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Inoculation of pure aluminium aided by electromagnetic field

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Abstract

The main aim of studies was to determine common influences of two different refinement methods on EN AW-Al99,5 primary structure and developing of refinement mechanism. The first method is inoculation with small amount of (Ti+B) less than obligatory standard PN-EN 573-3 and the second method is influence of electromagnetic field on aluminium crystallization process. These methods of structure refinement are particularly important in continuous and semi–continuous casting where products are used for plastic forming. Large columnar crystals zone result in forces extrusion rate reduction and during the ingot rolling delamination of external layers can occur.

Key words: Mechanical properties, Aluminium, Titanium, Boron, Electromagnetic field

1. Introduction

Columnar crystals which are parallel to heat flow, creates primary structure of pure metals independently from type of crystal lattice (fig.1a). This unfavourable structure for plastic forming of ingots can be eliminated by controlling of heat abstraction velocity from cast (fig.2), change in chemical constitution (fig.1b) and liquid metal convection (fig.3) [1].

Effective method of columnar crystals zone elimination is inoculation with introduction into metal bath of specified substances, called inoculants, increase grains density as result of creation of new particles in consequence of braking of grains growth velocity, decrease of surface tension on phase boundary of liquid – nucleus, decrease of angle of contact between nucleus and "washer" and increase of density of "washers" to heterogeneous nucleation. This leads to increase of equiaxed crystals zone, which guarantee of mechanical properties improvement, decrease of constituents segregation and limitation of hot cracks [1÷5].

Active base to heterogeneous nucleation for aluminium are particles which have high melting point i.e. TiC, TiB, TiB_2, AlB_2 and Al_3Ti (tab.1) [1÷7].

Table 1.
Characteristic of crystal lattice of "washres" to heterogeneous nucleation formation in aluminium [1, 2, 4, 8]

Phase	Melting point (circa) [°C]	Type of crystal lattice	Parameters of crystal lattice [nm]
Al	660	Cubical A1	a = 0,404
TiC	3200	Cubical B1	a = 0,431
TiB	3000	Cubical B1	a = 0,421
TiB_2	2900	Hexagonal C32	a = 0,302 c = 0,321
AlB_2	2700	Hexagonal C32	a = 0,300 c = 0,325
Al_3Ti	1400	Tetragonal $D0_{22}$	a = 0,383 c =0,857

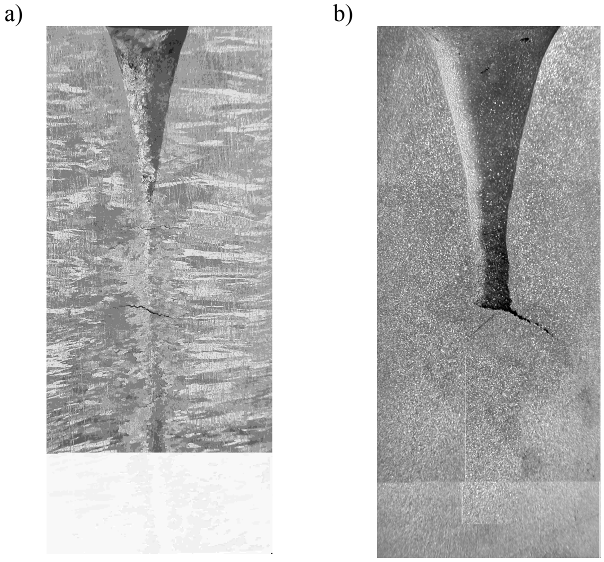

Fig.1.Macrostructure of aluminium ingot longitudinal section:
a) without inoculation,
b) after inoculation with (Ti+B)

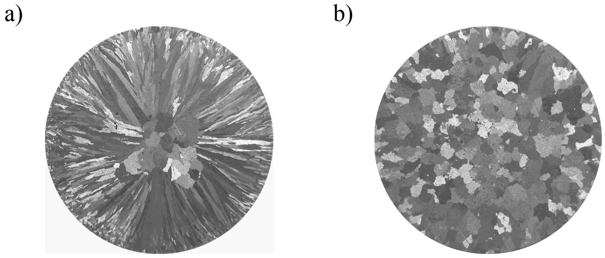

Fig.2. Macrostructure of aluminium ingot cross-section after pouring into: a) graphite mould, b) shell mould

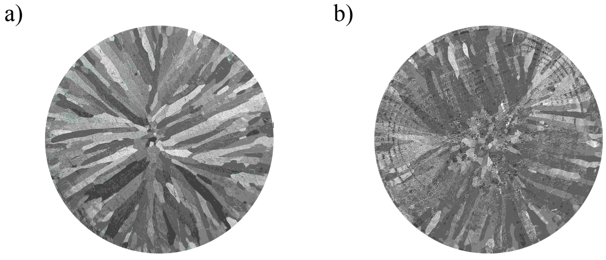

Fig.3. Macrostructure of aluminium ingot cross-section after casting: a) without influence of electromagnetic field, b) with influence of electromagnetic field

However, this method of inoculation of primary structure is limited for pure metals, because inoculants decrease the degree of purity specified in EN-PN standards. Moreover, inoculant influences negatively on physical properties i.e. electrical conductivity of pure aluminium (fig.4).

