OTKA (109436) final report

In the first research year the basic principles of laser assisted metal plastic joining were investigated.

1.1 Basic experimental conditions:
Exploring the state-of-the-art of this joining technique it can be seen that different base material combinations, different laser sources and heating methods are being developed continuously. Basically, the geometries of the joint may be sheet to sheet or pin to plate. To exploit the benefits of the laser hybrid joining we selected the transparent-absorbent type joining (penetration type) in pin to plate geometry in the first stage because of its lower complexity, which helps us separate the different main influencing factors

1.2. Selecting the laser source: The industrial laser sources could be divided roughly in 3 groups: the far infrared (CO_2), the near infrared and visible range solid state lasers (rod, disk, fiber), the diode lasers and finally the ultraviolet range. The CO_2 laser has no transmittance within any of the applied polymers, and the ultraviolet laser is only occasionally used, therefore from the SSL group the commonly used Nd:YAG laser source was chosen as the most suitable. The heating process was investigated in both of the possible operation modes ((cw) and pulse mode). Our results showed that the cw mode gives a faster joining process, but the joint strength was lower. Therefore in most of our experiment in the first year a pulse mode Nd:YAG laser was applied with Gaussian power distribution.

1.3 Selecting the base materials:
Based on these aspects, from the group of steels, an unalloyed structural steel (S235) and a high alloyed corrosion resistant steel (X5CrNi1810) were involved. From the nonferrous metals group, due to the need of high absorption and the low heat conduction (low heat loss) the most appropriate copper based (CuZn39Pb3) and alloyed aluminium based (AlMgSi0,5) alloy was selected.
Regarding the metals, higher absorption and low heat conduction was needed. From our results, it stands out clearly, that the ferrous based metals are more suitable in case of laser heating. The joints were created between 2- 7 seconds. As a comparison: with the highest absorber aluminium and copper based alloys this was not possible within 9 seconds with the same experimental setup. Comparing the unalloyed and high alloyed steel the S235 gives better results in strength. Therefore from among the metals, the structural steel was selected.

Regarding the plastic material, from the aspect of laser beam transparency and softening effect, polymers from the thermoplastic group was found to be suitable. Thus we investigated natural (PP, PMMA, PA, ABS, PC) and reinforced or modified (glass fiber, boron-nitride, talcum) polymers.
We developed a new measuring method which is suitable to determine the transparency not just in the beginning of the laser irradiation, but during the whole heating process (at the applied wavelength and power intensity of laser) as compared with the existing techniques in the market. As a part of this system development a laser power measuring device (Coherent PowerMax USB LM5000) was purchased.
Investigating the involved polymers, we found that the modification materials have a negative effect on the transparency of the plastic material. Thus it can be stated that the natural material has better transparencies. From the chosen natural polymers the PMMA has the highest transparency (92 %) as compared with other plastics. Therefore the PMMA was chosen for the basic experiment of joining. The transparencies in descending order were PMMA, PP, PA, PC, ABS.

1.4. Examining the fundamentals of the joining process:
The mechanism of the joint formation was the following: the sample was irradiated from the plastic side. The beam transmitted through the plastic and was mainly absorbed by the steel surface. The heated steel transferred the heat to the plastic which locally melted down and the steel pin penetrated into the plastic. The melted polymer flowed back along the lateral surface of the pin and formed a flash at the entrance hole. After cooling, a joint was created. It was possible to observe the shrinkage of the burr during the cooling phase by video records.
Investigating the effects of the two main influencing factors (laser power and interaction time) from the results we can summarize the following: The minimal laser power needed to create a joint was 120 W with a 5 mm spot diameter. The more the laser power, the deeper the penetration and the tearing forces until 200 W. Increasing the interaction time at the same laser power caused a higher steel pin penetration, but the maximal tearing force did not continuously increase due to the bubble formation in the joint interface which limited the connection area.
To analyse the bubble formation the laser heating process and the PMMA material were investigated. With DSC, TGA methods the characteristic temperatures were determined: the glass transition temperature was 117 °C and the decomposition temperature was 280-300 °C by a slow heating speed (10, 15, 80 °C/min).

We investigated the heating process of PMMA with different heating rates. A slow heating was carried out in a furnace up to certain, predefined temperatures and we examined the bubble formations. The results showed that above 170 °C bubble began to form in the PMMA.
In case of fast heating with laser beam there was more temperature oscillation using the shorter pulses, thus the temperature profile was very strongly influenced and controlled by laser pulse shapes.

