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# EFFECT OF SECOND PHASE PARTICLES TOPOLOGY ON THE ONSET TEMPERATURE OF ABNORMAL GRAIN GROWTH IN Fe - 3%Si STEELS

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The relations between regimes of dynamic annealing, state of secondary particles system and the onset temperature of abnormal grain growth are investigated. Two distinguish types of Fe-3%Si grain-oriented steels, after one and two stage cold rolling, were studied. The second phase particles remain unaffected in first type of steel during the heat treatment. Vice versa, the increased density of second phases was observed after annealing in the second type of the investigated materials. It is shown that start/onset of abnormal grain growth strongly depends on both volume fraction of second phase particles and annealing temperature. Texture and magnetic properties of the investigated samples are investigated within the current study.

*Key words:* second phase particles, abnormal grain growth, coercive force, electrical steel

**Učinak druge faze topologije čestica pri početnoj temperaturi abnormalnog rasta zrna kod Fe – 3%Si čelika.** Istraživani su odnosi između režima dinamičkog žarenja, stanja sustava sekundarnih čestica i početne temperature abnormalnog rasta zrna. Analizirane su dvije različite vrste Fe-3%Si zrnato usmjerenih čelika, nakon prve i druge faze hladnog valjanja. Čestice druge faze ostaju nepromijenjene kod prve vrste čelika tijekom toplinske obrade. Obrnuto, uočena je povećana gustoća u drugoj fazi nakon žarenja kod druge vrste ispitanih materijala. Prikazano je da početak abnormalnog rasta zrna uvelike ovisi i o opsegu čestica druge faze i o temperaturi žarenja. Ova analiza istražuje teksturu i magnetska svojstva ispitanih uzoraka.

*Ključne riječi:* čestice druge faze, abnormalan rast zrna, koercitivna sila, elektro-čelik

## INTRODUCTION

Electrical steels are undoubtedly the most important soft magnetic materials in use today. Applications vary in quantities from the few ounces used in small relays or pulse transformers to tons used in generators, motors, and transformers.

There are two principal types of electrical steels: grain oriented and non-oriented steels. The grain oriented (GO) electrotechnical steel sheets are widely used as core material in transformers, large rotating machines and other electrical equipments. For high magnetic induction and low core losses during premagnetization, this type of steel should have a sharp so-called Goss texture {110}<001> in the final state. This kind of texture provides favorable magnetic properties of the steel in the rolling direction (RD) [1]. Other type of electrical steels is non-oriented steel used in electric machines to transform mechanical energy into electrical energy and vice versa. The {100} orientation plane parallel to surface is a special feature of this kind of electrical steel which provide approximately the same magnetic properties in all directions of the sheet plane [2].

The Goss texture (110)[001] is a result of conventional technological rout applied for Fe-3%Si steels. Two essential requirements have to be fulfilled to foster the process of secondary grain growth:

1) the grain growth inhibitors such as MnS, AlN or MnS+AlN and etc., must be added;
2) the completely recrystallized microstructure with sufficient intensity of Goss texture component has to be obtained before secondary recrystallization [1,3].

The increase in the number of precipitates, change in grain size as well as texture state are account for magnetic properties change in material. The magnetic properties are characterized by coercive force $H_C$. This physical value drastically decreases with commencement of secondary recrystallization because of significant changes in microstructure and texture. The secondary recrystallization starts at particular temperature called an onset temperature of the secondary recrystallization - $T_{SR}$.

The intention of current study is to investigate the influence of inhibition system on the $T_{SR}$ in Fe-3%Si grain-oriented electrical steels.

