

János Takács - Péter Gál, MSc - Antal Lovas - Joachim Belt \*

## FLUX POKOVOVANIE ZOSILNENÉ ABSORPCIOU AI POVRCHOV PRI CHARAKTERISTICKEJ VLNOVEJ DĹŽKE CO<sub>2</sub> LASEROVÉHO ŽIARENIA

### FLUX COATING ENHANCED ABSORPTION OF AI SURFACES AT THE CHARACTERISTIC WAVELENGTH OF CO<sub>2</sub> LASER IRRADIATION

*Hliníkové tvrdé spájkovanie je metóda pre spojenie Al-častí v tesnom dotyku pomocou kvapalnej zliatiny, ktorá sa tavi nad 577 °C. Na základe použitého tepelného zdroja, je možno rozlíšiť konvenčnú metódu (plameň) alebo metódu energetického zväzku lúčov (laser). Laserová metóda umožňuje rýchle lokálne tavenie v okolí povrchov, ktoré majú byť spájané. Napriek vysokej hustote energie (reprezentovanej laserovým žiarením) taveninou Al je tavenie často neúspešné. Príčinou je nízka absorpčná schopnosť Al v blízkosti vlnovej dĺžky charakteristickej pre CO<sub>2</sub> laser. Z dôvodu zvýšenia absorpcie energie sa preto musia použiť povlaky, ktoré majú vysokú absorpčnú schopnosť.*

*V práci je diskutovaná zmena absorpčnej schopnosti Al povrchov spôsobená povlakmi vytvorenými NOCOLOK<sup>®</sup> Flux technológiou.*

*Aluminium brazing is a method for joining Al-parts in close proximity by introducing liquid alloys which melt above 577 °C. On the basis of an applied heat source, conventional (flame) and energy-beam brazing (laser) can be distinguished. The laser is capable to result in rapid local melting around the surface to be joined.*

*In spite of the high energy density (represented by laser irradiation) the melting of Al is often unsuccessful. The reason is the low absorption ability of Al at around the wavelength characteristic to the CO<sub>2</sub> laser. Coatings with a high absorption ability have to be used in order to facilitate the energy absorption.*

*In the present study the change in absorption ability of Al surfaces due to NOCOLOK<sup>®</sup> Flux coatings will be presented.*

#### Introduction

In principle, all kind of laser can be used for brazing as a heat source. However, their efficiency in the brazing process is very different, and depends highly on the absorption ability of the irradiated material around the characteristic wavelength of the applied laser. For this reason CO<sub>2</sub> laser can not be used for brazing of Al and its alloys [1, 2].

The explanation of this fact can be drawn from Fig. 1. where the room temperature absorption ability as a function of wavelength is plotted for several metals.

While the absorption ability for steels at 10.6 μm is appreciable, (~ 0.1) this value is very low for Al (lower than 0.02).

The absorption ability can be risen by applying appropriate coatings. Graphite is commonly used in laser technology to rise the absorption ability of sample surfaces. Due to the corrosion risk attributed to graphite, its application is undesirable in the case of Al joining. It is plausible that absorption ability can be enhanced by addition of good absorber material to the flux. This condition seems to be fulfilled automatically by applying the

NOCOLOK<sup>®</sup> Sil Flux, as a consequence of Si particles being dispersed in the NOCOLOK<sup>®</sup> Flux.

To verify this supposition the change of absorption ability at 10.6 μm was investigated on different Al surfaces covered by this type of flux. For the successful application of CO<sub>2</sub> laser irradiation to the Al brazing, the mechanism of NOCOLOK<sup>®</sup> procedure will be briefly described [3].

#### The NOCOLOK<sup>®</sup> aluminium brazing process and the NOCOLOK<sup>®</sup> Flux

The flux dissolves the aluminium oxide before the melting of filler material has started and protects the eutectic join against the oxidation during the whole process. The eutectics composition of filler material ensures lower melting point than that for the parts to be joined. (See Al-Si phase diagram in Fig. 2.) The Flux material (Trade mark: NOCOLOK<sup>®</sup> Flux) is also eutectic system with the composition of KAlF<sub>4</sub> + K<sub>2</sub>AlF<sub>5</sub>·H<sub>2</sub>O with the melting temperature of 562 °C - 577 °C, which is slightly below the melting