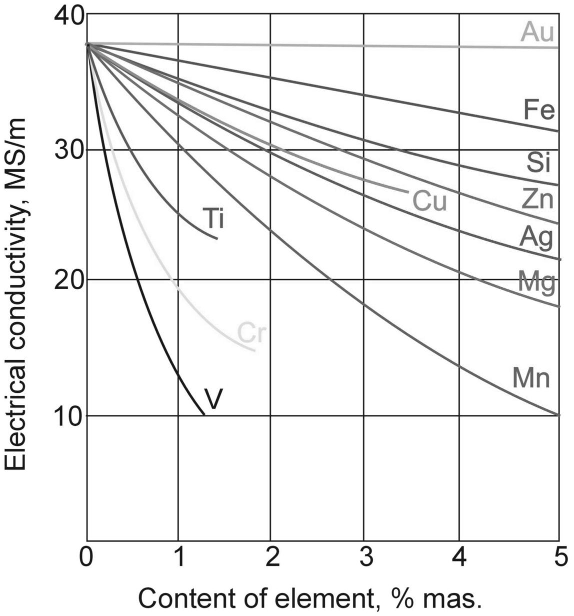

Fig. 4. Influence of different elements on electrical conductivity of EN AW-Al99,99 [9]

But introduction of small amount of inoculant can be strengthened by use of size reduction other method i.e. use of ultrasonic vibration or electromagnetic field to force liquid metal movement in mould.

Forced liquid metal movement influences in diversified way on changes in structure of casting i.e. by changes of thermal and concentration conditions on crystallization front, which decrease or completely stops the velocity of columnar crystals growth and by [1, 10÷15]:

- tear off of crystals from mould wall, which are transferred into metal bath, where they can convert in equiaxed crystals,
- parting of dendrite by coagulation and melting as result of influences of temperture fluctuation and breaking as result of energy of liquid metal movement,
- crystals transport from free surface to inside the liquid metal,
- crystals from over-cooled outside layer of bath are transported into liquid metal.

On the base of presented dates was accepted thessis that contribution of these both mechanism models of additional creystal nucleuses formation i.e. electromagnetic field influence and introduction of small amount of inoculant – less than in conventional modyfication process, should result in higher size reduction in pure metals structure. It is not possible, when we use one of these methods, we must use these two methods together.

2. Range of studies

The main aim of studies was to determine common influences of inoculation with small amount of (Ti+B) less than obligatory standard PN-EN 573-3 (concerning about aluminium purity) and of electromagnetic field on crystallization process and structure of aluminium EN AW-Al99,5 and developing of refinement mechanism.

After study of literature datas [1÷5] in investigations inoculant of type AlTi5B1 was used. This inoculant increas size reduction, because its introduction result in formation of “washers” to heterogeneous nucleation of aluminium (tab.1). Inoculants quantity was (25Ti+5B) ppm and was controlled in result of chemical analysis, which was executed with use of emission optical spectrometry method on plasma spectrometer ICP of type JY 138 ULTRAFACE of YOBIN YVON (tab.2).

Aluminium was melted in inductive furnace and temperature was measured with use of NiCr-NiAl thermocouple (pouring temperature was set to 740 °C). Metal was poured into the graphite mould with wall thickness 7 mm. Test castings as ingots with dimensions of 45 mm diameter and 180 mm length were casted with exactly specified parameters: pulse frequency of electromagnetic field, magnetic induction and time of electromagnetic field action. These parametres characterize used in investigations modified methody of casting in rotating electromagnetic field in form of impulse reverse electromagnetical field and became optimized on basis of earlier investigations of aluminium EN AW-Al99,98 [14, 15]. Statistical analysis with using stepwise regression of this results was selected to determine relations between variable factors of casting and parameters of structure. Following function was wanted:

$$SKR = f\,(f, B_i, M) \qquad (1)$$

where:
SKR – equiaxed crystals zone content on cross-section of ingot, which was measured on macrostructure with use of computer programme Multi Scan Base v. 13.01,
f – pulse frequency of electromagnetic field, variation level 0,5 and 1 Hz,
B_i – magnetic induction, variation level 35 and 45 mT,
t – time of electromagnetic field action, variation level 15 and 30s,
M – inoculant quantity (Ti+B), variation level (100Ti + 20B) and (200Ti + 40B) ppm;
Statistical analysis resulted in function shown below:

$$SKR = -3{,}74\cdot f + 0{,}41\cdot B_i + 0{,}14\cdot M + 0{,}04\cdot t - 25{,}6 \qquad (2)$$

statistical parameters of correlation:
- correlation coefficient R = 0,989,
- $R^2 = 0{,}978$,
- Fisher test F = 123,18,
- standard deviation s = 3,771,
- standard error of estimation b = 1,572,
- significance level $\alpha = 0{,}05$.

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Because time of electromagnetic field action does not influence on equiaxed crystals zone content on cross-section of ingot, that is possilble to accept 15 and 30s for investigated ingots. In result of this assumption i.e. omit of value (t) in equation (2), was obtained following dependence:

$$SKR = -3{,}74\cdot f + 0{,}41\cdot B_i + 0{,}14\cdot M - 24{,}67 \qquad (3)$$

statistical parameters of correlation:
- correlation coefficient R = 0,988,
- $R^2 = 0{,}976$,
- Fisher test F = 169,45,
- standard deviation s = 3,524,
- standard error of estimation b = 1,547,
- significance level $\alpha = 0{,}05$.

Graphic interpretation of equation (3) for choose value of variables is shown in fig.5÷7.