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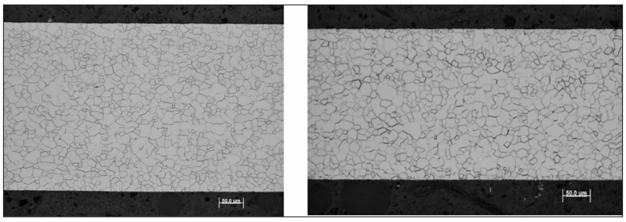
Table 1. **Chemical composition of the investigated steels, wt.%.**

| Sampl. | C | Mn | Si | Cu | P | S | Al |
|---|---|---|---|---|---|---|---|
| P | 0,032 | 0,235 | 3,031 | 0,5 | 0,01 | 0,0062 | 0,0165 |
| C | 0,039 | 0,16 | 2,84 | 0,52 | 0,01 | 0,009 | 0,015 |



**a)** **b)**

**Figure 1.** Primary recrystallized microstructures of investigated steels annealed at 850°C/5 min in wet atmosphere of cracked ammonium: a) steel C, b) steel P.

## EXPERIMENTAL PROCEDURE

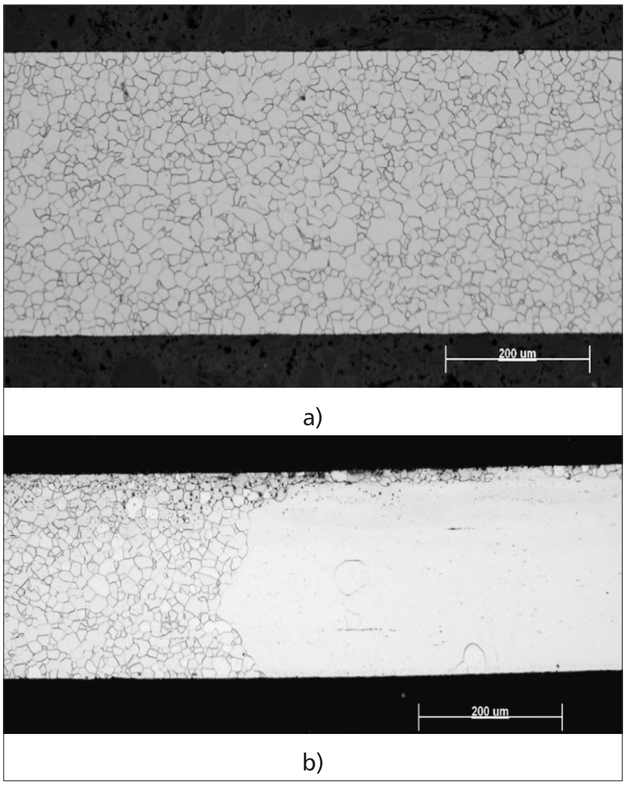
Grain-oriented electrical steels of different chemical composition were used in the experimental procedure. The samples were taken from industrial line after cold rolling process. The investigated steels P and C were after one-stage and two-stage cold rolling processes respectively. The respective total reduction for C and P steels are 87% and 90%. The chemical compositions of the investigated steels are presented in Table 1.

Generally, in order to obtain desired magnetic properties, cold rolled GO electrical steels are subjected to long-term annealing treatments in both pure $H_2$ and $N_2$-$H_2$ gas mixture.

The heat treatment was realized by furnace "Nabertherm" with an electronic control system C19/S19. The dynamic heat treatment conditions were applied in order to achieve the onset of secondary recrystallization in the investigated materials within short time in decarburizing atmosphere. The samples were heated up to elevated temperature with heating rate ~ 10°C/s and then cooled to room temperature. The annealing time was varied within the range of $t$=5-10min. The annealing temperature was changed in the range of $T$=850-1150°C. The moisture of the annealing atmospheres was varied within +20°C - +30°C.

The coercive force of the steel was measured by standard coercive-meter KPS-lc using samples of 3cm × 1cm.

The microstructures of the specimens were examined in the longitudinal cross-section and in the normal direction plane using light microscopy. The carbon extraction replicas were prepared in order to isolate precipitates from the metallic matrix. Second phase particles distribution was investigated by TEM in cold rolled and heat treated samples. The technique described[4] was applied to investigate both particle size and volume fraction.




a)

b)

**Figure 2.** Microstructures of investigated steels heat treated at 1000°C/5 min in wet atmosphere of cracked ammonium: a) Strong inhibition of primary recrystallized matrix in steel *C*; b) commencement of abnormal grain growth in steel *P*.