\* <sup>1</sup>Prof. Dr. János Takács, <sup>1</sup>Péter Gál, MSc., <sup>1</sup>Ass. prof. Antal Lovas, <sup>2</sup>Dr. Ing. Joachim Belt

<sup>1</sup>Budapest University of Technology and Economics, Dept. of Mechanical Engineering Technology Bertalan L. Street 2., 1111. Budapest, Hungary, Tel.: +36-1-463 1694

<sup>2</sup>SOLVAY Fluor und Derivate GmbH, Hans-Bickler-Allee 20, 30173. Hannover, Germany, Tel.: 49-511-8570

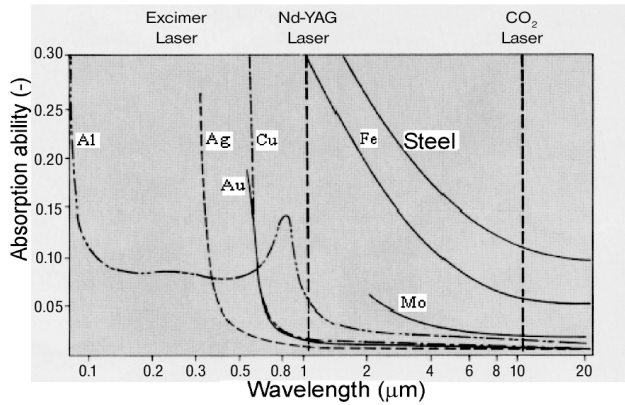
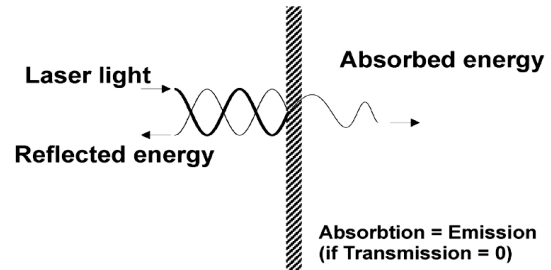


Fig. 1 Absorption ability of several metals at room temperature [6]



point of eutectic Al-Si [4]. The eutectic filler liquid with sufficiently low melting point can also be formed via local dissolution of Si-particles in the neighbouring Al parts. The Si particles are previously mixed into the Flux material. (Trade mark: NOCOLOK Sil. Flux) The oxide dissolving ability is the same for both type of Flux material.

The brazing process is schematically illustrated in Fig. 3. No change can be observed below 562 °C when the material is heated. (Fig. 3a) In the temperature range of 562 °C - 577 °C the Flux has melted and the aluminium oxide layer is dissolved. (see Fig. 3b) When the temperature rises beyond 577 °C, the filler material (Al/Si12wt % alloy) is also melted. In case of using NOCOLOK<sup>®</sup> Sil Flux the silicon particles dissolve in the Al forming an eutectic liquid near the surface (Fig. 3c). The mechanical joint between the parts is formed via the freezing of this liquid layer. After the completion of brazing process the surface is covered by the remainder part of Flux (no need to remove, see Fig. 3d).

$$a_{\lambda,T} = \varepsilon_{\lambda,T}$$

where  $a_{\lambda,T}$  is equal to the absorption coefficient and  $\varepsilon_{\lambda,T}$  is equal to the relative emission coefficient of a grey body at a given temperature and wavelength

According to this one can suppose that the determination of temperature dependence of the relative emission coefficient on the wavelength of CO<sub>2</sub> laser (10.6 μm) can supply useful information also about the absorption ability of the same partner materials. So, the intensity of infrared emission spectra was measured using filter on the wavelength of CO<sub>2</sub> lasers (10.6 μm) during continuous heating of a series of samples, covered by the same Flux composition used for the laser brazing. In the next chapter the measuring techniques, and the temperature dependence of the relative emission coefficient on the applied materials will be summarised.

