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Advances in continuous carbon fibre reinforced Aluminium matrix composites

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Abstract
Space structures require the usage of thermally stable materials with high -specific stiffness and strength. These must operate in harsh space environment with limited degradation to fulfil space requirements and be able to save mass, space and costs.
LKR possesses a gas metal infiltration process capable of manufacturing carbon-fibre reinforce Aluminium matrix composites (AluC MMC) with excellent properties but high standard deviations due to an outdated heating/cooling technology and process control.
This paper presents the development work conducted in research project SpAACe where the update of the manufacturing process led to significantly higher mechanical properties and reduced scatter.

1. Introduction

All materials used in space applications must fulfil a number of sometimes conflicting requirements such as mass, stability, strength, stiffness, and radiation resistance. Although materials used in space are not different from those for terrestrial application, the operational environment with its high energetic particles and ionizing radiation from sun, vacuum, large thermal variations or high velocity debris and meteoroids differs greatly. In special, space structures require the usage of thermally stable materials with high density-specific stiffness and high density-specific strength. These must operate in harsh space environment with limited degradation in order to fulfil space requirements and at the same time be able to save mass, space and costs.
Metal Matrix Composite (MMC) materials especially long fibre reinforced MMC´s offer such unique combination of material properties. A lot of MMC materials were investigated by the scientific community worldwide, but limited commercial products are on the market, especially for long fibre reinforced parts and especially from European suppliers (TiSiC from UK for SiC-long-fibre reinforced Titanium matrix MMC´s) to date. The reason for this is the difficulty of stable production leading to high production costs, the difficulty to achieve combined high specific stiffness and strength and the currently small market for such materials.

In 2013 to 2014 Aerospace & Advanced Composites GmbH (AAC) was contracted in an ESA (ESTEC) led Basic Technology Research Programme (TRP) to conduct a thorough material investigation series on latest European C-fibre reinforced MMC materials [1]. Novel materials with i.e. a maximum of density specific stiffness, density-specific ultimate strength, or ±0 coefficient of thermal expansion (CTE) over a wide temperature range (-150°C to +300°C) are searched for by ESA for current and future European Mars Robotic Exploration Preparation (MREP) Programmes. Mars surface missions such as ExoMars, Mars Sample Return or similar missions, tight mass constraints exist, particularly for mall landers or rovers.
In this TRP project of AAC a total of 154 MMC candidate materials have been reviewed and traded by application of a set of selection criteria which were defined and agreed with ESA. For the ranking of the different materials 4 Ashby criteria [2]: a: natural frequency of vibrating tubes, B: deflection of tubes under load, C: stress of beam under bending and D: strength of strut under pure tension as well as the specific stiffness. Besides these criteria, procurement issues (ITAR), maturity of production cost and production limitations (i.e. size limitation, machining and coating ability) were considered.

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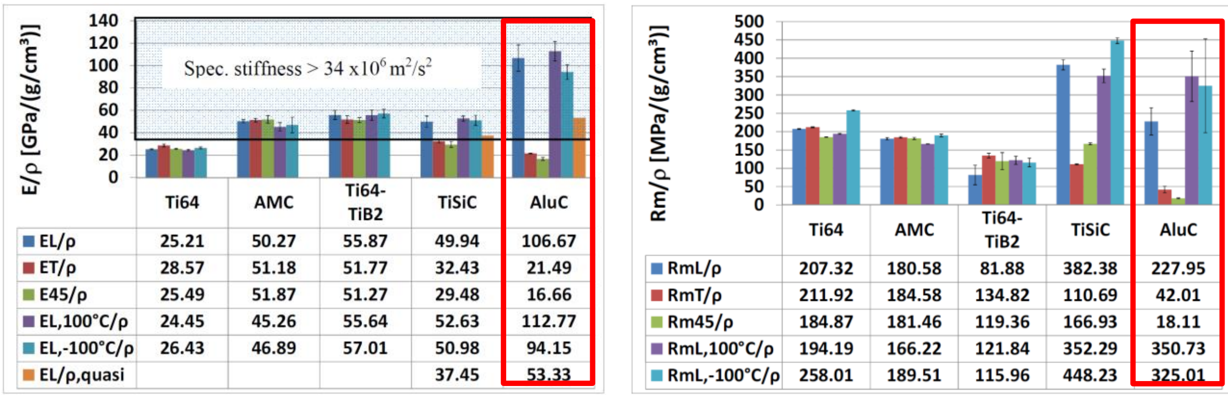
In this search for novel high specific stiffness materials, a total of 4 candidate materials have been selected for detailed investigations together with Ti6Al4V (ELI grade 23) as a reference material for comparison. The 4 final candidate materials were:

