UDK 669.14.018.583
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 57(6)681(2023)

SUSTAINABLE AND STRATEGIC SOFT-MAGNETIC Fe-Si-Al ALLOYS PRODUCED BY SECONDARY METALLURGY

TRAJNOSTNE IN STRATEŠKE MEHKOMAGNETNE Fe-Si-Al ZLITINE, IZDELANE S POSTOPKI SEKUNDARNE METALURGIJE

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Prejem rokopisa – received: 2023-12-11; sprejem za objavo – accepted for publication: 2023-12-15

doi:10.17222/mit.2023.1072

In Slovenia, Fe-Si-Al alloys for non-oriented, silicon-steel sheets are designed and manufactured in a sustainable manner. Ferrous scrap is recycled, and, therefore, CO_2 emissions are greatly reduced. However, sustainable technologies based on secondary metallurgy have many limitations. Impurities that cannot be effectively removed from the steel melt increase the complexity of the material's behavior during processing and use. One of the most contaminating elements is copper (Cu). In this review, the focus is on phenomena related to the Cu impurity during the specific steps of the metallurgical processing of selected Fe-Si-Al alloys. The identified challenges concerning the efficiency of some technological phases related to the presence of Cu in Fe-Si-Al, non-oriented electrical steels might motivate further (inter)disciplinary research, basic or applied. In order to follow the set goals of the EU and achieve climate neutrality by 2050, silicon electrical steels produced with sustainable circular-economy approaches must be recognized as a strategic material for the EU.

Keywords: silicon steel, secondary metallurgy, impurities, copper, magnetic losses, EU Green Deal

V Sloveniji so zlitine Fe-Si-Al za neorientirane silicijeve jeklene pločevine zasnovane in izdelane na trajnosten način. V postopkih sekundarne metalurgije recikliramo jekleni odpad, s čimer se znatno zmanjšajo emisije CO_2. Vendar pa imajo trajnostne tehnologije, ki temeljijo na sekundarni metalurgiji, številne omejitve. Nečistoče, ki jih ni mogoče učinkovito odstraniti iz jeklene taline, znatno povečajo kompleksnost obnašanja materiala med predelavo v končni proizvod in v njegovi rabi. Ena najbolj obremenilnih nečistoč je baker (Cu). V pregledu je poseben poudarek namenjen pojavom, ki so povezani z nečistočo bakrom in učinkovitostjo posameznih proizvodnih faz v izdelavi Fe-Si-Al neorientiranih elektropločevin. Identificirani izzivi, povezani s prisotnostjo bakra v zlitinah Fe-Si-Al lahko motivirajo nadaljnje (inter)disciplinarne raziskave, bodisi temeljne bodisi aplikativne. Da bi sledili zastavljenim ciljem EU in dosegli podnebno nevtralnost EU do leta 2050, bi morala biti silicijeva elektro jekla, proizvedena s pristopi trajnostnega krožnega gospodarstva, prepoznana kot strateški material.

Ključne besede: silicijevo jeklo, sekundarna metalurgija, nečistoče, baker, magnetne izgube, Evropski zeleni dogovor

1 INTRODUCTION

The synthesis and processing of metallic materials is the largest single source of greenhouse-gas emissions in the world.[1] Metals production accounts for 40 % of all industrial greenhouse-gas emissions, 10% of global energy consumption, 3.2 billion tonnes of minerals mined, and several billion tonnes of by-products every year. Therefore, metals must become more sustainable.[1] The European Green Deal is the EU's new growth strategy, aiming to transform the EU into a fairer and more prosperous society, with a modern, resource-efficient and competitive economy, with no net emissions of greenhouse gases by mid-century.[2] Specific priority topics of the EU Green Deal have been defined, i.e., Clean Energy, Circular Economy, Efficient Renovations, Sustainable Mobility, Sustainable Food, Preserving Biodiversity, all of which contribute to climate action and zero-pollution ambitions. However, achieving a climate-neutral and circular economy requires the full mobilisation of industry. All industrial value chains will have a key role to play. To meet the EU's energy and climate targets for 2030, EU countries need to establish integrated national energy and climate plans that outline how the EU countries intend to address five areas: energy efficiency, renewables, greenhouse-gas-emission reductions, interconnections, and research and innovation. Therefore, the whole industry value chains along with the research and innovation sector will have a key role to play.[2] Becoming the world's first climate-neutral continent by 2050 is a once-in-a-lifetime opportunity to modernise the EU's economy and society towards a sustainable future.

