Effect of production technique on a high silicon al-si-ni sintered alloy structure and properties

Kaverynskyi V.[1], Bagliuk G.[1], Verbilo D. G.[1], Kyrylyuk S. F.[1], Sukhenko Z.[1], Rud A. D.[2]

Institute for Problems in Material Science, Ukraine[1]

Institute for Metal Physics, Ukraine[2]

Abstract: *An experimental study was carried out to develop a technique for manufacturing a high silicon Al-Si-Ni sintered alloy with a coefficient of thermal expansion close to steels. A stage of hot forging was added to overcome porosity and improve silicon inclusions morphology to obtain better mechanical properties. Liquid and solid phase regimes of sintering were studied for the material made of the elemental components powders mixture as well as of a pre-alloyed powder. The impracticality of liquid-phase sintering modes is shown for this type of material primarily because of the tendency to liquid-phase exudation. It was proposed to carry out a solid-state vacuum sintering with subsequent hot deformation. Only the pre-alloyed powder is suitable for the technique; the other way, it leads to the course and sometimes, not equilibrium intermetallic inclusions will form in place of nickel particles because of the insolubility of Ni in solid Al. It is shown the possibility of successful plastic deformation of this material after sintering despite its extremely low ductility, which helps successfully overcome porosity and improve the characteristics of the microstructure of the material.*

KEYWORDS: ALUMINIUM, SILICON, NICKEL, SINTERING, HOT FORGING, COEFFICIENT OF THERMAL EXPANSION

1. Introduction

Lightweight materials with the given physical properties are crucial for aerospace equipment production [1, 2]. For precise navigation devices, the coefficient of thermal expansion (CTE) is extremely important to be conformed to one of the contact materials such as steel, copper alloys etc. It is known that device details instability contributes up to 20 – 50 % of its total error [3].

Light materials on aluminium base are well proven owing to a number of their advantages such as corrosion resistance, plasticity, manufacturability, non-toxicity and relatively low cost [4]. However, CTE aluminium is rather high – $23.4 \cdot 10^{-6}$ K^{-1} (for 20 – 100 ºC) [5], which is significantly more than for steel ($11 – 15 \cdot 10^{-6}$ K^{-1} [6]) or copper ($16.8 \cdot 10^{-6}$ K^{-1} [7]). Thus, it seems to be an important task to reduce the CTE of aluminium-based materials with the mentioned destination. It is achieved mainly through alloying.

According to data from [4] Cr, Si, Be, and Cu reduce the CTE of aluminium. A low density of silicon and its efficiency in CTE decreases, making it primary for the purpose. However, we practically can't increase its content by more than 30 – 35 %, which would significantly reduce mechanical properties. But the CTE value obtained at such Si content is usually not enough. Nevertheless, it could be additionally reduced by an additional alloying with other elements. Among them, Ni is proven to be one of the most effective and harmless mechanical properties. 5 – 7 % of Ni is usually enough to obtain the desired CTE at 25 – 30 % of Si.

Casting technology is quite inadmissible for such high silicon materials with Ni mainly because of a wide range of solidification temperatures from ~800 – 900 °C (liquidus) to 557°C (solidus) [8, 9, 10]. Mentioned fact causes a significant liquation and a tendency for primary Si inclusions to grow large and course. So, the Si content in cast alloys often doesn't surpass 20 – 23 % [11]. Thus, powder metallurgy seems to be an appropriate manufacturing technique for the materials considered here.

However, there are questions of solid or liquid state sintering workability and efficiency as well as about the usage of a powder of a previously made alloy (then –pre-alloyed powder) or a mixture of the elemental powders. Some studies of liquid phase sintering for Al-Si, Al-Cu, Al-Si-Cu, and Al-Zn alloys [12 – 17] report its positive effects, such as porosity overcoming and better components consolidation. But all of them consider materials with Si content not more than 12 – 14 % and with other alloying elements, like Cu, Zn, or Mn. Thus, the production methods acceptable for the material considered here need a special study.