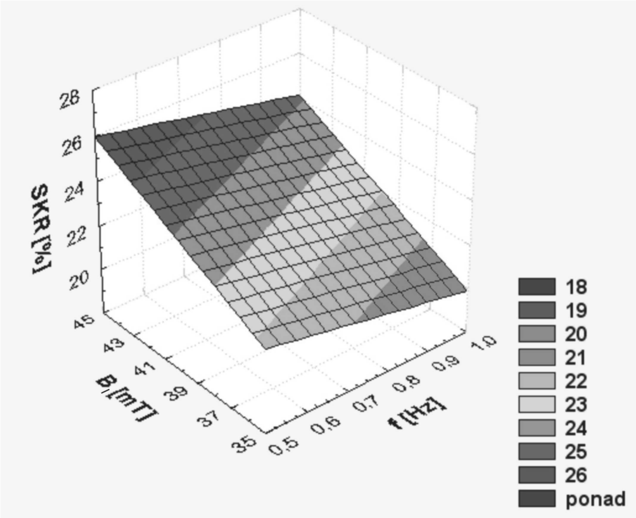

Fig. 5. Equiaxial crystal zone content on cross-section of ingot (SKR) in pulse frequency of electromagnetic field (f) and magnetic induction (B_i) function, for inoculant quantity M=(200Ti + 40B) ppm

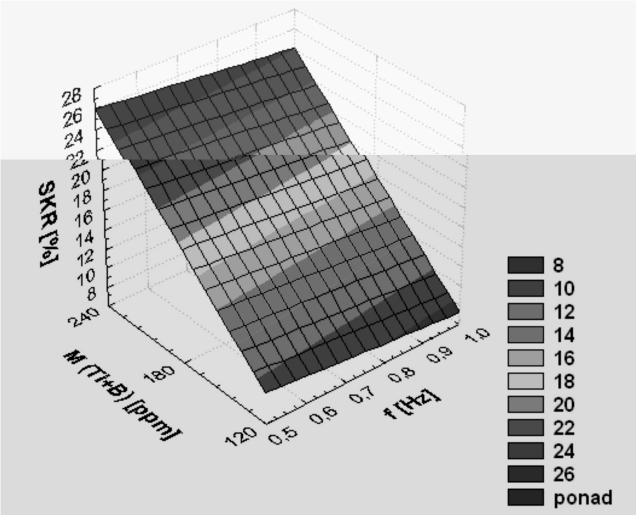

Fig. 6. Equiaxial crystal zone content on cross-section of ingot (SKR) in pulse frequency of electromagnetic field (f) and inoculant quantity (M) function, for magnetic induction B_i = 45 mT

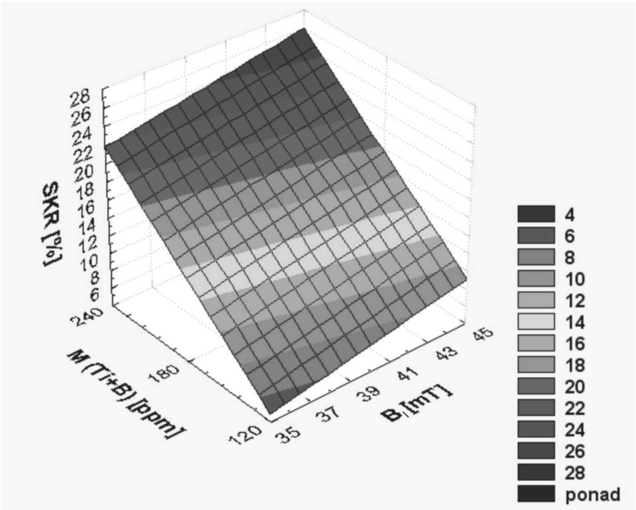

Fig. 7. Equiaxial crystal zone content on cross-section of ingot (SKR) in magnetic induction (B_i) and inoculant quantity (M) function, for pulse frequency of electromagnetic field f = 0,5 Hz

With decrease in pulse frequency of electromagnetic field and with increase in magnetic induction, increse in equiaxed crystals zone is observed. It results from high velocities that are attained in liquid metal and which lead to columnar crystals tearing occurring on crystallization front and additional crystal nucleuses formation.

Moreover, inoculant content (Ti+B) strongly influences on size reduction in aluminium structure.

Variable factors of casting with use of impulse reverse electromagnetical field in investigations with EN AW-Al99,5 were f = 0,5 Hz, B_i = 50 mT and t = 30 s.

In aim of measurements realization of size reduction in aluminium EN AW-Al99,5 structure were made metallografic examinations macro- and microscopic on Nikon light microscope with magnification from 100x to 600x. Surfaces of samples which were prepared for microstructure analysis were etched with use of solution of: 0,5ml HF, 99,5ml H_2O. Surfaces of samples which were prepared for macrostructure analysis were etched with use of solution of: 50g Cu, 400ml HCl, 300ml HNO_3 and 300ml H_2O. Value of size reduction in structure was represented by equiaxed crystals zone content (SKR) on cross-section of ingot of aluminium EN AW-Al 99,5 and average area of equiaxed crystal (PKR), were calculated by computer program Multi Scan Base v. 13.01 to processing and image analysis after macroscopic metallographic research.

X-ray examinations of investigated Al was made using DRON 2.0 diffractometr with Co anode. X-ray tube was supplied with the current I = 10mA under voltage of U = 25kV. Diffraction examinations were performed within the range of angles 2θ from 35° to 100°. The measurement step was 0,1° in lenght while the pulse counting time was 1s. Ivestigations of diffraction and thin foils were made on the JEM JEOL 2000 FX transmision electron microscope with system EDS at the accelerating voltage of 200kV. Thin foils for TEM investigations were electropolished with use of 20 ml $HClO_4$ and 80ml CH_3OH.

In aim of determination of thermal conditions of solidification was used mobile thermovision system of type Inframetrics 760. Accuracy of measurement of thermovision camera with helium cooling in Sterling circulation was 0,02°C in range -50°C ÷ 1500° C.

Electrical conductivity was measured with use of Thomsons bridge on sample with dimensions ϕ45 and 15mm length, which were cuting directly from ingots and on sample with dimension ϕ 10 and 400 mm length, which were casted in shell mould.