### Measurement of the absorption ability

The absorption ability cannot be measured directly. Therefore the emission coefficient was measured for the individual partner materials. According to the Kirchoff law (which is the basis of our measurements) [7]:

### Experimental settlement and procedure

The essence of measurements is the continuous monitoring of the surface temperature of a sheet radiator on which the samples are placed. The spectral distribution of the emissivity is compositional dependent. Besides it may also depend on the tribological character of the surface at the observed wavelength. So, the local

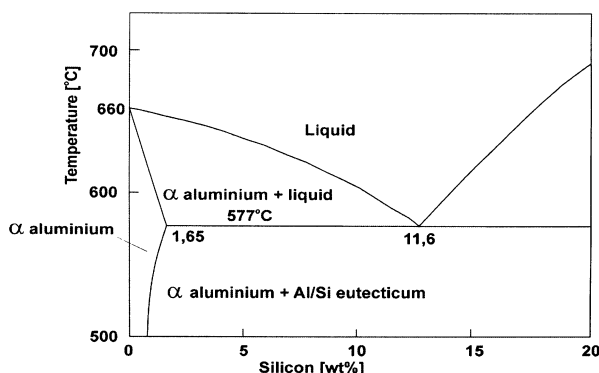


Fig. 2 Al-Si phase diagram [4]

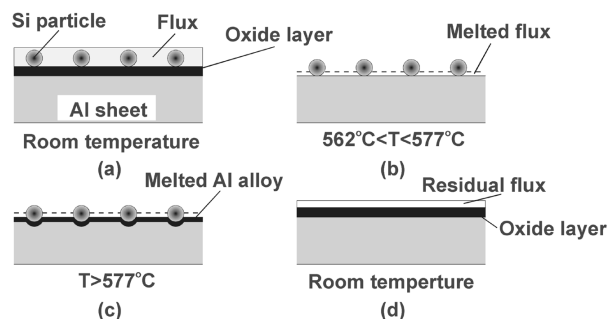


Fig. 3 The NOCOLOK<sup>®</sup> aluminium brazing process [3]

temperature on the surface of a given sample measured by thermovision can significantly differ from the physical temperature measured by a thermocouple at the same point of the surface [10]. Therefore, the first task was to ensure a nearly constant temperature on the surface of the sheet radiator. In order to improve the temperature homogeneity of the heated sheet surface, it was covered with a 5 mm thick Cu plate. In addition, Ni-Cr powder was layered under and on the surface of the Cu plate. The samples were pressed into the Ni-Cr powder in order to ensure perfect heat contact between the samples and the surface-radiator. Ni-CrNi thermocouples are used for the measurement of the physical temperature at several points of the surface embedding them into the powder layer. The experimental arrangement of the system is illustrated in Fig. 4.

The temperature was continuously increased during the experiments from the room temperature up to the melting point of aluminium, then the radiator was cooled down, switching the controllable power supply off. During the whole process the thermal emission was continuously monitored using a THERMOVISION® 880 LWB type infrared camera.

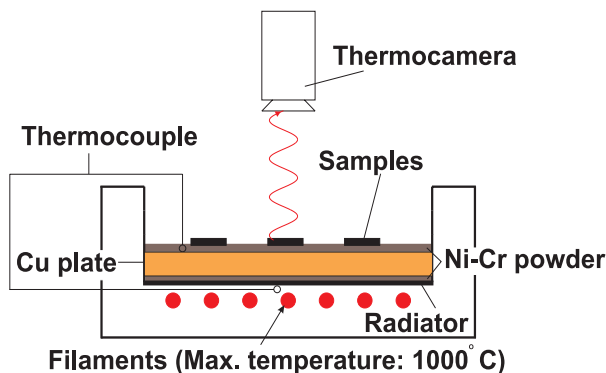


Fig. 4 Experimental arrangement for the thermal emission measurements

### Sample preparation

Sample geometry are 20 x 20 mm squares or Ø10 mm circles, being cut or punched either from a simple 1 mm thick Al-plate (without laminated layer) or from a laminated Al-plate.

After mechanical flattening, burring-off, and degreasing in acetone, the samples were clad, using NOCOLOK Flux (water-based dispersion of Flux powder) and NOCOLOK Sil-Flux (Flux, containing Si powder) with three different concentrations in each case (Flux: 5 %, 10 %, 15 %, Sil Flux: 10 %, 15 %, 20 %). From the sets of Flux 15 % and Sil Flux 20 % several samples were dried in a furnace (150 °C, 2 hours heat treatment in a box-furnace) in order to remove the chemically bonded crystal water. Another fraction of samples was dried at the room temperature. The specimens were pressed into the NiCr powder layered on the surface. In this way a perfect heat-contact between the samples and the heated surface is ensured.