One of the ways to overcome porosity appearing in powder materials is hot deformation processing. However, Al-Si materials are known to be brittle and even hot deformation may need a special technique. Nevertheless, it is possible, and we had research experience in the area of Al-Si deformation-heat treatment study, and some results of structure and properties improvement were achieved, but for cast alloys with lower Si content [18]. Here such a method is to be allied to the material produced through the powder metallurgy and with a significantly higher amount of Si.

2. Experimental material and technique

Approaches studied here are devoted to both a mixture of the pure initial components powders and a powder made of the pre-alloyed cast product. For the first one were used the following powders: Al 10 – 300 μm, Si 0.5 – 70 μm, and Ni 20 – 120 μm. The pre-alloyed powder was obtained through grinding in a planetary ball mill of the quickly cooled strips 30 – 50 μm strips. The obtained average particle size was 106±5 μm with variation from 4 to 380 μm.

The average chemical composition of the powders according to the spectral analysis is given in Table 1.

Table 1

Chemical composition of the powders, wt. %

Material type	Al	Si	Ni	Fe	P
Pre-alloyed	base	31.32	6.51	0.068	0.097
Mixture of elemental powders		32.91	5.91	0.17	0.090

The powders were compacted under a pressure of 500 – 600 MPa. Steel covering shells were used due to it was assumed that they might optimize the subsequent deformation (forging) process, bringing it closer to the mode of all-around compression to reduce a brittle fracture probability. The outer diameter of the shells was 32 mm, the inner diameter was 28 mm, and the height, including the bottom and cover, was 37 mm. The bottom was welded before pressing. The top covers were set up after pressing and sintering.

The sintering occurs in a vacuum furnace. The temperature was set at 560 ºC for the liquid phase sintering and 530 ºC for the solid phase one. The sintering duration was 40 – 45 minutes for the liquid state one and 2 hours for the solid state one. Also, a preheating was used to 350 ºC and kept at that temperature for about 30 minutes for the adsorbed water and hydrogen purification. The longer sintering time for the solid-state process was due to slower diffusion in the solids.

A special die was constructed to carry out the hot forging to provide a deformation regime close to all-around compression. The forging temperature was 520 – 540 ºC.

Dilatometer measurements were performed on an induction quartz dilatometer assembled based on an induction transducer model 275-01 for measuring linear movements, the relative measurement error of which is ±0.5 %.

Mechanical tests were carried out on cylindrical samples with a working part length l0 = 20 mm and ∅ 3 mm with deformation velocity $\varepsilon' = 2 \cdot 10^{-3}$ c^{-1}.

The study of the phase composition of the alloys was carried out on a DRON-4M diffractometer using Cu$_{K\alpha}$ and Co$_{K\alpha}$ radiation.

Electronic microscopy study and X-ray microanalysis of the present phases and structural entities composition were performed using TESCAN MIRA 3 device.

The microstructure of the samples was studied by optical microscopy. Metallographic studies were carried out on a NEOPHOT-2 optical microscope.

3. The results of the experiments and their discussion

Liquid phase sintering was found to lead to the liquid phase exudation both for materials from elemental components powders and from the pre-alloyed powder. Because of this reason porosity remained at the level of 20 – 25 % after such sintering.

The liquid phase exudation phenomenon, which is uncontrolled, is highly undesirable because of material loss and the chemical composition perversion. Also, it causes inner defects such as pores and hollows to appear, which couldn't be subsequently repaired. A liquid phase exudation is usually associated with poor wettability of the solid phase by the liquid. However, no information about its appearance during Al-Si alloys sintering was found in the literature. Usually, such a problem occurs for other systems, for example, Al-Sn [16].