To understand the melting phenomena of PMMA more deeply, melt flow index (MFI) measurements were carried out under special measuring conditions in order to determine that temperature range when the polymer has better flow characteristics, thus less resistance against the penetration of the steel pin. The MFI index and the viscosity results showed that the values changed more than an order of magnitude between 200 °C and 240 °C. Thus the melting of polymer is very sensitive to the small temperature differences above the glass transition temperature.

The solidification process was observed at the end of the joining process. Intensive bubble formation was detected in the flash by video camera based observations and some shrinkage effect was also identified. But this burr had no significant effect on the tearing force.

To analyse the structure of the joint the cross section of the bond (microscopy) and the torn surfaces were investigated by SEM after the tearing process.
From the cross sections it can be seen that the polymer has good wetting properties in the joint and at the flash. It filled the roughness of the pin surface (mechanical adhesion). The bubbles found were formed by the partial overheating of the pin.
The scanning electron microscope investigations show that a thin layer of plastic remained on the steel pin but steel or oxide was not detected on PMMA torn surfaces after the tearing process.

In the **second year** of the research work the characteristics of the laser beam - metal - polymer interaction, the influencing factors of the interaction and their effects were determined.

2.1. **Determination the properties of the base materials influencing the properties of the joint**
The investigated properties of base material which are important form the viewpoint of the joint (the penetration depth, bubble formations and tearing force) were already shown by our earlier results.

From the side of metal: composition, macro geometry, surface micro geometry (value and structure), coatings. Form the side of polymer: composition, additives (colouring materials), humidity of polymer, production technology (extruding, injection moulding), macro geometry (thickness), surface micro geometry (value and homogenity).

Modifying the metals
For the possible base metal materials unalloyed structural steel (S235), high alloyed corrosion resistant steel (X5CrNi1810), copper based alloy (CuZn39Pb3) and aluminium based (AlMgSi0,5) alloys were investigated.
Due to our previous result from the viewpoint of composition the unalloyed Fe based metals were the most suitable for the joint, because of the highest absorption the lower heat conduction, therefore the surface of the S235 steel was modified with the most appropriate way.
The goal of modification was to change the micro geometry and the coating on the face surface of the steel pin. The micro geometry was varied in case of turning process from Ra 1 to 10 µm and we found that the higher the average roughness the higher the penetration and the maximal tearing force because of the complex effect of higher absorption and higher shape locking. From the same roughness value the type of the manufacturing process were changed to grinding and Al oxide particle blasting. The grinded face surface reflected the laser beam therefore the penetration and the maximal tearing force was decreased. The Al oxide particle blasted surface increased the penetration with 100 % the tearing force increased just with about 10 %. The oxidized surface increased the penetration depth with about 250 % but the penetration was decreased with 20 %. Thus the ferrite oxide has not a strong connection with the metal surface, but very effectively increased the absorption.

Modifying polymers
From the thermoplastic group we investigated natural (PP, PMMA, PA, ABS, PC) and reinforced or modified (glass fiber, boron-nitride, talcum) polymers were involved. The most appropriate polymer compositions were the PMMA and the PP, The reinforcing additive materials decreased the transparency.
We can classified the laser-polymer interactions into 3 models which characterize the transparency properties of plastics. Outside the critical parameter range the transparency value is constant, but reaching a critical area the investigated plastic materials start to decompose and transparency decreases radically.
Outside the critical parameter range the PMMA has 92 %, the PP has a 65 % and the PA has a 40 % average transparency value. The PC and ABS polymers are not transparent in the investigated region. The used colouring material additives did not help the transparency in case of ABS. The investigated additives in the PP decrease the transparency. In case of 20 % glass fiber content the transparency reduced to 20 % and the BN and talcum eliminated the transmitted part of laser in the investigated region.

In case of the effect of production technologies the extruded and injection moulding processes were investigated the most suitable polymers (PMMA, PP). The result showed that the transparency decreased with injection moulding in case of both plastic. The 2 mm thick extruded PMMA sheet had 93% transparency, but the injection moulded has monotonously decreasing transparency with high deviation. In case of PP the transparency had a low value and the burning of the plastic were also started. Thus the injection moulding process does not helped the transparency, this way it is less suitable this way for the laser assisted metal plastic joining technology.
The modification of the surface of the plastic changes locally the optical properties which has important role during the heating. A high value and homogenous transparency of plastic is needed for heating the face surface of steel pin.

2.2. Declaration the factors which are important from the point of view of laser heating
The influencing factors could be divided for three different main groups: the base materials, the laser beam, the technology.
The main factors in case of base materials are the same which were discussed from the side of metal and polymer.
The main factors in case of laser beam are: the wavelength, laser power, power distribution and operation mode (pulsed or continuous wave).

