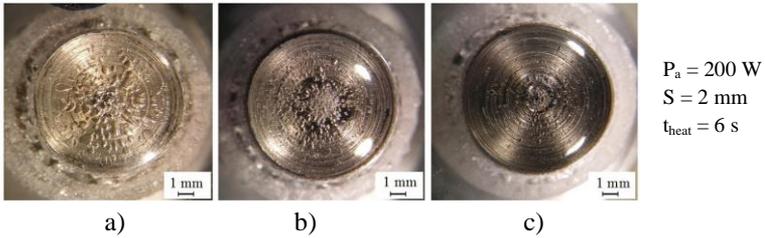


Fig. 9. Upper view of samples created at the same heating time of 6 s and clamping force of 3,2 N (a), 6 N (b) and 9,2 N (c)

The tearing force of the bond can be effectively improved by using an appropriate micro- and macrogeometry of steel and plastic. Increasing head surface roughness improves the efficiency of laser absorption, resulting in higher temperature, deeper penetration and larger contact area. The increased contact area and the growing mechanical adhesion due to increasing roughness cause together the significant improvement of bond strength. The effect of surface roughness on penetration and tearing force is showed in Figure 10. However, the bond strength can be enhanced with increasing plastic sheet thickness as well. The greater thickness ensures bigger sheet stiffness which can reduce bond weakening deformation during tearing. Utmost bond strength can be improved by using shape locking geometry steel pins, which allow a better utilization of plastic sheet strength, and a doubling of tearing force. The effect of sheet thickness and different shape locking pin geometries on tearing force can be seen in Figure 11.

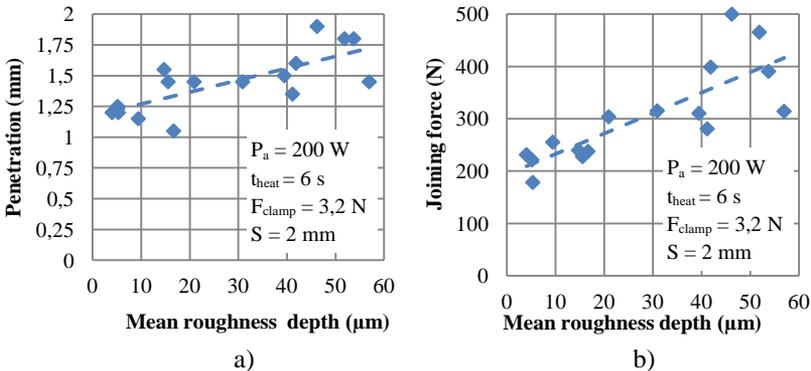


Fig. 10. Effect of surface roughness on penetration (a) and joining force (b)

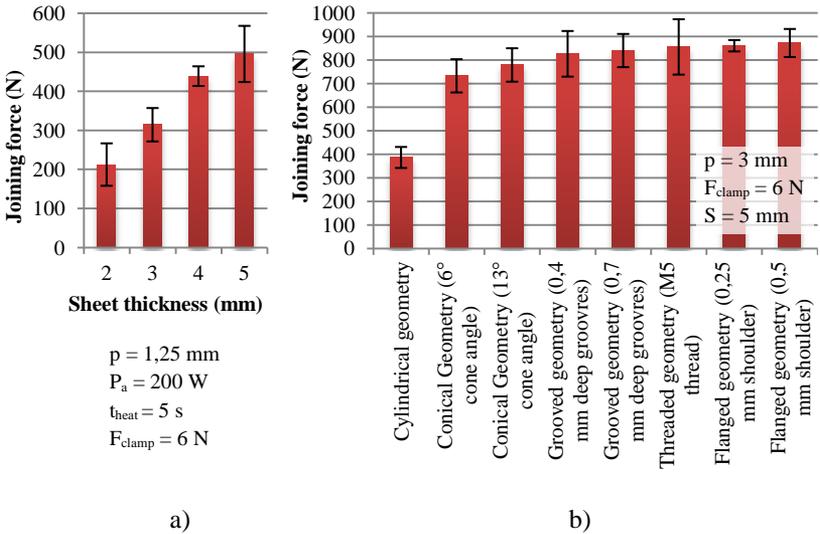


Fig. 11. Joining forces in the case of different sheet thicknesses (a) and shape locking pin geometries (b)

Finally the LAMP joining is compared with the most commonly used adhesives for joining metals and plastics. In the case of applied joining geometry, materials and adhesives the results showed that strength of LAMP joining is similar or even better than those made with adhesives. Taking into consideration the further advantages of laser, like rapidity, lack of added materials, good controllability as well, we get a technology which can be an alternative solution compared with present applied technologies in the future.

6. New scientific results - theses

1. I developed a method which is suitable for polymer transparency measurement, and which is able to determine the polymer transparency and the change of transparency as a function of time in the case of different laser beam wavelengths, power densities, radiation time ranges, laser modes, pulse settings, polymer types, thicknesses and additives under conditions of laser transparent metal-plastic joining.

During carrying out the method the power of laser beam with chosen parameters is measured by applying a thermoelectric principle power measuring unit in defocus position. After the measured power before irradiation becomes constant, the plastic sheet is got into the way of the laser beam at a defined beam diameter, in a short time, and the power change is recorded as a function of time.

If the value of power becomes constant during irradiation again, the transparency can be calculated as a quotient of constant power values before and during irradiation. In other cases, the transparency can be estimated with proportioning between constant power value before irradiation and value obtained during free cooling, which is dependent on inertia of measuring device [S1, S6, S8].