The microstructure of the material after the liquid phase sintering is shown in Figure 1.


a)   b)
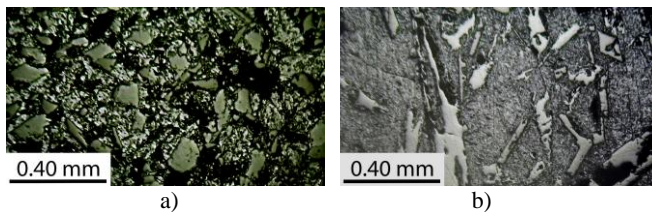
Figure 1 – The material structure after liquid phase sintering without subsequent deformation: a) from components mixture, ×40; b) from the pre-alloyed powder, ×40

A significant porosity seen from the structure remains after liquid phase sintering (Figure 1 – a) means that the liquid doesn't tend to fill the pores but leaks out. In the material made of the pre-alloyed powder (Figure 1 – b), we don't observe such a significant porosity on a micro scale, but rather macro defects (hollows). Silicon inclusions look similar to ones in the cast metal.

Because of the fact of liquid phase exudation, the decision was made to focus on the solid-state sintering with subsequent hot deformation. It should be noted that the task was to obtain a material of a certain composition which couldn't be changed significantly, so the composition correction to prevent the exudation was not an option.

A sample made of the powders mixture structure after solid phase sintering and subsequent hot forging at 530 – 540 ºC is given in Figure 2.
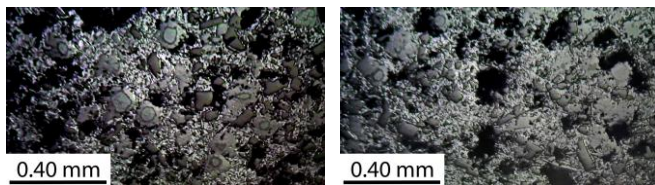
Figure 2 – The structure of the material made of the powders mixture after solid phase sintering and hot forging at 530 – 540 ºC

Even after hot plastic deformation, such material remains porous. Silicon inclusions sizes are 50 – 70 μm. Also, specific entities having the shape of spherical inclusions with reams are observed in the structure. They are nickel particles surrounded by a layer of intermetallics, which was confirmed by X-ray micro spectral analysis. Figure 3 shows an SEM image of a part of such inclusion with the results of local analysis.
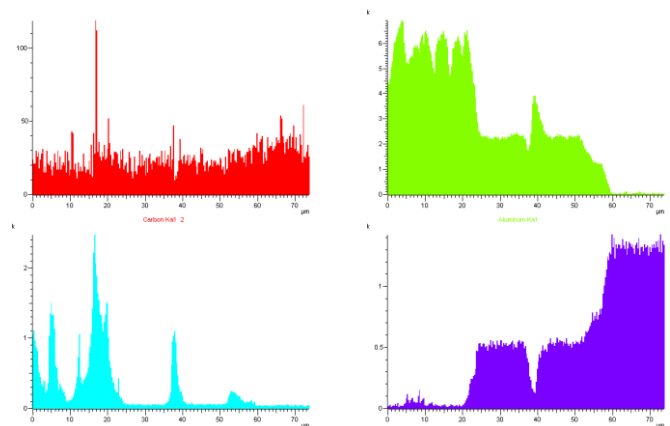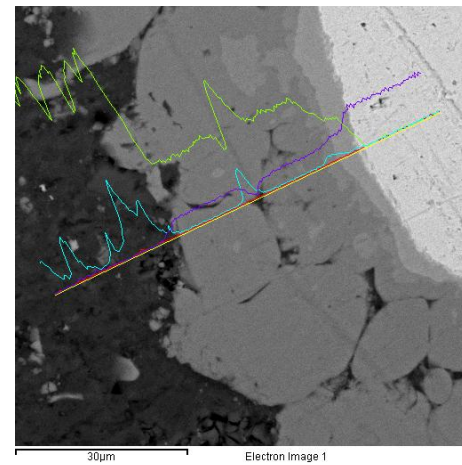
Figure 3 – Results of local X-ray microspectral analysis of the intermetallic structural component in a sample from a mixture of powders after solid phase sintering

The intermetallic shell is not dense, and it has cracks, but hot deformation, in this case, does not destroy such entities. Most of the intermetallic shell consists of $NiAl_3$. On the side of nickel particles, there is a rather thin (2 – 3 μm) layer of other intermetallic with a greater Ni concentration, which is probably Ni_2Al_3. Therefore, the nickel has not completely formed intermetallic compounds, so the final structure is non-equilibrium. The formation of such large-sized aggregates is because nickel almost does not dissolve in solid aluminium.