Deformability was estimated on basis of sheet press forming test with use of Erichsen method with according to norm PN-EN ISO 20482. Diameter of used stamp was 20 mm. Sheet of thickness 1 mm, were obtained as result of cold rolling of sample with double roll pass.

Full experimental plan with results of equiaxed crystals zone content (SKR) on cross-section of ingot of aluminium EN AW-Al 99,5 and average area of equiaxed crystal (PKR) measurements are shown in table 2.

3. Results and analysis

Selected results of metallographic research are presented on fig.8÷11. Aluminium EN AW-Al99,5 has columnar structure in initial state (fig.8). After inoculation with 25ppm Ti and 5ppm B, increase in size reduction of primary structure is observed (fig.9).

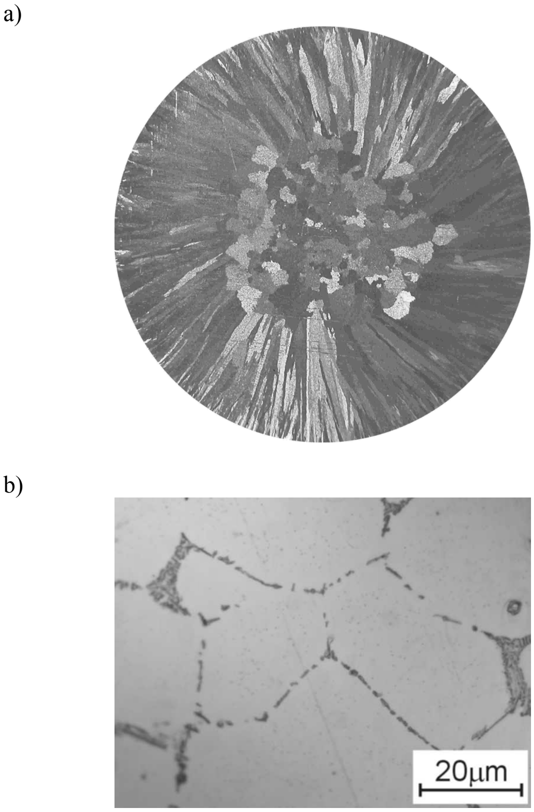

Fig. 8. Macro- (a) and microstructure of equiaxed crystals zone (b) of alumnium EN AW-Al99,5 without inoculation

Table 2.
Range and results of investigations

Sample number	Type of Al	f* [Hz]	B_i* [mT]	t* [s]	Assumption content Ti [ppm]	True content Ti [ppm]	Melting loss Ti [%]	Assumption content B [ppm]	True content B [ppm]	Melting loss B [%]	PKR [mm^2]	SKR [%]
1	99,5	–	–	–	–	–	–	–	–	–	26,32	24,34
2		–	–	–	25	53	–	5	4	20	3,83	51,44
3		0,5	50	30	–	–	–	–	–	–	0,68	32,05
4		0,5	50	30	25	55	–	5	4	20	0,57	45,24

* - IRPM – impulse reverse electromagnetic field

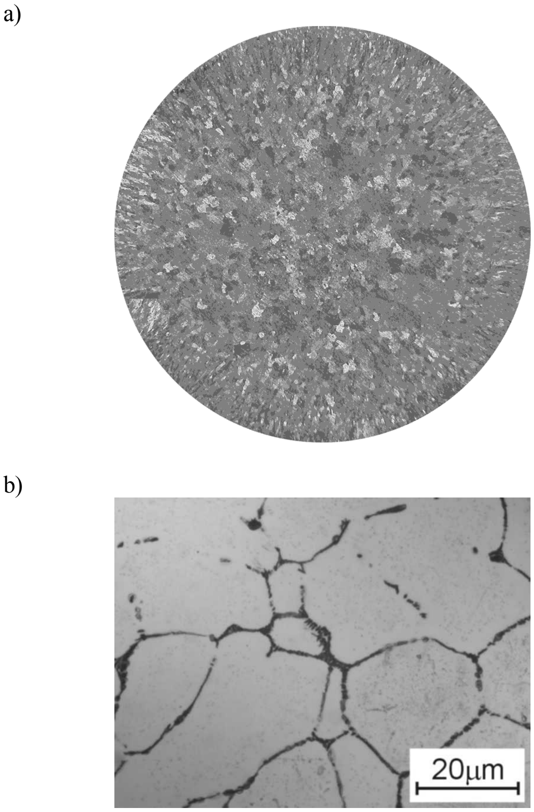

Fig. 9. Macro- (a) and microstructure of equiaxed crystals zone (b) of alumnium EN AW-Al99,5 after inoculation with 25ppm Ti + 5ppm B

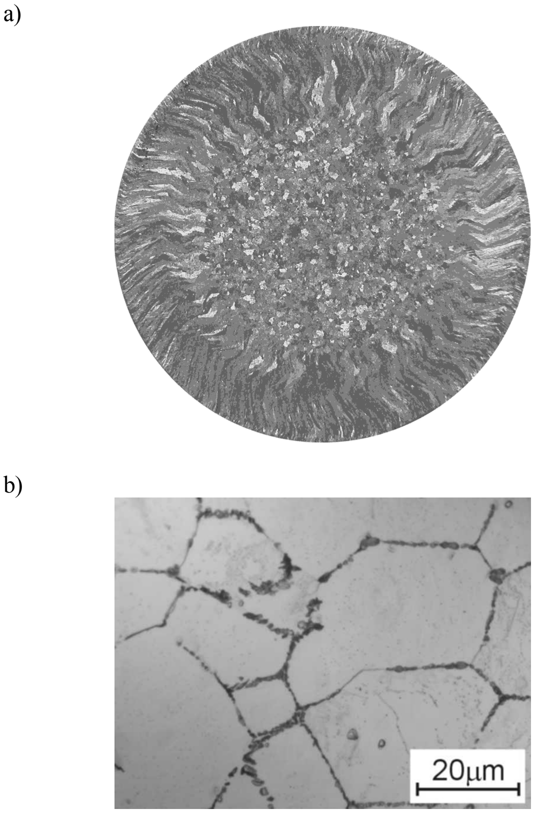

Fig. 10. Macro- (a) and microstructure of equiaxed crystals zone (b) of alumnium EN AW-Al99,5 after casting with influence of electromagnetic field