It is clear from our results that the visible or near infrared wavelength are applicable. The power range is in some ten or hundred watts (in our case the average power was mostly 200 W). According to the applied Gaussian power distribution (TEM0,0) the temperature distribution of the face surface of the pin shows a maximum value in the middle of the surface, which is responsible for the more intensive bubble formation in the plastic material over the center of the joining.
The pulse peak power, the pulse time and the pulse frequency has a high effect onto the heating process. With the same average power it is possible to heat a slower and quicker the metal depending on the pulse shape. The shape of pulse also has an effect on the transparency of plastic to. We determined a critical value of power density in different ceases where the plastic is not able to tolerate the laser and the decomposition is starting during the heating process hindering the creation of the joint.

The main factors in case of technology are: the interaction time, the spot size (the power density), clamping forces (value and time dependence), concentricity (beam and the metal pin).
With constant interaction time and laser power and decreasing spot size, the power density increases quadratic way, and it reaches a value which can be tolerated by the plastic material only for a short period. Reaching the decomposition temperature it induces a radical destruction of optical properties in the material. The result is a transparency value converging to zero. Thus heating of steel pin becomes less effective. From the viewpoint of power density the plastic is the critical part.
In case of the clamping force a spring was applied in earlier. Due to the penetration type joining process the clamping force was linearly decreased with increasing penetration therefore the pressure got lower and while the temperature increased monotonously. In this part of the research work a permanent pneumatic pressure was applied with special fix and jigger equipment (cooperation with external expert). The result showed that at the same force range the penetration depth was about the same but the application of pneumatic cylinder the maximal tearing force was higher approximately 10 %. Therefore the further increasing of force is planned.
The effect of the concentricity of the laser beam and the steel pin was also investigated to determine the robustness of the system elements positioning. The result showed that the shifting of the centre

line of laser and steel pin form 0 to 1.5 mm caused more than 60 % decrease of penetration, but the maximal tearing force was decreased only by 20 %. The shifting usually is in the range of some tenth of mm, but it is important to reduce the misalignment.

2.3. Mechanical investigation to qualify and optimize the strength

We investigated the static maximal force of the joint because of the undefined contact surface.
We determined: that the tearing force starts to increase with the longer heating time but when it reaches the maximum (optimum), the tendency changes, and the force begins to decrease. In the formation of tearing force, the top and the lateral surface of the steel pin play an important role. The deeper penetration should cause a higher strength, but the bubble formation reduces the connection at the interconnection surfaces and the plastic material itself also becomes less enduring. The two contrary effects should cause the reduced strength in the range of higher penetration depths.
During our research we reached the 800 N tearing force in case of 5 mm thick PMMA plastic and 5 mm in diameter steel pint which could be alternative to adhesion bonding.

In the **third research year** the limitations and the possibilities was researched comparing the developed process.

3.1. Determining the possibilities and limits of extending the joining formation

The limitations of existing LAMP process

Time of the process: In the investigated LAMP (Laser Assisted Metal Plastic) joining process the pin to plate geometry was in the focus manly when the metal surface was heated from the plastic part. Therefore the plastic had to be show some transparency in order to create the bond. The result showed that in case of the transparent type plastics above a certain laser power density the plastic parts were damaged. The pin to plate geometry ensure connection just in spots. The number of applicable spots determined and limited the strength of the joint and if the sealing is necessary on that case it is not satisfying.

Pre-treatment of surface: It was proved with our results that the surface preparation has an high impact on the strength of the joint. If the no prepared surface were used (rolled, turning) the strength of the joint was limited. The usual chemical cleaning for example using aceton can help the process but in a limited way.

Base materials and material properties:

Investigating the base materials in case of metal part the structural steel, stainless steel, aluminium, cupper material were examined. Because of the reflection properties of metal the application of aluminium and cupper material were limited.
In case of plastic parts more plastic types were investigated but just some of them were applicable for the LAMP joint because of the low laser transparency. The strengthening additives which is useful for other viewpoint are harmful in case of the value of transparency. Therefore the application of thermoplastic in a wide range is limited by the low transparency effect if the joining process occurred form the plastic side.

Laser properties

Because of the applicable thermoplastic materials the usable wavelengths and therefore the laser source types are limited. Just the laser beams from the visible range and the near infrared region (for example solid state lasers: SSL) can be applied. The commonly used CO_2 laser is not possible to use because of the high absorption and lack of transparency in plastic parts. Thus the applicable laser sources are limited.