The etch pit patterns method was utilized in order to investigate the orientation of grains in heat treated samples according to procedure described in [5]. Both, the light microscopy and the SEM technique were applied to achieve a high quality resolution of etch pit patterns in the investigated samples.

The indentation of the second phase particles in iron matrix was realized by EDX analyzer INCAx-sight model 7557.

## RESULTS AND DISCUTION

Figures 1 – 3 present microstructures development in heat treated sheets *P* and *C* for 10 min at various temperatures in atmosphere of cracked ammonia. As follows from the figures, progress in grain size is proportional to value of thermal exposition within the range of 850°C – 1050°C. The microstructure is fully recrystallized after 10min. annealing at 850°C with mean grain size about 30-40mm in both steel sheets (see Figures.1.a, b). Thermal exposition at respective temperature leads to start of

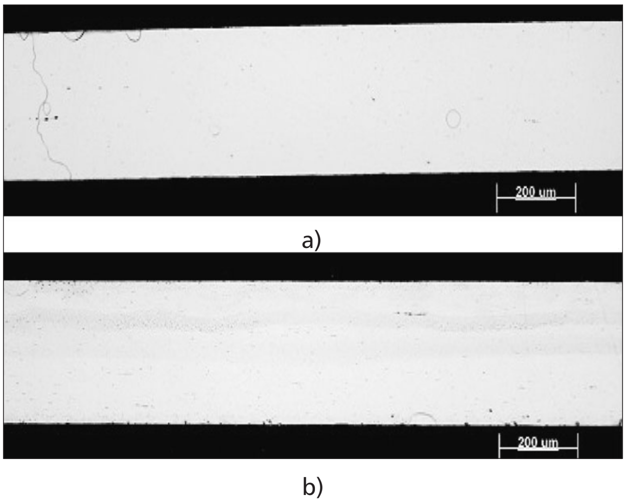



**Figure 3.** Abnormal grain growth in C (a) and P (b) steels. The materials are treated at 1050°C for 5 min in wet atmosphere of cracked ammonium.

abnormal grain growth (AGG) in each material. For instance, the beginning of AGG in steel P treated at 1000°C/10min is presented in Figure 2.b. Vice versa, no microstructural changes were observed in steel C treated under the same circumstances (see Figure 2.a). As follows from Figure 2, the heat treatment conditions provide strong inhibition of primary recrystallized matrix in steel C and lead to abnormal growth of Goss grains, consuming the inhibited surrounding matrix, in material *P*.

Huge grained microstructure consisting of abnormal ferrite grains was observed in the investigated steels P and C after annealing at 1050°C (see Figures 3. a, b). In both materials, the applied heat treatment parameters provide appropriative selection conditions for abnormal growth of grains with Goss orientation and mean size ~1-1.5mm. Increasing the annealing temperature breaks the selectivity conditions for AGG which in turn become apparent in non-uniform grains distribution; this phenomenon is noted in Figure 4.

The electron scanning micrographs (SEM) of etch pit figures obtained from heat treated steels C and P are presented in Figures 5 a and b respectively. The investigated samples where treated at 1050°C for 5 min in wet atmosphere of cracked ammonium. As one can see, the annealing circumstances are appropriative for AGG process with high density of cube on the edge texture component.

The etch pit figures make possible to predict the orientation of certain grain by the way already described [5]. Examples of Euler angles and Miller's indexes calculated from etch pit pattern are presented in Table 2.