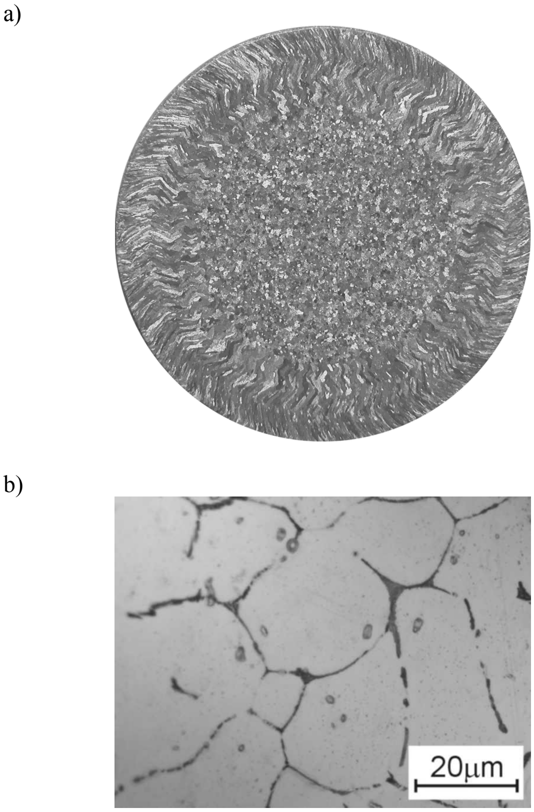

Fig. 11. Macro- (a) and microstructure of equiaxed crystals zone (b) of alumnium EN AW-Al99,5 which was casted with influence of electromagnetic field and with inoculation with 25ppm Ti + 5ppm B

Increase in size reduction of aluminium structure after inoculation with Ti and B, result from “washers” of type TiC or TiN (fig.12) and Al_3Ti (fig.13) to heterogeneous nucleation formation. These “washers” are high-melting and have analogy in crystal lattice with Al (tab.1). As result of X-ray analyses it was identified “washer” of type titanium carbide TiC or TiN. It is not possible to identify TiC or TiN because θ angle is the same for these phases. Moreover on basis of literature datas [1, 2, 4, 5] is possible to say, that important contribution in size reduction of Al structure have “washers” of type TiB, TiB_2, AlB_2.

Whereas, increase in size reduction in aluminium EN AW-Al99,5 structure, after casting with influence of impulse reverse electromagnetic field (fig.10) result from high velocities that are attained in liquid metal and which lead to columnar crystals tearing occurring on crystallization front and additional crystal nucleuses formation. Effect of mechanical erosion of crystallization front is strengthened by melting of dendrite as result of influences of temperture fluctuation, which result from large diversification of temperature in ingot, which was casted with influence of electromagnetic field. Diversification of temperature are presented on figure 14 as temperature fields distribution on free surface of ingots, which were casted without and with forced movement as result of influence of electromagnetic field. Temperature fields distribution were recorded with use of thermovision camera.

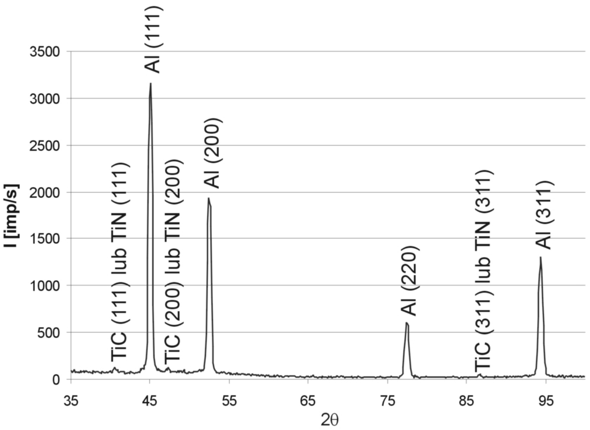

Fig. 12. X-ray diffraction of aluminium EN AW-Al 99,5 after inoculations Ti and B