2. I defined with 3 to 10 s duration, flash lamp pumped Nd:YAG laser radiation transparency measurement that in the case of poly(methyl methacrylate) and polypropylene plastics used for joint creation
 - a. three cases can be distinguished based on irradiation time dependent plastic transparency changes [S6, S8]:
 - i. plastic transparency do not change during irradiation time
 - ii. plastic transparency is constant in the first part of irradiation time, and decreases monotonically in the second part of irradiation time
 - iii. plastic transparency decreases monotonically during the whole irradiation time
 - b. In the case of fixed laser beam parameters (pulse power, pulse time, pulse frequency) a critical average power density value, as well as in the case of fixed average power density and pulse time a critical pulse power density value can be established; below this critical value transparency is constant in time, but above this value transparency

decreases during irradiation. Further enhancement of average power density or pulse power density results in a higher-value reduction of transparency [S1, S6, S8].

Both the increase of average power density and pulse power density increases the amount of inputted heat on area unit during radiation, which accumulates under the volume of laser beam spot. The accumulated heat degrades the plastic, causing bubble formation and therefore transparency decrease. The above described 3 cases can be distinguished based on starting moment of degradation [S1, S4, S6, S8].

3. I defined through measurements, that in the case of laser transparent, penetration joining of 2 to 5 mm thick poly(methyl methacrylate) sheet and 5 mm diameter S235 steel pin, joined by flash lamp pumped Nd:YAG laser where the laser spot is concentric with the head surface of the pin and has the same diameter than the head surface, the penetration of steel pin into the plastic material increases monotonically with separate increasing of pin surface roughness (R_z 3 - 60 μm), average power (120 - 200 W), heating time (4 - 11 s), initial value of spring created clamping force between pin and sheet (3,2 - 9,2 N), because the increase of average power and heating time as well as the growing laser absorption caused by increasing surface roughness results all in a rising temperature of the pin. The rising temperature causes decreasing of polymer viscosity and therefore deeper penetration. By increasing the initial value of clamping force the pin can penetrate deeper at a given plastic viscosity [S1, S7, S9, S10, S11, S13].
4. I defined through measurements in the case of laser transparent, penetration joining of 2 to 5 mm thick poly(methyl methacrylate) sheet and 5 mm diameter S235 steel pin, joined by flash lamp pumped Nd:YAG laser where the laser spot is concentric with the head surface of the pin and has the same diameter than the head surface, in the line of the joining force (the force needed to tear the pin out of the sheet), that:
 - a. by separate increasing of heating time (4 - 11 s) and average power (120 - 200 W) joining force increases initially, because increased heating time and average power results in deeper penetration, larger contact area and at the same time more intensive bubble formation. The force increases until bubble formation do not reduces the role of head surface to an extent that it can't be compensated by increasing role of lateral surface [S7, S11, S12].

- b. The speed of bubble formation can be reduced and joining force can be improved by increasing the initial value of spring created clamping force, because increasing pressure shifts the temperature range of intensive plastic decomposition upwards [S1, S2, S7, S10, S11, S13].
 - c. Both the increase of steel pin surface roughness (R_z 3 - 60 μm) and plastic sheet thickness (2 – 5 mm) increases tearing force. Increasing surface roughness of the pin improves mechanical adhesion, which is supported by increasing penetration due to higher face surface roughness. By increasing sheet thickness stiffness of the sheet increases as well, that reduces mechanical load originating from bending deformation during tearing [S7, S9, S10, S11, S13].
5. I defined through measurements in the case of laser transparent, penetration joining of 2 to 5 mm thick poly(methyl methacrylate) sheet and 5 mm diameter S235 steel pin, joined by flash lamp pumped Nd:YAG laser where the laser spot is concentric with the face surface of the pin and has the same diameter than the head surface, that the joining force (the force needed to tear the pin out of the sheet) is dependent from laser pulse properties. In the case of constant 200 W average power, 4 s heating time, 3,2 N clamping force and 1 μm average surface roughness
- a. at a given pulse power density, a critical pulse energy can be defined by increasing the pulse time. Above this critical value penetration and therefore joining force decreases, because at high pulse energies pin temperature is lower and plastic viscosity is higher [S11].
 - b. a pulse energy value can be defined at which penetration and joining force is independent from pulse power [S11].
6. I defined through measurements in the case of laser transparent, penetration joining of 2 to 5 mm thick poly(methyl methacrylate) sheet and 5 mm diameter S235 steel pin, joined by flash lamp pumped Nd:YAG laser where the laser spot is concentric with the head surface of the pin and has the same diameter than the face surface, that the joining force can be doubled compared to cylindrical geometry by using subservient macrogeometry. First reason for improvement is the dominance of macroscopic shape locking against mechanical adhesion, which is typical for cylindrical geometry. Second reason is the change in load conditions of the plastic sheet due to shape locking, and therefore better utilization of sheet strength originating from sheet thickness increment [S12].
7. I demonstrated, that it is able to create pulse mode, Nd:YAG laser assisted, laser transparent, penetration joining of poly(methyl

methacrylate) sheet and S235 steel pin that's joining force nearly ranges with same geometry joining made with Loctite 454 and Loctite 496 adhesives, because the forming adhesion at the face and lateral surface in the case of laser joining is equal with those forming during adhesive joining. Therefore laser joining offers an alternative against adhesive joining that do not makes necessary the application of added materials [S1, S3].

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