The purpose of the article is to present the role of the soft-magnetic material silicon steel and give an overview of the state of the key challenges related to the material produced via circular-economy routes. Since silicon steels are designed and manufactured in a sustainable manner also in Slovenia, a special focus is dedicated to the phenomena related to Cu impurity during the specific steps of the metallurgical processing of selected Fe-Si-Al alloys.

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2 SOFT-MAGNETIC SILICON STEELS

Soft-ferromagnetic materials are alloys that magnetise and demagnetise with low hysteresis losses.[3,4] A magnetic material is considered "soft" when its coercive field strength is of the order of or lower than the earth's magnetic field (about 40 A/m). A soft-magnetic material can be employed as an efficient flux multiplier in a large variety of devices, including transformers, generators, and motors, to be used in the generation, distribution, and conversion of electrical energy, and a wide array of apparatus, from household appliances to scientific equipment.[5] Silicon steels are the most important soft-magnetic material occupying most of the soft-magnetic market. Almost 97 % of soft-magnetic materials produced today are silicon electrical steels made of Fe-Si or Fe-Si-Al alloys. Over 12 million tons are produced annually, around 80 % of which are non-oriented (NO) grades, the remainder being grain-oriented (GO) silicon steels. In electrical machines NO grades are used almost exclusively.[6,7] The development of new classes of high-specific-power electrical machines and drives is constantly required.[8]

In Slovenia, non-oriented electrical steels are produced by SIJ Acroni d.o.o., Jesenice.[9] Their SIWATT brand includes non-oriented electrical steel that is essential for the economical production, conversion, distribution and use of electricity. Various grades of Fe-Si-Al electrical steels under SIWATT are characterised by magnetising ability, high magnetic permeability and low losses and are built into the magnetic cores of electric motors and generators.

For the optimal design of the soft-magnetic material ex-ante material's characterization is necessary, including a definition of the chemical composition of the steel along with extensive thermodynamic and microstructure modellings, and magnetic-loss prediction. Ex-post chemical, mechanical, metallographic and magnetic characterization analyses are dedicated to the optimization of the material's properties. These generally include analyses of the steel's chemical composition and purity, mechanical tests, microstructure analysis and grain-size measurements, the texture characterization and the magnetic-loss measurements. Many of their properties derive from an interplay of processing, microstructure, and chemistry, on scales that reach from manufacturing dimensions. This turns research on magnetic materials not only a multi-physics and multi-scale problem, but also requires close collaboration between characterization, processing, and theory.[3]

3 THE METALURGY OF SILICON STEELS PRODUCED USING CIRCULAR-ECONOMY ROUTES

The accumulation of impurities in the recycling of steel impacts the quality of secondary steel. Understanding impurity levels is crucial in the context of the proliferation of circular-economy policies, expected high recycling rates, and the growth of scrap consumption.[10] The complexity of the materials' behaviour, and consequently, the metallurgical processing of silicon steels for magnetic applications increases with the complexity of steels' chemical composition. Si being a ferrite stabilizer increases the ferrite phase fraction of Fe.[11] The addition of Si to iron brings notable changes in the physical, mechanical and magnetic properties of the Fe. The most notable effect regards the electrical resistivity. Alloying with Al instead of Si leads to similar physical and structural effects, while not causing material embrittlement. However, Al is very reactive and prone to oxide formation, but it brings an increase in magnetostriction.[5] When circular-economy approaches are involved, i.e., secondary-metallurgy routes, the steel's cleanliness becomes an important factor for silicon steels that are to be further processed into grain-oriented or non-oriented electrical steel sheets and coils. In the electric-arc-furnace (EAF) steelmaking process, the main raw material is ferrous scrap, and an inspection of its quality is the first crucial step in producing clean steel. Ferrous scrap is the most recycled material in the world, and through its utiliza-

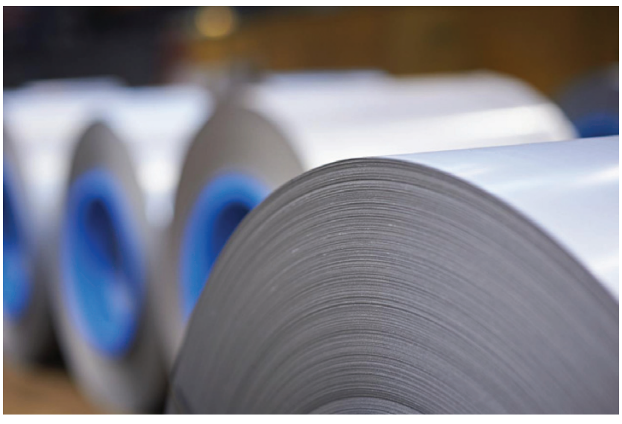

Figure 1: Coils of soft-magnetic Fe-Si-Al steel (SIWATT brand of SIJ Acroni d.o.o., Jesenice, Slovenia)[9]