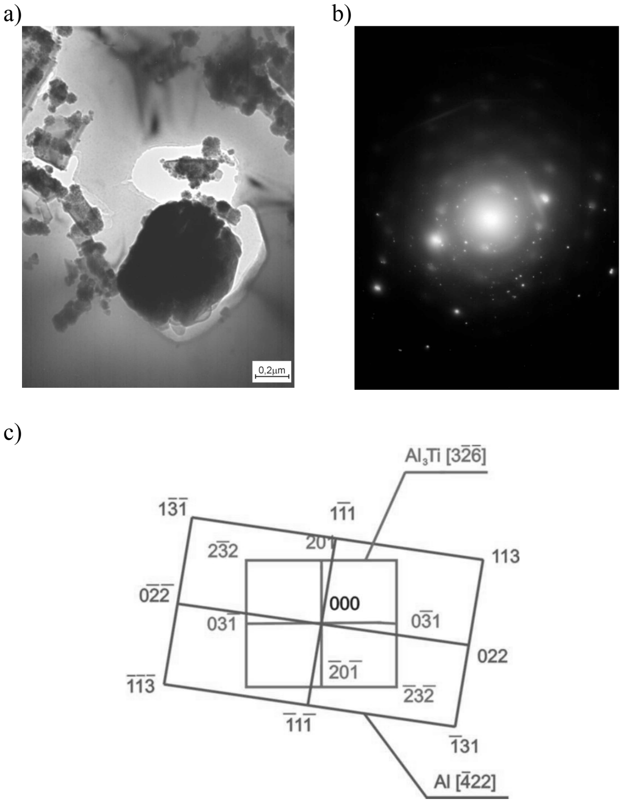

Fig. 13. a): Structure of thin foil from alumnium EN AW-Al99,5 after inoculation with (Ti + B) – 30000x , b): diffraction pattern from the area as in fig. a, c): solution of the diffraction pattern from fig. 13b

Moreover, common influence of impulse reverse electromagnetic field and inoculation with (Ti+B) (tab.2 and fig.11) result in larger equiaxed crystals zone content and smaller size of macrograin than in standard sample (fig.8) and comparable in sample which was casted only with inoculation (Ti+B) (tab.2 and fig.9) but it has larger size of macrograin than sample which was casted with influences of electromagnetic field and inoculation.

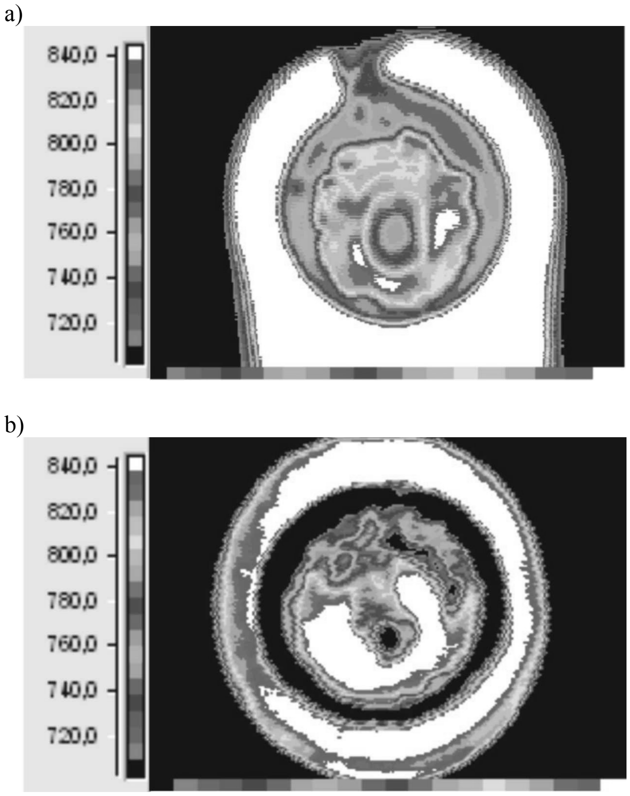

Fig. 14. Temperature distribution (pouring temperature 900°C after 5 s from moment of end of pouring) on free surface of solidify ingots which were casted:
a) without forced movement,
b) with forced movement as result of influence of electromagnetic field

In table 3 are presented results of electrical conductivity measurements of aluminium EN AW-Al99,5 which was casted with influences of electromagnetic field. Size reduction as result of use of electromagnetic field does not influence negatively on value of electrical conductivity.

In table 4 are presented results of electrical conductivity measurements of aluminium EN AW-Al99,5 after inoculation with (Ti + B). With increase in inoculant content decrease in electrical conductivity is observed (fig.15). Decrease in electrical conductivity result from influence of Ti (fig.4), which segregation on grain boundary of Al is observed (fig.16).

Table 3.
Results of electrical conductivity measurements for sample with area of cross-section 1589,625 mm^2 and length 15 mm

Type of inoculation	R [μΩ]	ρ [μΩ·m]	γ [MS/m]
lack	2,0	0,24	4,16
IRPM	2,0	0,24	4,16
IRPM and (25ppm Ti + 5ppm B)	2,0	0,24	4,16

where:
R – resistance (measured value),
ρ - resistivity,
γ - conductivity.

Table 4.
Results of electrical conductivity measurements for sample with area of cross-section 78,5 mm^2 and length 400 mm

Quantity of inoculant		R [μΩ]	ρ [μΩ·m]	γ [MS/m]
Ti [ppm]	B [ppm]			
0	0	1,494	0,0293	34,11
25	5	1,551	0,0304	32,85
50	10	1,624	0,0318	31,37
100	20	1,668	0,0327	30,53
150	30	1,702	0,0334	29,93
200	40	1,734	0,0340	29,38

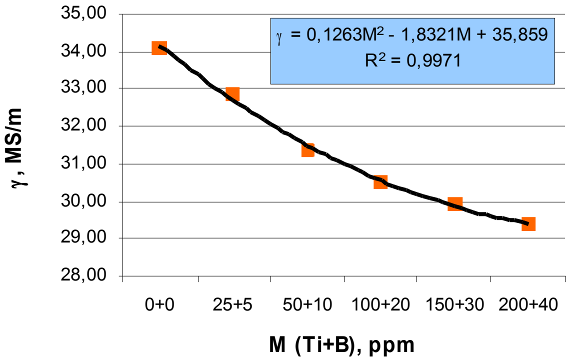

Fig. 15. Electrical conductivity (γ) of aluminum EN AW-Al99,5 in quantity of inoculants M (Ti + B) function

Value of size reduction in ingots primary structure strongly influences on deformability of sheet. After rolling, delamination of external layers occur in ingots, which were casted without inoculation or only with influence of (Ti + B) inoculation (fig.17a, b and c) or only with influence of electromagnetic field. The best quality has sheet from ingot which was casted with common influence of (Ti+B) inoculation and electromagnetic field (fig.17d). It is confirmed by press forming test. With increase in size reduction in primary structure increase in Erichsen number (IE) i.e. depth of entry of stamp into sheet till to appearance of crack (value in mm), is observed (fig.18).