Table 2. **Calculated Euler angles and Miller's indexes for marked grains in Fig. 5.**

| | | $\varphi 1$ | $\Phi$ | $\varphi 2$ | *h* | *k* | *l* | *u* | *v* | *w* |
|---|---|---|---|---|---|---|---|---|---|---|
| **C** | 1 | 85 | 90 | 44 | 1 | 1 | 0 | 0 | 0 | 1 |
| | 2 | 90 | 90 | 39 | 3 | 4 | 0 | 0 | 0 | 1 |
| **P** | 3 | 90 | 90 | 45 | 1 | 1 | 0 | 0 | 0 | 1 |
| | 4 | 87 | 90 | 41 | 5 | 6 | 0 | 0 | 0 | 1 |

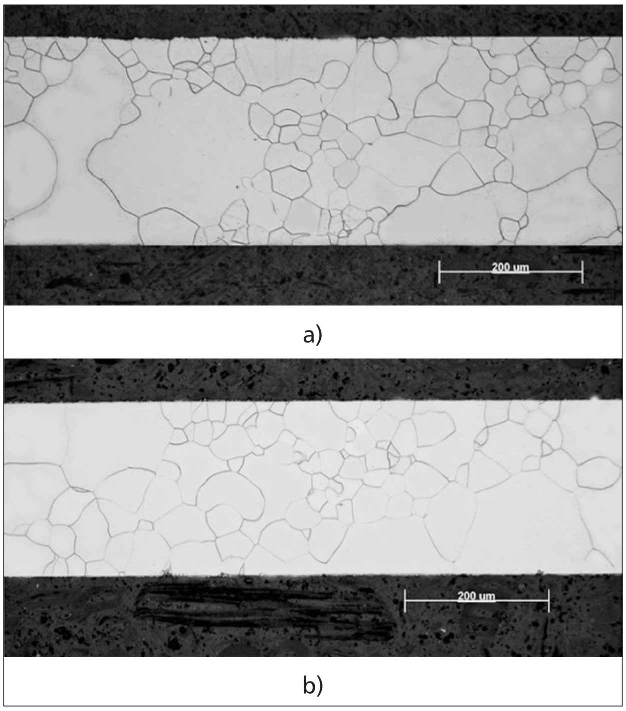



**Figure 4.** Microstructures of heat treated steels at 1100°C/5 min: a) steel C; b) steel P.

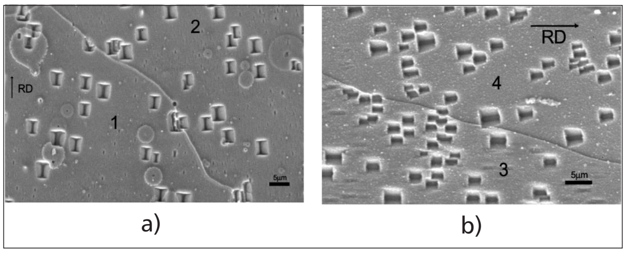



**Figure 5.** SEM micrographs of etch pit figures: a) steel C and b) steel P. The samples were annealed at 1050°C for 5 min in wet atmosphere of cracked ammonium.

Results of orientation measurements for ~ 100 grains obtained by this way are shown in Figure 6. The more representative sections were subjected to investigations. The diagram in Fig. 6 shows the intensity for major orientations i.e Goss, Cube and {111}[uvw] deformation texture components. As follows from the presented dia-

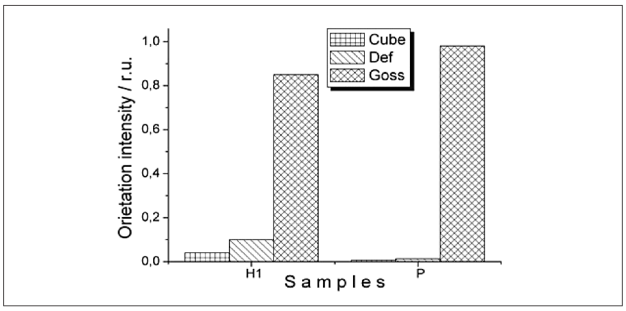



**Figure 6.** Orientations intensity in the investigated samples C and P annealed at temperature 1050°C. The heat treatment accounts for AGG and diffusion controlled grain growth in C and P samples respectively.

grams, the "cube on edge" components are predominant in the both investigated steels after annealing at 1050°C. The developed grains having size of some millimeters with Goss/near Goss texture orientations were obtained in steel P (~98% of the total amount of investigated grains pose Goss orientation). A strong near Goss texture component was observed in material C too with volume fraction ~ 85%. The surrounded "parasitic" grains have either rotated cube texture component or some component of {111} fiber (deformation texture). The average size of "parasitic" grains was approximately 20-50 $\mu$m.