### Experimental results

The increase in emissivity was often found in the low-temperature range of our heating experiments (Fig. 5 - 10). In principle, the change of emissivity is not expectable if a phase transformation or a compositional change is absent. In fact, the change in  $\epsilon(T)$  can be resulted from the sensitivity limitation in the low temperature range. In spite of the possible sensitivity limitation- there is a definite compositional dependence of the observed slope of  $\epsilon(T)$  in this temperature range, i. e. the emissivity is lower for lower concentrations. So the upraise can be detected, when the Flux concentration is low (between 5 - 15 %). It was found that slope of  $\epsilon(T)$  increases with decreasing concentration of Flux in the covered layer. For the laser brazing the higher concentration of Flux will be good because of its higher absorption coefficient. Therefore, it was highly suspected that the low-temperature increase of  $\epsilon(T)$  is connected with a compositional change (water evaporation) during the experiments. This hypothesis was verified by continuous heating-cooling experiments where several samples (including 5, 10, 15 and 20 % Flux content, with and without Si particles) were continuously heated from the room temperature to 700 °C, and then cooled below 100 °C again with the reference samples, monitoring continuously the actual temperatures as was previously described.

The main feature of the heating-cooling curves can be characterised by subtracting the measured  $\epsilon$  values obtained during the heating and cooling experiments. The difference increases with decreasing Flux content, as it is obvious from Figs. 5 - 10. It is clear that  $\epsilon(T)$  increases at the heating side, especially at around 10 - 15 % of Flux content. The measured  $\epsilon(T)$  values seems to be higher for the Si-Flux samples.

Another interesting feature of the curves is that  $\epsilon(T)$  value at the cooling side is nearly independent from the initial Flux concentration (about 0.42 for all cases), which hints again to the possible compositional change being nearly completed during the heating process. *The main conclusion is that temperature dependence of the relative emission coefficient at around 20 % Flux content is nearly identical both on the heating and cooling side, consequently the effect arising from the possible compositional change is negligible.* The quantity of chemically bonded water may contribute to the observed differences in the lower temperature range. At higher temperature this difference is negligible.

The melting process of the Flux material takes places at around 570 °C dissolving the thin  $Al_2O_3$  layer on the surface of Al. Rising the temperature eutectic Al-Si melt is formed via the diffusion of Si into the Al surface. Comparing the heating and cooling side of the  $\epsilon(T)$  curves a definite difference can be seen in the case of samples covered either by Flux or Sil Flux.

The difference arises from the phase transformation (melting) and from the chemical reactions (alloy formation,  $Al_2O_3$  dissolution) started at high temperatures.

The cooling side of the  $\epsilon(T)$  curves are practically unaffected by the Si content of the Flux. On the contrary Si addition causes

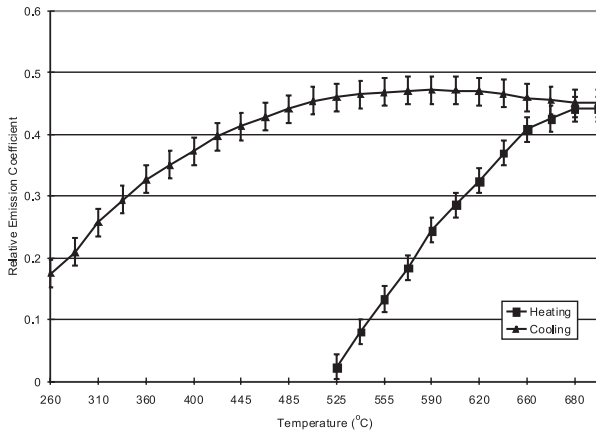


Fig. 5 The temperature dependence of Relative Emission Coefficient (5 % Flux, dried on air)