Al-MMC$_p$ – AMC 640xa	(AA6061 Aluminium alloy + 40 vol% SiC particle reinforcement)
Al-MMC$_f$ – LKR AluC	(Al 99,85 pure Aluminium + 60 vol% C/M40-fibre reinforcement)
Ti-MMC$_p$ –	(Ti6Al4V Titanium alloy + 30-35 vol% TiB2 particle reinforcement)
TiMMCf – TiSiC	(Ti3Al2.5V Titanium alloy + 33-35 vol% SiC fibre reinforcement)

The four candidate materials as well as the reference underwent a mechanical test campaign at earth conditions. Results (density-specific stiffness, density specific strength as well as CTE) of this test campaign are shown in the following figures 1 and figure 2.



	Ti64	AMC	Ti64-TiB2	TiSiC	AluC
EL/ρ	25.21	50.27	55.87	49.94	106.67
ET/ρ	28.57	51.18	51.77	32.43	21.49
E45/ρ	25.49	51.87	51.27	29.48	16.66
EL,100°C/ρ	24.45	45.26	55.64	52.63	112.77
EL,-100°C/ρ	26.43	46.89	57.01	50.98	94.15
EL/ρ,quasi				37.45	53.33

a) Density specific stiffness

	Ti64	AMC	Ti64-TiB2	TiSiC	AluC
RmL/ρ	207.32	180.58	81.88	382.38	227.95
RmT/ρ	211.92	184.58	134.82	110.69	42.01
Rm45/ρ	184.87	181.46	119.36	166.93	18.11
RmL,100°C/ρ	194.19	166.22	121.84	352.29	350.73
RmL,-100°C/ρ	258.01	189.51	115.96	448.23	325.01

b) Density specific ultimate strength

Figure 1: Comparison of (a) density specific stiffness and (b) density specific strength of the assessed high-performance materials; LKR material AluC in red Box; [1]

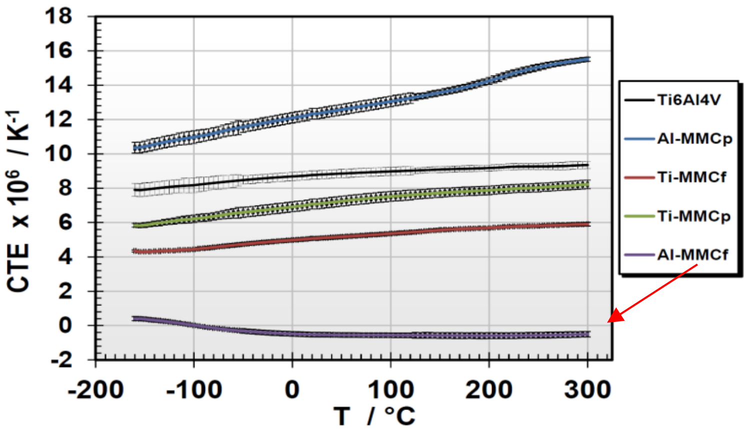


Figure 2: CTE profiles in the temperature working range between -150°C and +300°C [1]

Although showing excellent mechanical properties for specific stiffness (E_L=106,67 GPA/(g/cm³)), specific strength (227,95 MPA/(g/cm³)) and a CTE of ~0 x 10^6/K^{-1} for both low as well as elevated working temperatures, results of AluC material of LKR showed high standard deviations in mechanical properties due to the lack of process robustness as well as poor results in stress crack corrosion tests. In addition, mechanical results of AluC results were low when being compared to theoretical possible material properties (Figure 3). For these reasons, it was down-selected and not considered for the final test campaign at Martian environment conditions in this project.