The best structure was obtained after the solid phase sintering and hot forging at 520 ºC for the samples made of the pre-alloyed powder. Before the forging, the sintered samples were affected by preliminary hot deformation from sides with a setting degree of ~20 − 30 % at 510 – 520 ºC. The structure of the material obtained in such a way is shown in Figure 4.

Silicon and intermetallic inclusions are dispersed (1 – 7 μm) and evenly distributed in the metal matrix. Thus, according to the obtained structure, such a manufacturing method looks the most acceptable for further production of the material.

Despite the shown disadvantages, the hot forging of the specimens after the liquid phase sintering was also studied here. In Figure 5 are shown the structures of the samples from the powders mixture and hot deformation. The porosity has decreased compared with the sintered state but remain. Those specific intermetallic entities observed after solid phase sintering were not found here.

The structures of the samples made of pre-alloyed powder after liquid-phase sintering and hot forging are shown in Figure 6. In the samples, affected by deformation at 530 – 540 ºC without preliminary lateral hot deposition of the sample, areas with a typical cast structure are still observed – Figure 6 (a). Defects (pores) are also observed in these areas. However, most of the structure demonstrates equiaxed polyhedral silicon inclusions with a range of sizes from ~5 μm to ~60 μm and even more. Their average size, in this case, is 26.4±4.7 μm.
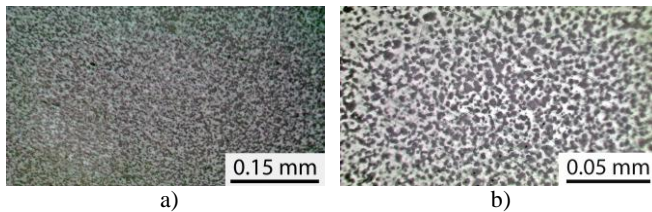
Figure 4 – The structure of the material from the pre-alloyed powder after solid phase sintering and hot forging at 520 ºC: a) ×40; b) ×500.
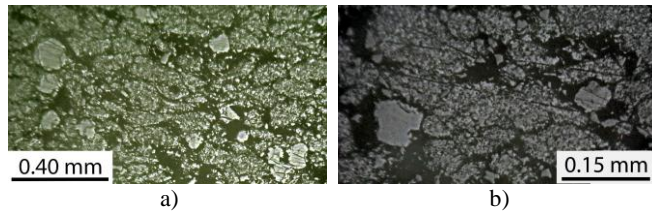

Figure 5 – The structure of the material from the powders mixture after liquid phase sintering and hot deformation at 540 – 550 ºC: a) ×40; b) ×100.
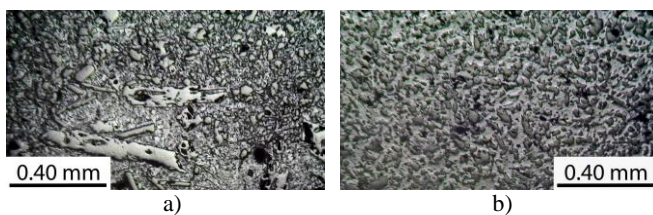

Figure 6 – The structure of the material from the pre-alloyed powder after liquid-phase sintering and hot deformation: a) forging at 530 – 540 ºC, ×40; b) forging at 520 ºC with additional lateral pressing at 510 – 520 ºC, ×40

Lowering the forging temperature to 520 ºC with adding the previous pressing from the sides at 510 – 520 ºC leads to some structure improvement (Figure 6 – b). However, the silicon inclusions are still larger than the ones obtained after the solid phase sintering. Here were not found those cast-looking structures and significant pores. The inclusions sizes vary from ~5 μm to ~40 μm with an average size of 19.8±2.3 μm.