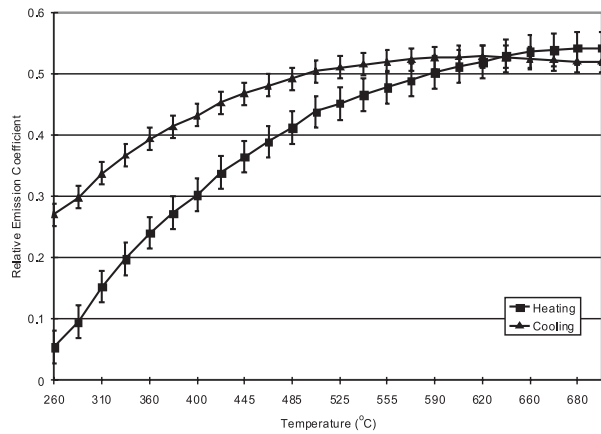


Fig. 6 The temperature dependence of Relative Emission Coefficient (15 % Flux, dried on air)

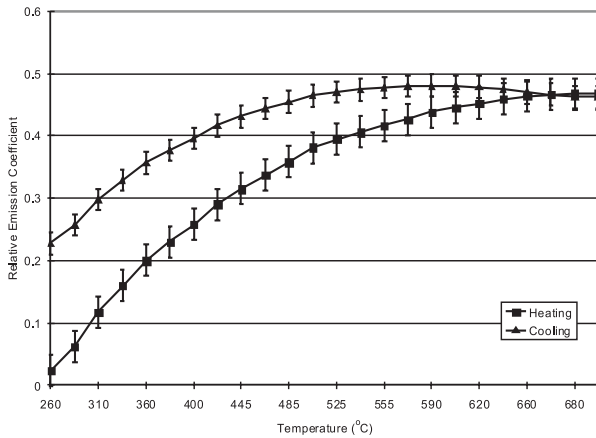


Fig. 7 The temperature dependence of Relative Emission Coefficient for 10 % Sil Flux, cladding, dried in air

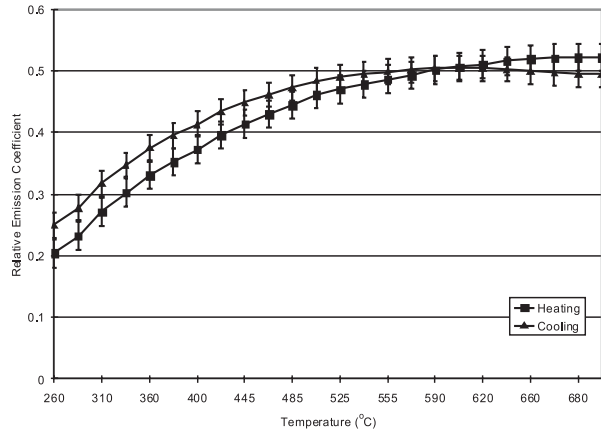


Fig. 8 The temperature dependence of Relative Emission Coefficient for 20 % Sil Flux, dried on air

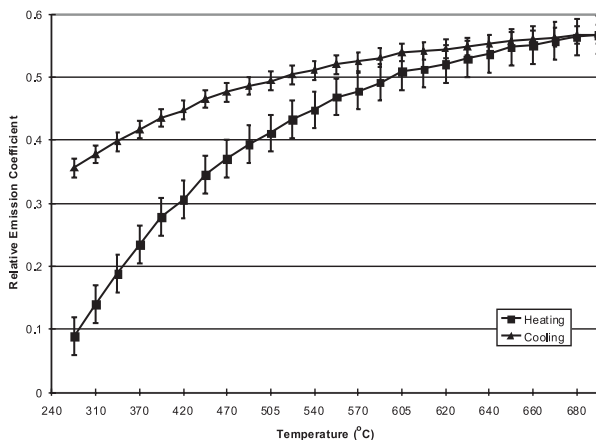


Fig. 9 The temperature dependence of Relative Emission Coefficient for 15 % Flux, dried in furnace

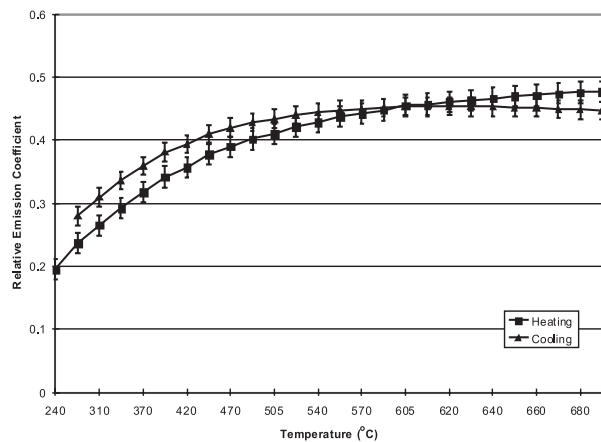


Fig. 10 The temperature dependence of Relative Emission Coefficient for 20 % Sil Flux, dried in furnace

a significant change in  $\varepsilon(T)$  on the heating side. Flux with Si addition has a higher emissivity than one without Si powder.