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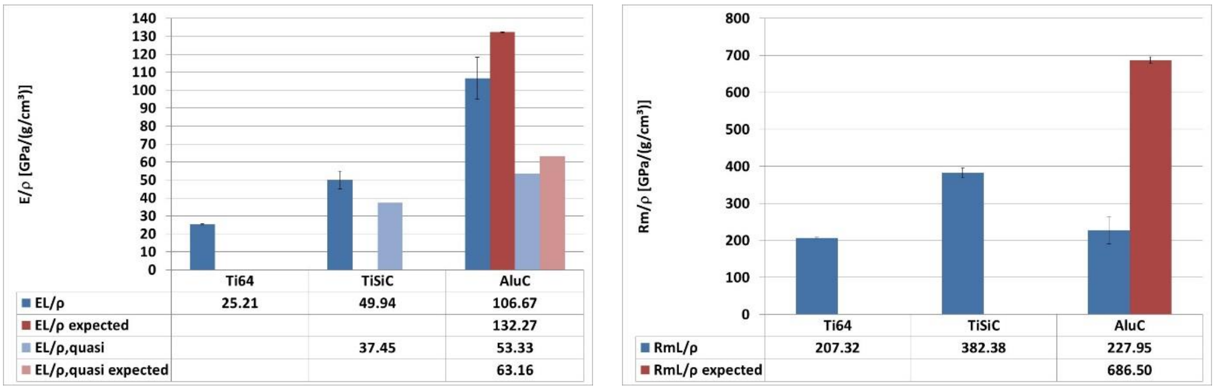

(a) Specific stiffness (b) Specific strength

Figure 3: Measured (a) specific stiffness and (b) strength of Ti64, TiSiC and LKR Al-MMC (blue) compared to theoretical max. values (red) in uni-directional (L) and quasi-isotropic layup (transparent).

LKR´s AluC MMC with continuous C/M40 fibre reinforcements has the following advantages:

- Single filament diameters are less than 10 µm (TiSiC 100-150 µm)
- Compaction density up to 65% volume fraction are possible (compared to 35% in TISIC)
- Dominant positive effect of fibres on maximum stiffness, strength values, CTE
- Low density of Aluminium matrix compared to Titanium matrix
- Near-0 Coefficient / slightly negative Coefficient of Thermal Expansion

Although proving competitiveness, the process robustness of the applied gas-infiltration process for AluC production built one of the weak points which required significant improvement so to gain process robustness and raise mechanical properties towards theoretical maximum values.

As a result of this ESA TRP project, AAC and LKR set up a research project "Space Applications of Aluminium Composite" (SpAACe, FFG No.859755) with the following three major goals:

1. Robust and advanced gas-pressure-infiltration process delivering high performance Al-MMC incl. pre-processing (winding / preforming / compaction) and tempering
2. Increasing density-specific material properties to achieve worldwide competitive, high performance C-fibre reinforced, lightweight, SCC-resistant MMCs
   (Key performance indicators: +15-25% higher specific stiffness; >+50% higher specific strength (baseline = 227 MPa/(g/cm³) at present); enhanced corrosion resistivity through selected matrix alloy,coating,anodization
3. Material data base for lightweight-MMC (AluC) assessed according to space standards

This paper presents intermediate research results of SpAACe. These include results on significant improvements of mechanical properties through an advanced gas metal infiltration process, resulting process stability and conduction of a systematic fibre/matrix/process investigation.

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2. Gas Metal Infiltration Process and Advancement

Extensive research has been carried out to find suitable processes to produce a range of MMC with different characteristics in terms of matrix's alloys, fibres nature and state of both phases during the formation of the material. Those processes could be categorized in the three big groups, depending on the state of the matrix when the composite is processed. Thus, it can be differentiated between the three main process routes for manufacturing metal matrix composites:

- Solid State processing through powder-metallurgical processes with addition of short fibres or particles (Direct hot pressing (DHP), Rapid Sinter Pressing (RSP), Hot Isostatic Pressing (HIP), Spark Plasma / Field Assisted Sintering, Powder spraying / rolling / forging / extrusion etc.) ; High-pressure diffusion welding of stacks of long-fibre reinforced MMC-prepregs.
- Liquid State Processing such as Squeeze casting or gas-pressure-infiltration of unidirectional aligned long-fibre, short-fibre fabrics or particles made of carbon fibres, boron or silicon carbides.
- Deposition processing: such as Ion platting or Plasma spraying.