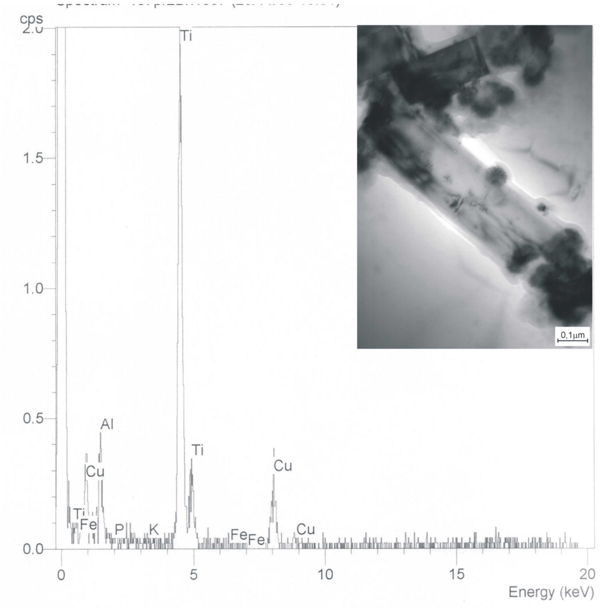

Fig. 16. Segregation of Ti on grain boundary of Al

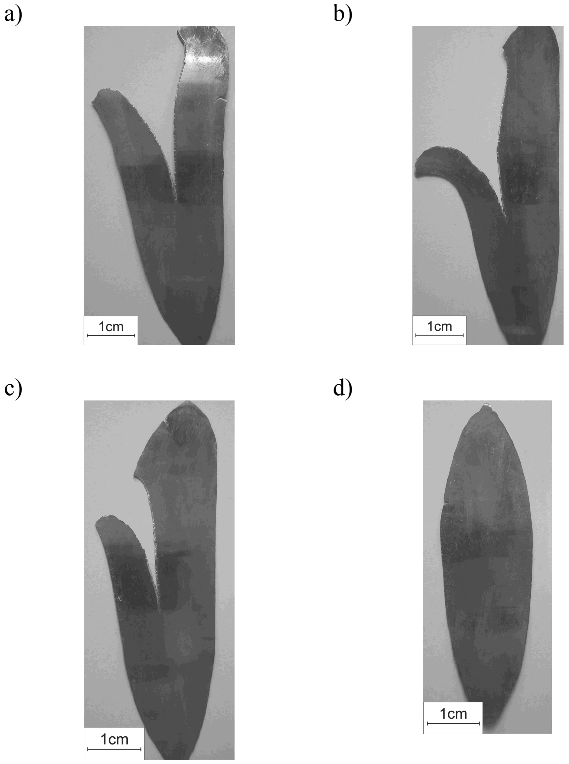

Fig. 17. View after rolling of sample of aluminium EN AW-Al99,5: a) without inoculation, b) after inoculation with 25ppm Ti + 5ppm B, c) after casting with influence of electromagnetic field, d) which was casted with influence of electromagnetic field and with 25ppm Ti + 5ppm B inoculation

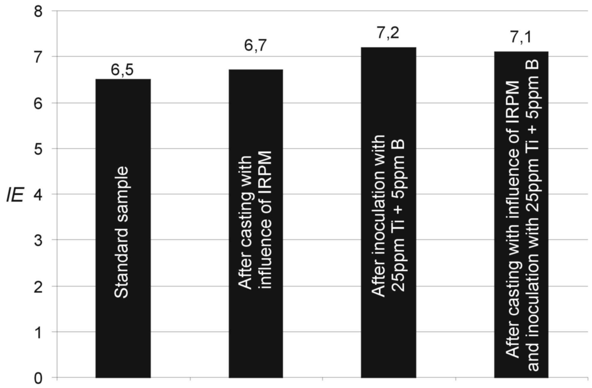

Fig. 18. Results of press forming test

4. Summary and conclusion

Based on conducted studies it was affirmed, that common influence of both presented methods of shaping of aluminium EN AW-Al99,5 primary structure i.e. influence of internal factor as inoculation with small amount of (Ti + B) and influence of external factor as electromagnetic field, lead to larger size reduction in structure than influence of these factors separately. On one hand this results from “washers” to aluminium heterogeneous nucleation formation – internal factor. On the other hand this results from mechanical and thermal erosion of crystallization front – external factor. However very important is influence of forced liquid metal movement on concentration distribution of inoculants and impurities on cross-section of ingot, what results from [13] too. Increase in grains density after increase in “washers” density in central area of ingot result from forced liquid metal movement, which leads to convection transport of Cu and Ti from crystallization front to inside of metal bath. Proof of this thesist are results of determination of Cu and Ti concentration in near surface and central areas of investigated ingots with use of emission optical spectrometry. In aluminium EN AW-Al99,5 ingot which was casted with influence of electromagnetic field and with (Ti + B) inoculation, increse in concentration of Cu and Ti in central area is observed (fig.19a). Whereas in aluminium EN AW-Al99,5, which was casted only with (Ti + B) inoculation, concentration of Cu and Ti in near surface and central areas of ingot is similar (fig.19b). Second testifying proof about convection transport of Cu and Ti from crystallization front to inside of liquid metal is analysis of macrostructure of investigated ingots and counting of all macrograins in equiaxed crystals zone. Macrostructure of aluminium EN AW-Al99,5 ingot which was casted with common influence of electromagnetic field and with (Ti+B) inoculation has smaller equiaxed crystals zone than in ingot which was casted only with influence of external factor. But this first ingot has smaller size of macrograin in equiaxed crystals zone than in ingot which was casted only with (Ti+B) inoculation.