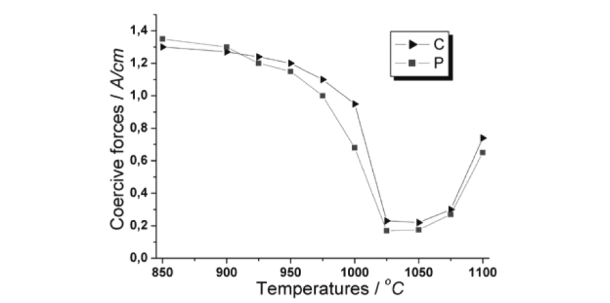


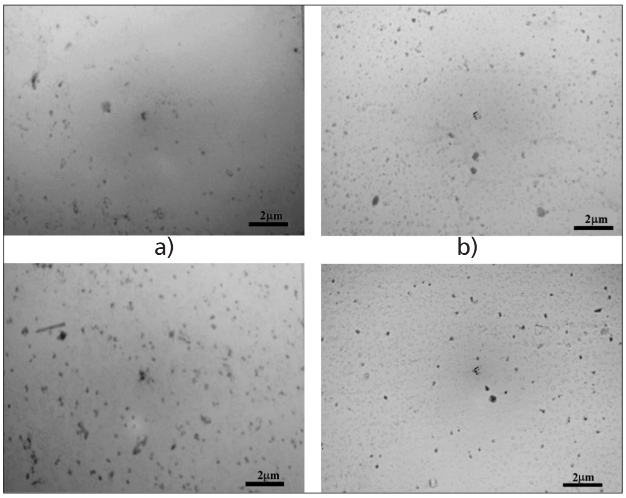

**Figure 7**. Dependence of coercive force on annealing temperatures in C and P steels treated for 5 min in wet atmosphere of cracked ammonium.

Magnetic properties are very sensitive to change in number of precipitates, grain size as well as to texture state of the investigated materials. Figure 7 presents the change in coercive force $H_C$ as function of annealing temperature. The rapid decrease in $H_C$ value observed in sample P after annealing for 10 min at temperature 1000°C. The differences in $H_C$ values are observed because of significant change in grain size (bimodal grain size distribution is observed) due to beginning the AGG (see. Figure 2,b). Whereas the similar yielding in coercive forces is observed for material C annealed at 1025°C, see Figure 7. This phenomenon confirms the difference in onset temperatures $T_{SR}$ for steels C and P. The plateaus behavior in $H_C$ curve within temperature range of T=1025-1050°C for both P and C samples would lead one to conclude that no microstructural changes occur. The microstructure development in Figures 3 a and b support this suggestion. In both materials, the abnormal grain growth is completed within above mentioned temperature range.

Dispersion of second phase particles in heat treated steels C and P are presented in Figure 8. As one can see, the intensity of second phases in steel C is higher than in steel P. Hence, the difference in $T_{SR}$ can be attributed to difference in volume fraction of precipitates $F_V$ for investigated C and P materials. The carbon extraction replicas in Figures 8 b and d confirm that dynamic heat treatment condition does not influence neither morphology nor distribution parameters of the second phases in the investigated steels. The volume fraction of second phases is drastically increased after annealing in material C from 0,135% (for untreated materials) to 0,28% (for annealed one), viz. Figure 8 a and c. As one can conclude from the above mentioned facts, the precipitation and coarsening of second phases take place during heat treatment (1050°C/5 min) of steel C. The differences in the inhibition systems can be attributed to difference in thermo-mechanical history. It means that steel P was subjected to pre-annealing treatment before cold rolling process. Contrarily, material C was immediately subjected to two stage cold rolling conditions after hot rolling treatment. Hence, pre-annealing process in P material has led to second phase particles coarsening before cold rolling reduction, the similar phenomena was observed [6].