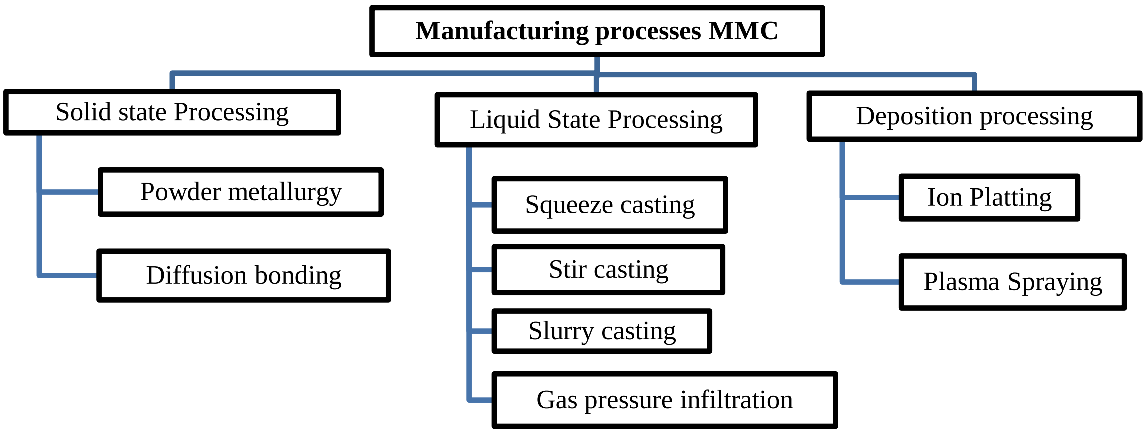

Figure 4. Most common MMC manufacturing processes

These manufacturing methods determine the microstructure and interfacial bonding conditions between reinforcements and matrix. Thus, they have great influence on the mechanical properties of the final materials [3].
In addition to these primary processing methods, additional secondary processing techniques are thermo-mechanical treatment; rolling, forging, extrusion which are applied to bring MMCs to near-net shape for final finish operations or application.

Since its on beginning in 1994 LKR has conducted research and development in the field of Aluminium and Magnesium alloys development as well as process investigations and improvement. Besides the fields of metal alloy research, also high-performance particle, short fibre and long fibre reinforced light metals have been field of R&D activities. For this, LKR has used devices such as the gas-pressure-infiltration process or squeeze casting technology since 20 years (Figure 5).

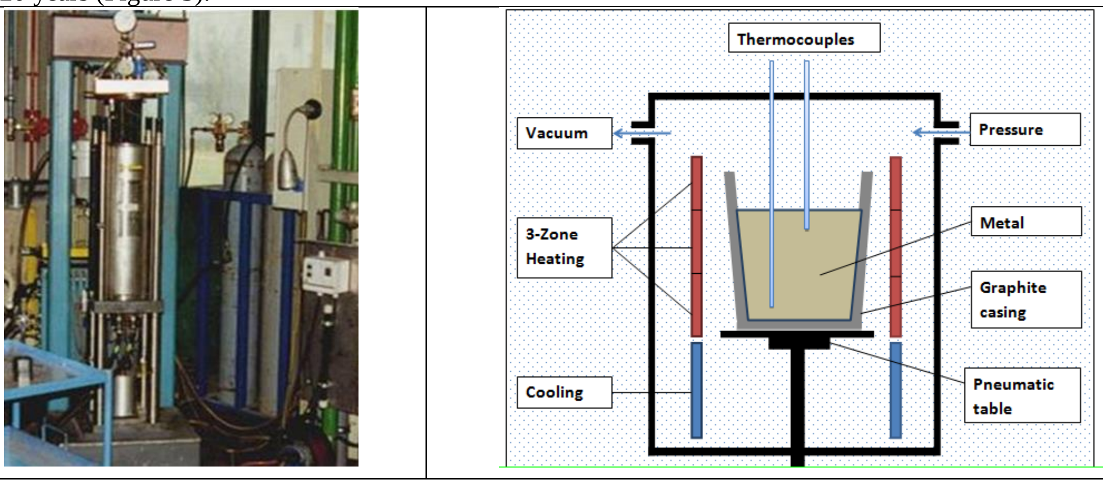

Figure 5: Gas-pressure-infiltration process facility of LKR (left) including facility schematic (right);