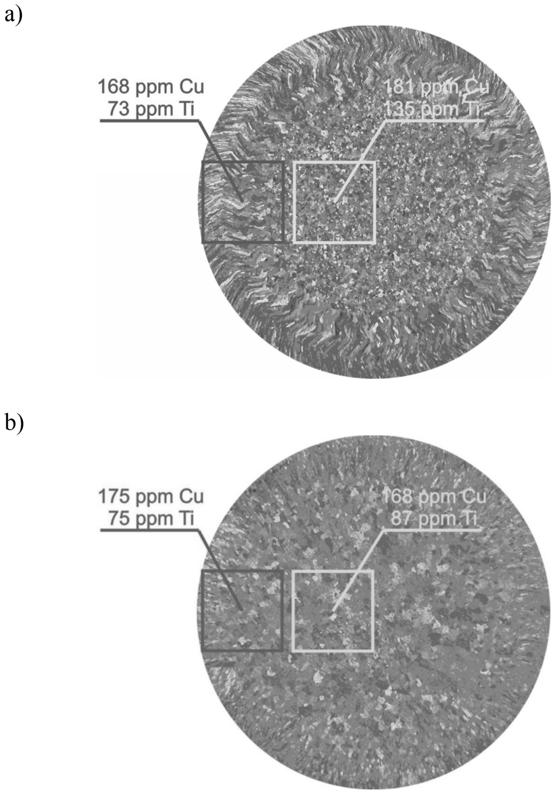

Fig. 19. Concentration of Cu and Ti in near surface and central areas of investigated ingots of aluminium EN AW-Al99,5:
a) which were casted with influence of electromagnetic field and with inoculation with 25ppm Ti + 5ppm B, b) after inoculation with 25ppm Ti + 5ppm B

Based on conducted calculations of value of macrograins in equiaxed crystals zone (fig.20) following dependence have been formulated:

$$n_{wz} > n_w + n_z \quad (4)$$

where:
n_{wz} – value of macrograins in equiaxed crystals zone of ingot which was casted with common influence of internal and external factors,
n_w – value of macrograins in equiaxed crystals zone of ingot which was casted only with influence external factor,
n_z – value of macrograins in equiaxed crystals zone of ingot which was casted only with influence internal factor.

About 60% macrograins in equiaxed crystals zone result from mechanical and thermal erosion of crystallization front as effect of forced liquid aluminium movement by use of electromagnetic field. Wheras residual 40% macrograins in this zone result from “washers” to aluminium heterogeneous nucleation formation, which as effect of liquide metal movement with high velocities are concentrated in central area of ingot.

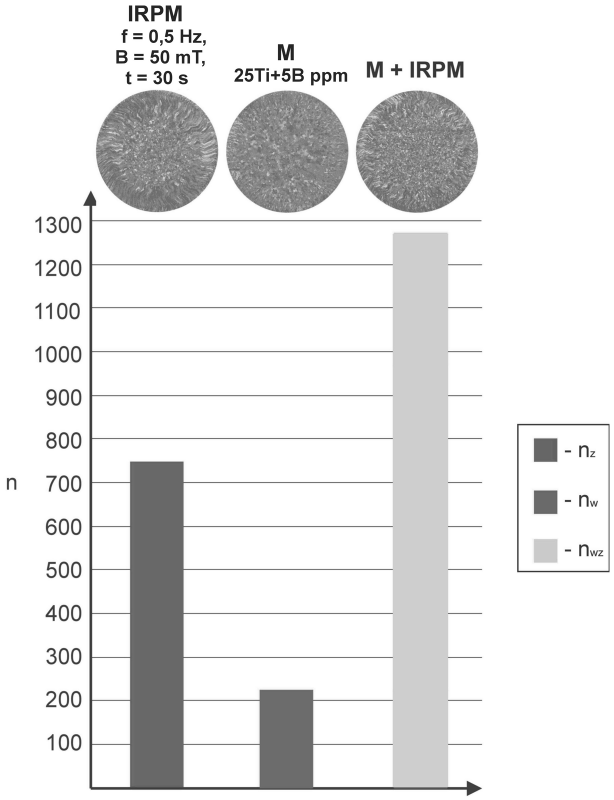

Fig. 20. Value of macrograins in equiaxed crystals zone of investigated ingots of aluminium EN AW-Al99,5

Summarize, based on conducted studies following conclusions have been formulated:

1. Even inoculation with small amount of (Ti + B) strongly increase size reduction in pure aluminium structure. It result from reactions, which proceed between modifying elements and modified metal or impurities of charge. These reactions lead to formation of active “washers” to heterogeneous nucleation of aluminium as high-melting small particles of type Al_3Ti, AlB_2, TiB_2, TiB and TiC or TiN, which have analogy in crystal lattice with Al.
2. Influence of electromagnetic field on solidification process of pure aluminium ingots creating mechanical and thermal erosion of crystallization front, which increase deformability of sheet because aided size reduction of their structure, which creates mainly by introduction of small amount of inoculant (Ti + B) – less than obligatory standard PN-EN 573-3 concerning about aluminium purity and no lowering indeed electrical conductivity of aluminium.

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