### Control experiments with CO<sub>2</sub> laser

Applying the results of thermovision measurements, pure and Flux-covered aluminium sheets were irradiated by laser beam. Except the exposure time, all of the applied irradiation parameters were the same during the experiments. Experimental arrangement can be seen in Fig. 11.

additional agent is not needed for the successful laser-brazing of aluminium parts.

### Conclusions

- The CO<sub>2</sub> laser energy absorption in Al surfaces can be significantly enhanced by applying surface coatings with NOCOLOK Flux or NOCOLOK Sil Flux prior to the laser irradiation.
- The emissivity (so the absorption ability) is improved by increasing Flux concentration in the coating.

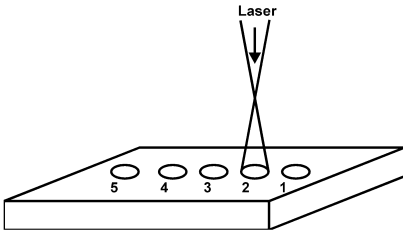
<p>Al without 20 % Sil Flux P: 600 W v : 0 Focal diameter: 2 mm Shielding gas: argon Exposure time: 1: 0.2 s 2: 0.4 s 3: 0.6 s 4: 0.8 s 5: 1 s</p>		<p>Al with 20 % Sil Flux P: 220 W v : 0 Focal diameter: 2 mm Shielding gas: argon Exposure time: 1: 0.2 s 2: 0.4 s 3: 0.6 s 4: 0.8 s 5: 1 s</p>
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Fig.11. Experimental arrangement and irradiation parameters for laser brazing

In spite of the highest power density applied, melting of pure aluminium sheet cannot be detected due to the low absorption ability of pure aluminium. On the contrary, the melting has definitely started on the flux-covered sheets using the same power density.

Therefore it can be established that absorption of CO<sub>2</sub> laser energy in aluminium is highly enhanced if surface cladding by NOCOLOK<sup>®</sup> Flux or by Sil Flux is applied. Consequently, further

- The increasing absorption ability (caused by the Flux covering) is also supported by the CO<sub>2</sub> laser irradiation experiments.

### Acknowledgement

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### Literatúra - References

- [1] PRIMKE, K.: *Az alumínium hegesztése és forrasztása* Műszaki Könyvkiadó, Budapest, 1975. p.: 314
- [2] HANEBUTH, H.: *Laserstrahlhartlöten mit Zweistrahltechnik* Meisenbach Verlag Bamberg, Erlangen, 1996. p.: 157
- [3] TIMSIT, R. S., JANEWAY, B. J.: *A novel brazing technique for aluminium* Welding Journal, June 1994. pp.: 119s-128s
- [4] *The NOCOLOK<sup>®</sup> Flux brazing process* Reklám anyag SOLVAY Fluor und Derivate GmbH.
- [5] VERÁ, J., KÁLDOR, M.: *Fémtan Nemzeti Tankönyvkiadó*, Budapest, 1996. p.: 636
- [6] DAWES, C.: *Laser welding* Abington Cambridge, 1992. p.: 258
- [7] RUDOWSKI, G.: *Az infratelevízió és alkalmazásai* Műszaki Könyvkiadó, Budapest, 1982. p.: 226
- [8] GREENSTEIN, M.: *Optical absorption aspects of laser soldering for high density interconnects* *Applied Optics*, Vol. 28, No. 21, pp.: 4595-4603
- [9] WITHERELL, C. E., RAMOS, T. J.: *Laser Brazing* Welding Journal, October 1980. pp.: 267s-277s
- [10] KISS, Gy., SKLÁNITZ, A., SZILÁGYI, A., TAKÁCS, J.: *Surface Treatment and Modification by Laser Beam* *Bewertung, Behandlung und anwendung von Konstruktionswerkstoffen in Fahrzeugbauteilen VII. Kolloquium*, Dresden, 1990. pp.: 1-15.