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The process is carried out in a pressure casting equipment, with a maximum pressure range of 100 bar. During the process, the pressure in the autoclave, the temperature of the three stages of the heating system and the temperature data detected by three thermocouples is monitored continuously with a sampling time of 1.5 s.
The composites are prepared by Gas Pressure Infiltration method and the process is done as follows. The first step is the winding and alignment of the fibres longitudinally in a processing container. This step begins with a continuous roll of fibre yarns (Toray M40J) containing 6000K fibres of 5 micrometres. The container is closed, and the fibres compressed so to get a sound compaction for an envisaged fibre volume fraction of 60% in the Al-MMC composite.
This case is then introduced at the bottom of the autoclave inside one additional working graphite die. This controls the melt reservoir as well as the melt flow during processing and pressure application. On top of the securely placed inner case, a block of solid aluminium (Al99.85%, or AlSi7Mg) is placed at room temperature and then, after positioning the thermocouples in their different prepared points, the autoclave is secured and closed.
The process starts with vacuum application in the whole system. This leads to combustion of the fibre sizing at low temperatures and prevents oxidation of the carbon fibre surfaces at temperatures beyond 500°C. The metal stock heats up beyond melt temperature by three resistance heating rings stacked vertically around the whole mounting inside the pressure chamber (see Figure 6). The melt fills all open cavities in the die as well as all interspaces in the fibre stack due to vacuum and additional pressure application.
After infiltration of the alloy, the heating is discontinued, the water cooling system activated, and the gas pressure applied. The system is cooled down under high pressure until it reaches room temperature. The pressure is released, and the die removed from the autoclave. After gas infiltration process the final C-fibre reinforced Aluminium plate has to be machined out of the manufacturing die as well as the fibre placement container. As a result, LKR manufactures plates as shown in Figure 5.

The LKR gas infiltration process was developed in 1995 and since then only minor changed. Within project SpAACe is goal to advance mechanical properties through stabilization of process robustness and update of process relevant technologies. Three steps have been done to advance the gas infiltration process:

1. <u>Change of heating system:</u> The original resistance heating system was exchanged by a low-volume, low weight coil tube heating cartridge systems. This is a) much lighter compared to the former heating and b) possesses far quicker reaction times to changes of process control parameters. In this way, a total of 12.3 kg could be saved which in terms of heat-up energy stands for a total of 4040.55 kJ or 1.12kWh which is saved. This means on the one hand a faster heat up time as well as a faster cooling time due to the omission of the "heavy" resistance heating.
2. <u>Change of process control</u>: Using the novel coil heating system led to the need for modification of the process control so to speed up activation and change times during gas infiltration process.
3. <u>Change of cooling system:</u> When running the gas infiltration process, the final infiltrated dies get vertically transferred into a cooling section by a pneumatic table (see Figure 4). This cooling section has been modified towards higher flow rates of the cooling water so to increase cooling rates in the die. It is mandatory to fall below 500°C as well as 300°C as quick as possible to oppress the evolution of Al_4C_3 carbides on carbon fibres as much as possible. These reduce the fibre cross-section and led to notch effects at the same.

These three modifications led to a far better control of the gas metal infiltration process, to a more stable and reproducible process as well as to an overall quicker process in terms of heat-up times, cool-down times as well as higher cooling gradients.

3. Al-MMC Coupon Plates

During the first project phase an initial test matrix was conducted to generate a) reference values when varying process parameters with the "old" process system and b) values when working with the modified process system (see following Table 1 and Table 2).

Table 1: First project phase with "old" system for reference investigation purpose

Al-MMC process test	Fibre-type	Matrix	Fibre volume fraction	Cartridge type
633	M40J-6K	Al99,85	59	New
634	M40J-6K	Al99,85	59	New
635	M40J-6K	Al99,85	59	Used
636	M40J-6K	Al99,85	59	Used
637	M40J-6K	Al99,85	60	New
638	M40J-6K	AlSi7Mg	60	New
639	M40J-6K	Al99,85	65	Used
640	M40J-6K	Al99,85	65	Used
641	M40J-6K	Al99,85	59	Used
642	M40J-6K	Al99,85	59	Used

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Table 2: Second project phase with updated gas metal infiltration process at LKR

Al-MMC process test	Fibre-type	Matrix	Fibre volume fraction	Cartridge type
648	M40J-6K	AlSi7Mg	59	Used
649	M40J-6K	AlSi7Mg	59	Used
650	M40J-6K	AlSi7Mg	59	Used
651	M40J-6K	AlSi7Mg	65	Used

Out of tests conducted in table 1 (not presented in this paper) it was concluded that process phase 2 should be run with the modified gas metal infiltration process and an AlSi7Mg matrix system. AlSi7Mg was defined as this together with high cooling rates contributes to the suppression of disadvantageous Al_4C_3 carbides. The positive effect of adapted process and this matrix alloy on the mechanical properties will be visible in the results section.
In each of the process test campaigns one coupon plate was manufactured with maximum dimensions of (160 x 65.5 x 2.0 mm). Figure 6 shows the resulted Al-MMC plate of gas metal infiltration process run V651 in demoulded state and the according cross-section view on the carbon-fibre reinforced AlSi7Mg Aluminium alloy matrix with a final fibre volume fraction of 65%.