We determined that in case of solid state laser that not just the average power is important but the pulse peak power too. It will determine the interaction type between the plastic and the laser. If the peak pulse power is too high the plastic will damage therefore it hinder the joint creation. So the pulse mode has also a limitation in this case.

Investigated extension possibilities of LAMP process

Time of the process: to expand the joint geometry into the direction of higher strength and solid joint the **sheet to sheet geometries** were investigated. In the first case plastic side joining were applied. The joints were created successfully. The sealing were solved but the process speed were also limited by the transparency limit of different plastic parts.

In order to increase the productivity more we investigated the metal side heating as an extension in case of sheet to sheet sample construction. In case of **metal side heating** the higher power density were able to use. During this heating situation the heat conduction of the metal part will just limit the time for the joining which is higher than it was earlier.

Due to the metal side heating construction higher laser power was able to use, because the metal parts is able to endure higher power density in this power range. Thus the process become faster than it was earlier.

Pre-treatment of surface: To enhance the strength other surface preparation was used. The surface modification of the metal parts was investigated and the result showed that it has a high influence on the strength of the joint. The surface micro and macro geometries were modified in order to get a better connection between the metal and the plastic parts in case of pin to plate and sheet to sheet geometries. We declared more preparation which is able to enhance the strength (grain blasting, conical and grooved shaped pins).

Base materials and material properties:

The most important property of plastic part in case of LAMP joining is the value of the transparency. Due to the metal side heating this property was not so important therefore wider range of thermoplastics are applicable. Thus the usable material type were expanded comparing with the earlier state when the PMMA plastic was applied.

Laser properties

Changing to metal side heating other wavelength laser sources become applicable. Not just the visible and near infrared range (SLS, diode) laser sources were able to use, but it was possible to extend the applicable laser sources to the very often used CO_2 laser too from far infrared range (FIR) and (VIR) like diode laser.

We extended the applicable laser power in to the higher ranges form some hundred watts to kilowatt ranges.

In case of metal side heating the pulse mode can be used without the earlier limitation due to the fact that the metal parts can tolerate the high peak pulse power or densities.

3.2. Durability test of the joint

The developed LAMP joint are created for construction in the different industrial segment like automotive industry. Therefore the static strength of the joint is not enough information about the bond. Therefore we expanded the test of the joints with: long term static tearing test, fast tearing test, vibration test, tearing test with different temperatures and heat shock tests.

From the result of long term (7 month) static tearing can be see that the tearing force basically increasing between 10-30 % because of ageing of plastic material.
Increasing the tearing speed the macro geometry of the steel pin has a high influence on the dynamic tearing force. In case of cylindrical pin the tearing force is decreasing but in case of flanged pint the it is increasing by enhanced tearing speed.
The dynamical tests showed that about the half load of the static tearing force there are no damage below 15 000 - 20 000 cycle. Using higher load the cycle decrease suddenly to some thousand and hundred cycles.
The thermal behaviour of the sample were tested during tearing test with different temperature (-40…+ 80 ˚C). The changing in the maximal tearing force less than 20-30 %.
From the heat shock test (cycle: 10 min on -30 ˚C than 10 min on 80 ˚C) it can be seen that after some cycle 10-15 the strength is decreasing about 30 %, because the plastic part is sensitive to the heat differences.
The result show that it is not enough to know the static strength of the joint. In different situations the strength very strongly depending on the circumstances. For further application the expectable circumstances have to be tested in every cases.

3.3. Determining the complex criteria system

In case of the criteria system we have to divide the process into two type.
In the first part the transparent type LAMP joining to reach the right temperature in the interaction zone the plastic part have to be a transparency in the range 60-100 % in order to let enough laser energy to surface of the metal part. The surface of the metal part have to be high absorption on the surface to heat up the interaction surface. The metal surface or the preparation of the surface have to be let the laser beam increase the temperature of metal to the softening of plastic but not overheating. Not just the temperature but the temperature over time have to be fitted to the selected plastic material. Other criteria is the clamping force which have to be applied during the process. The clamping force have to be high enough to ensure the heat transfer between the two materials during the whole process. Not just the heating but the cooling too.

In the second type LAMP joining when the metal side is heated the plastic part has less criteria. The transparency is not a viewpoint. The absorption of the metal part is more important. In this case the heat conduction is have to be high enough to conduct the necessary temperature to the interface surface. The heat conductivity and the thickness of the metal part become an important *role. The speed of the process have to be high enough not to deform the metal part but slow enough to reach the necessary temperature on the other side to create the bond to the plastic.*
The exact values of this criteria can be seen in our research papers in the investigated situations.