X-ray diffraction quantitative phase analysis was carried out to clarify the phase composition of the material produced by different techniques. Its results are presented in Table 2.

The phases detected by this method were the solid solution of Al, Si and an intermetallic compound $NiAl_3$. The lowest amount of $NiAl_3$ was found for types made from a mixture of powders. The possible and main reason for this may be the incompleteness of the intermetalide formation reaction. For one, after the liquid phase sintering, an additional reason may be the mentioned liquid phase exudation since $NiAl_3$ is part of the eutectic that melts and flows out at sintering temperatures. The most $NiAl_3$ was found in the samples made from pre-alloyed powder after the solid phase sintering. This amount looks very close to the equilibrium for the studied composition.

The method also allowed the estimation of the coherent scattering region sizes for the present phases of the studied material (table 2).

The sintering method has a valuable effect on the coherent scattering region sizes, especially for Al and Si-based phases. After solid-phase sintering, they are ~1.5...2 times larger than after sintering with the presence of the liquid phase. The effect is observed even though the specimens were made from completely different initial powders, which additionally indicates it.

Only for the samples made of pre-alloyed powder solid phase sintering was it possible to estimate yield strength. Such samples exhibited some but not significant ductility: total elongation was not more than ~2 %. Such specimens exhibited the highest ultimate tensile strength and hardness.

Table 2

*Quantitative estimation of the phase composition of the studied specimen based on the X-ray diffraction results, weight %, and sizes of the X-rays coherent scattering regions, Å ***

Specimen type	Al	Si	$NiAl_3$
From the powders mixture, after the solid phase sintering at 350 ºC (~30 min) + 530 ºC (~ 2 h) and forging at 530 – 540ºC	62.37 ± 0.83	26.09 ± 0.36	11.55 ± 0.17
	900	1020	660
From the pre-alloyed powder, after the solid phase sintering at 350 ºC (~30 min) + 530 ºC (~ 2 h) and forging at 520 ºC	56.76 ± 0.51	24.83 ± 0.27	18.40 ± 0.31
	920	940	480
From the pre-alloyed powder, after the liquid phase sintering at 560 ºC (40 – 45 min) and forging at 530 – 540 ºC	59.92 ± 0.61	25.88 ± 0.30	14.19 ± 0.32
	520	560	460
From the pre-alloyed powder, after the liquid phase sintering at 560 ºC (40 – 45 min) and forging at 520 ºC with previous pressing from the sides at 510 – 520 ºC	58.87 ± 0.44	26.78 ± 0.23	14.35 ± 0.20
	660	700	450
From the powders mixture, after the liquid phase sintering at 560 – 580 ºC (~40 min) without forging	67.08 ± 0.74	22.59 ± 0.26	10.33 ± 0.15
	470	660	460

* - upper value – weight % of the phase; lower value – size of the X-rays coherent scattering regions, Å

The structure patterns obtained after different ways of manufacture reflect the mechanical properties. The data obtained through tensile tests and hardness measurements are presented in Table 3.

Table 3

Mechanical properties depending of the way of the material obtaining

No.	Samples' type	σ_Y, MPa	σ_U, MPa	Hardness, HB
1	From pure components mixture. Liquid phase sintering and hot forging at 530 – 540 ºC.	—	81.3 ±27.2	68.1 ±5.3
2	From the pre-alloyed powder. Liquid phase sintering and hot forging at 530 – 540 ºC.	—	116.6 ±10.5	76.9 ±4.5
3	From the pre-alloyed powder. Solid phase sintering and hot forging at 520 ºC with preliminary hot deformation by pressing from sides.	220.7 ±10.3	261.2 ±20.9	95.8 ±3.2
4	From the pre-alloyed powder. Liquid phase sintering and hot forging at 520 ºC with preliminary hot deformation by pressing from sides.	—	139.7 ±12.4	82.5 ±5.7