Figure 6. Resulting Al-MMC coupon plate after manufacturing (left) and cross-section of the Al-infiltration.

Out of these Al-MMC plates different samples were manufactured according to an internally defined coupon preparation milling plan for mechanical tests and microstructural investigations (see Figure 7). Mechanical tests consisted of short beam 3-point bending tests (3P), interlaminar shear stress tests (ILS), 0°-tensile tests (T). For micro-structure investigation (C-samples) light microscopy, high resolution SEM, and computer tomography tests were used.

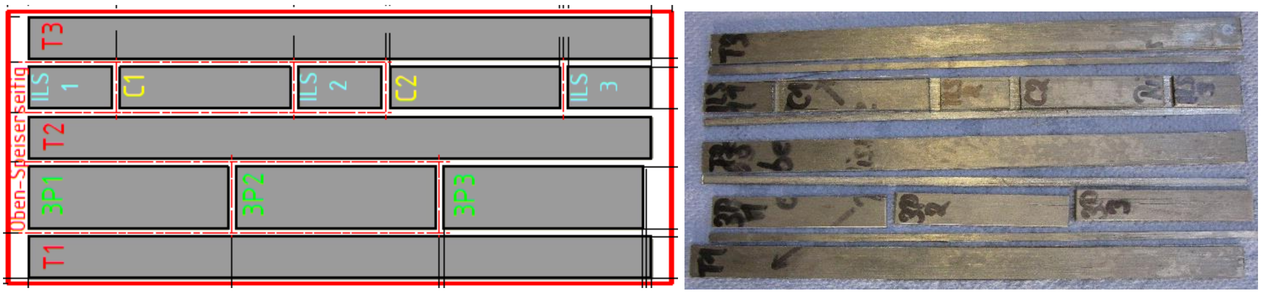

Figure 7. Sample plan (left) and final, milled Al-MMC samples out of coupon plate for mechanical testing.

ILS-tests were conducted on series of three parallel, short, double-sided notched coupons with dimensions of 20 x 10 x 2 mm. 3-point bending tests were run on series of three parallel rectangular coupons with dimensions of 48 x 15 x 2 mm. The three, parallel tensile test specimen possessed maximum dimensions of 150 x 10 x 2 mm. All samples were tested by AAC, Aerospace & Advanced Composites GmbH, an ESA-certified test-house in Austria.

4. Results of mechanical tests

The following Figures 8 – 11 show the mechanical test results of the coupon plate tests samples manufactured with the process configurations listed in tables 1 and table 2.
In phase 1 (process trials with "old" process setting) it was investigated whether the use of new or used fibre cartridges leads to a prominent role for the final mechanical properties, but no significant correlation could be drawn. V639 and V640 possess a higher fibre volume fraction of 65%. This leads to slightly higher ILSS and bending stress levels for V639 but not for V640. V638 shows reduced ILSS levels, moderate bending strength, and slightly reduced Young´s modulus. But its ultimate tensile strength turns out significantly higher than all other plate versions of phase 1. Summarized in short it can be stated that all varied parameters do not lead to a significant increase of mechanical properties by a factor of two or more so to approach theoretical values as targeted in figure 3.

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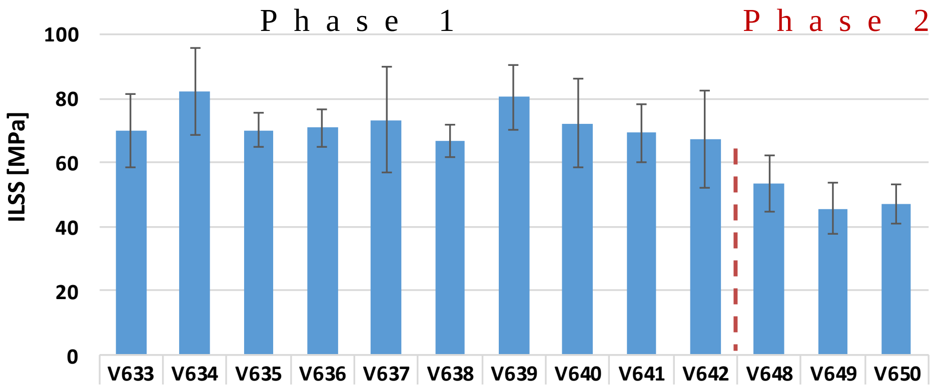