Qualitative identification of second phases was performed by EDX analyses which revealed that second phase particles are consisted of cooper, manganese and aluminum as shown in Figure 9. It is well known that Cu and Mn elements in 3% Fe steels are usually coupled with sulphur and Al with nitrogen [1]. Hence, the qualitative analyses shown in Figure 9 lead one to suggestion that created second phase particles are CuS, MnS and AlN. These statements are additionally supported by TEM diffraction patterns obtained from carbon extraction replicas.

Both theoretical and experimental estimation of volume fraction of the second phase particles was conducted by way presented [4, 7]. Theoretical method of volume fraction estimation is based on Zener equation. As was already pointed out by Zener, the dispersed particles could slow down the progress of grain boundary migration in metals [7]. This assumption has led to a famous formula derived by Zener:




c) d)

**Figure 8.** Distribution of second phases in the investigated materials: a) cold rolled steel C; b) cold rolled steel P; c) annealed steel C at 1000°C/5min; d) annealed steel P at 1000°C/5min in (all samples were treated in wet atmosphere of cracked ammonium).

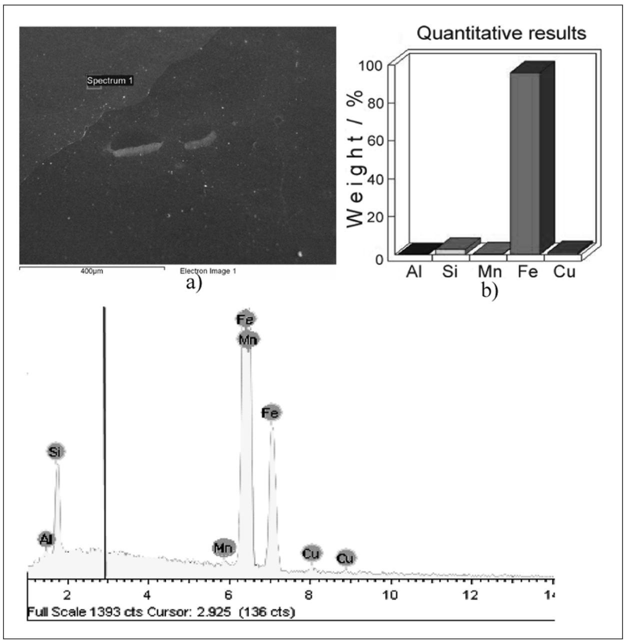



**Figure 9.** EDX analysis of second phase particles: a) SEM image of sample P; b) qualitative result of EDX analysis of the marked particle in figure a; b) quantitative result of EDX analysis.

$$R_C = 4r / 3f \quad (1)$$

where $R_C$ is the critical grain size after which the normal grain growth is not possible (Zener limit), $r$ the radius of pining particles and $f$ is the volume fraction of particles.

Estimating the volume fraction by equation 1 can be a satisfactory revising for the experimental data. The equation 1 is applicable for the present study, considering normal grain growth between the primary and secondary recrystallization processes. The mean grain as well as particle sizes can be estimated with sufficient accuracy by applied SEM technique.

Therefore, taking into account the obtained value of the mean grain and particle size, the following values of volume fraction were obtained 0,2% and 0,3% for heat treated P and C materials respectively. As one can see the calculated results are in good agreement with experimental ones.

## CONCLUSIONS

In three-phase particle containing Fe-Si steels, the onset temperature for abnormal grain growth is influenced by the volume fraction of the precipitates. The critical dependence of volume fraction of the second phases on both thermo-mechanical processing and chemical composition of steel is confirmed.

High temperature annealing within range of 1025-1050°C under dynamic heat treatment conditions leads to abnormal grain growth in Fe-3%Si steels. The orientation estimation by etch pit method sows that sharp Goss texture can be developed by this way.

Applying higher temperature than onset one ($T_{SR}$) breaks the selectivity condition for abnormal grain growth resulting in highly inhomogeneous microstructure.

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