All the other studied types of samples have significantly lower strength and hardness values, which are caused by larger and rough-shaped silicon inclusion in addition to porosity. Among them, the strongest were the samples made of the pre-alloyed powder after liquid phase sintering and forging at 520 ºC with preliminary hot lateral deformation, which was almost nonporous. The highest ultimate tensile strength shown for such material was 145.4 MPa. Samples made of pure components mixture have the lowest strength and are very brittle. These facts additionally prove the benefit of using the pre-alloyed powder and the inexpediency of liquid phase sintering for the material.

The considered material also is to have a coefficient of thermal expansion (CTE) close to those of steel. To measure it, were carried out dynamometric tests. These are given in Table 4.

Table 4

CTE values of the material at the studied temperatures

Samples' type	CTE, K^{-1}, $\times 10^{-6}$ at the temperature, ºC					
	50	100	150	200	250	300
Cast state		13.2		14.2		
Liquid phase sintering and forging	10.51	12.95	13.18	13.49	13.82	14.13
Solid state sintering and forging	13.81	14.98	15.42	15.84	16.19	16.52
Solid state sintering and forging + annealing at 410 – 420 ºC for ~1 hour	13.93	15.10	15.72	16.30	16.69	16.99

Because liquid phase exudation leads to an increase in silicon content, the samples after the liquid phase sintering have lower CTE values. For the specimens after the solid-state sintering and hot forging, its value at lover temperatures is $13.81 \cdot 10^{-6}$ K^{-1}, which is quite close to steel CTE. However, for low-alloyed steels, it is lesser – $11 \dots 12 \cdot 10^{-6}$ K^{-1}. Nevertheless, it is closer to it than ones of pure aluminium and cast Al-Si alloys. The closest CTE is observed for Cr and Ni alloyed stainless steels, high Cr instrumental steels, and special steels with Cr and Co. Annealing at 410 – 420 ºC for ~1 hour after deformation might lead to some rise of CTE, especially for the higher temperatures.

4. Conclusions

An experimental study was carried out to find out an appropriate manufacturing method to produce the high silicone Al-Si-Ni alloy (25 – 30 % Si and 5 – 7 % Ni) with a close to steel CTE value. The hot forging stage was applied to obtain better mechanical properties. Liquid and solid phase sintering types were studied.

The best structure and mechanical properties values were obtained for the material made of the pre-alloyed powder using the following method: pressing (500 – 600 MPa) of the powder into steel shells, vacuum sintering (30 min at 350 ºC then 2 h at 530 ºC), lateral 20 – 30 % pressing at 510 – 520 ºC, hot forging at 520 ºC. Such a technique allows obtaining of a non-porous material with homogenously distributed fine spherical silicon inclusions with sizes of 1 – 7 μm.

It was found that liquid phase exudation during the sintering makes such a technique impractical for material production. Not only does it lead to uncontrollable distortion of the chemical composition and promotion of pores and hollows formation, but it also tends the silicon crystals in the liquid to grow, which further rough cast-like structures with significantly lower strength.

It was shown high insolvency of elemental powder mixture usage as the raw martial for the solid-state sintering. It is because Ni doesn't solve in solid Al and tends to form rough intermetallic aggregates that could include non-equilibrium compounds. Instead, a pre-alloyed powder of the given composition would be a far better option. It could be obtained in different ways, for instance, by grinding a quick solidified strip.

It was found that the manufacturing method also has a significant influence on the sizes of the X-ray coherent scattering regions for the Al and Si phases. They appear ~1.5 – 2.0 times larger after the solid phase sintering than after liquid phase one, either for specimens manufactured from the powders mixture or the pre-alloyed powder.

Obtained as a result, this study material has high mechanical properties (σ_Y ~ 221 MPa, σ_U ~ 261 MPa, and hardness ~ 96 HB), and its CTE is proven to be appropriate.

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