Figure 8. Interlaminar shear test results of coupon plate test samples V633 – V650

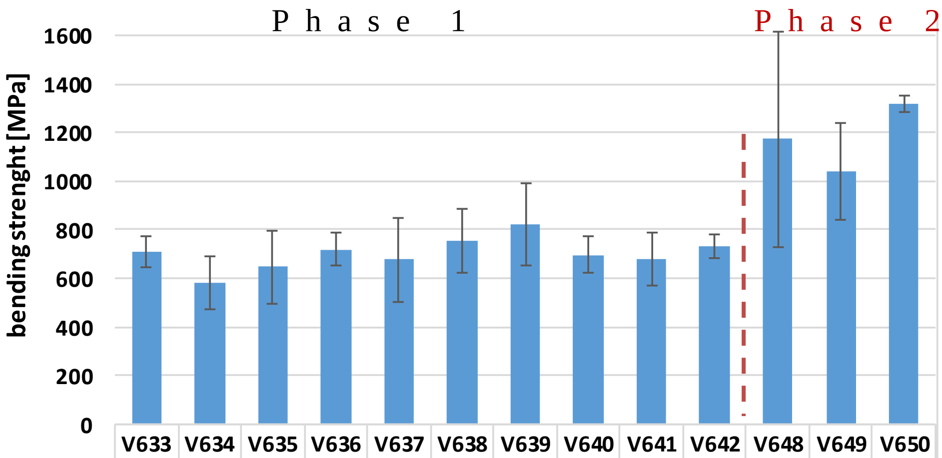

Figure 9. Bending test results of coupon plate test samples V633 – V650

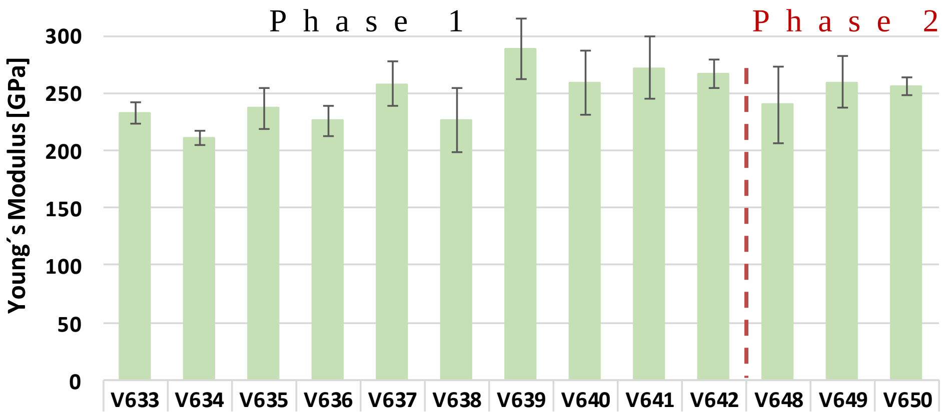

Figure 10. Young´s Modulus results of coupon plate test samples V633 – V650

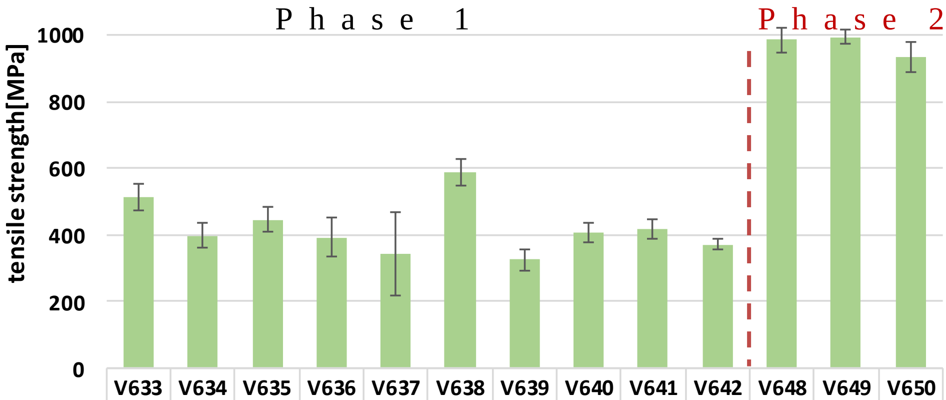

Figure 11. Tensile test results of coupon plate test samples V633 – V650

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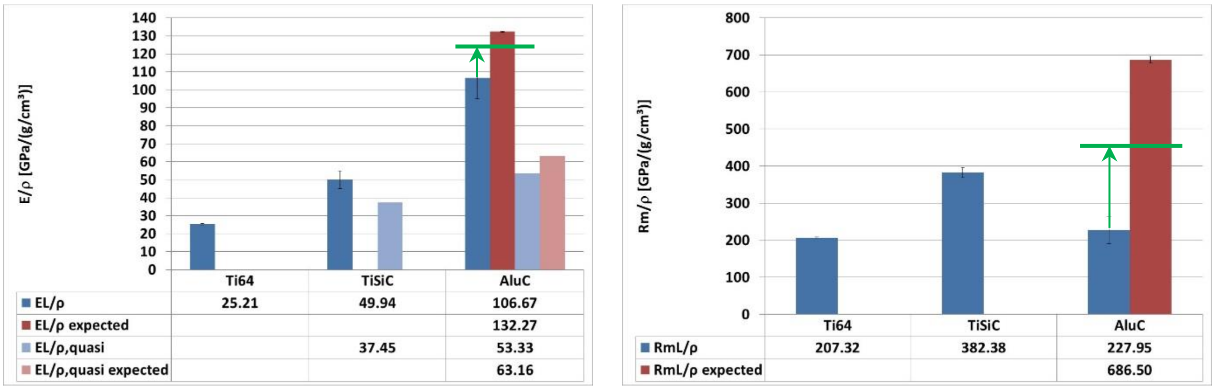
In contrast, mechanical results of phase 2 material samples show distinctly increased mechanical properties for bending strength as well as tensile strength. In terms of bending strength, values increased from 600-800 MPa in the first phase up to beyond 1000MPa in the second phase (V649) and even up to 1250 MPa with a very narrow scatter band. In terms of tensile tests, ultimate strength raised up to values between 934 – 993 MPa with a std. deviation of 2-5%. The Young´s Modulus values remained at ~250GPa. Only interlaminar shear strength reduced down to values between 53 – 45MPa.

Given the values of phase 2 and a calculated density of 2.1 g/cm³, Young´s Modulus of V649 turns out to 123 GPa/g/cm³. This is +15% higher compared to measured values of past material projects of AAC and ESA and 6.7 % less then theoretical Young´s Modulus of figure 3. Density specific ultimate strength of Al-MMC materials of phase 2 reach maximum values of 472 MPa/g/cm³. This is +107 % compared to past material projects of AAC and ESA (compare Figure 1) but still 31.1% below theoretical maximum values plotted in Figure 3. Figure 12 shows the achieved intermediate achievements in green compared to reference materials and "old" AluC data of the past project.



(a) Specific stiffness (b) Specific strength

Figure 12: Improved (a) density specific stiffness and (b) density specific ultimate strength (green horizontal line) achieved compared to Ti64, TiSiC and AluC values (blue) and theoretical values (red) of (see also Figure 3).

5. Conclusion and Outlook

Project SpAACe is a currently running project of LKR Ranshofen and AAC Aerospace & Advanced Composites GmbH with the tasks to update the metal gas infiltration process at LKR and to stabilize process conditions so to advance long carbon fibre reinforced Aluminium Al-MMCs in their mechanical properties. Results of two project phases have been presented. In the first phase, with the "old" process setup, different process parameters and material parameters were changed. But these did not lead to significant improvements of investigated mechanical results. In the second phase, after installation of new process technology items, process stabilization as well as faster heating and cooling times could be achieved. This in combination with the change of matrix alloy to AlSi7Mg has led to a significant improvement of mechanical results.

After definition of a final process parameter definition set and a final matrix alloy system, a big size, ESA-like test campaign will be conducted both at earth-like environmental conditions at low and elevated temperatures (-100°C to +100°C). Besides 0°, this test matrix will also include the screening of AluC material with fibres aligned at ±45° and 90°. Test are also intended under Martian-like conditions so to derive information on mechanical performance at harsh environments. Afocus will be put on metal coating and protection surfaces so to prevent stress crack corrosion.

6. Acknowledgements

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