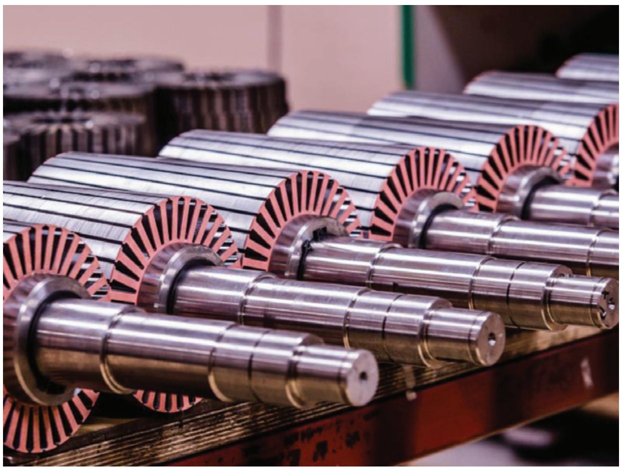

Figure 2: Rotor magnetic cores for electric motors[9]

tion, CO_2 emissions can be significantly reduced. According to the statistical data of The World Steel Association,[12] the imports and exports of scrap rose globally to >200 million metric tonnes in total, and that value is still increasing. However, efforts must be focused on the overall chemical composition of the steel. Since different qualities of scrap steel are used, the contents of impurity elements Cu, Sn, Sb, As, Ti, etc. can vary, and the control of the residual elements must be appropriate.[13,14] It is well known that the Cu content constantly increases in secondary steels. The first cross-national comparison of impurity accumulation in recycled steel[10] has shown that the content of Cu impurity was higher in Western Europe and Japan than in Ukraine, Vietnam, and China. The main sources of copper contamination are automotive scrap, waste electric and electronic equipment (WEEE) and demolition waste. Cu is mixed with cut metal scrap during shredding and is difficult to separate, unless done manually. This scrap is not fully separated from copper parts in electric motors and electrical wires. Another Cu source is scrap with a high Cu content, like rebars and stainless steel. Because information on the type of consumed scrap in all investigated countries was not available, the variation of the Cu content between countries was considered only in terms of the separation practices during recycling.[10]

4 CHALLENGES CONCERNING COPPER IMPURITY

Impurities that cannot be effectively removed from the steel melt increase the complexity of the material's behavior. One of the most contaminating elements in secondary steels is copper (Cu).

4.1 Decarburization

The decarburization of industrial cold-rolled sheets made of Fe-Si-Al steels produced by secondary metallurgy is performed by annealing in a wet gas mixture at temperatures around 850 °C. The principal reaction that controls the decarburization[7,15,16] is equation (1):

$$[C]_{Fe} + H_2O(g) = CO(g) + H_2(g) \qquad (1)$$

The exact technological parameters depend mainly on the chemical composition of the steel and are carried out according to prescribed technological methods. The decarburization is determined by the chemical reactions between the gas mixture and the carbon at the steel's surface. The temperature of the annealing, the composition of the gas mixture and the chemical composition of the steel are the influencing parameters that determine the kinetics and the mechanism of decarburization. The decarburization process of steel consists of several steps, among which is the diffusion of carbon to the steel surface among the most influential ones. One of our previous studies shows that the carbon surface segregation in the Fe-1.8Si-0.5Al alloys was hindered by the presence of larger amounts of copper (e.g, up to 0.6 *w*/% Cu) in the steel.

Carbon diffusion within the matrix of silicon steel is considered the rate-limiting step in the early stages of decarburization.[17] Elucidating the effectiveness of this metallurgical process in complex alloy systems such as secondary Fe-Si-Al steel is of utmost importance.

4.2 Magnetic properties

The core loss of the non-oriented electrical steel is defined as the dissipation of electrical energy in the form of heat during magnetization by an alternating current. In spite of the fact that the core-loss prediction in laminated magnetic circuits has generated relentless scientific research efforts, the almost forty-year-old Statistical Theory of Losses is still prevailing in this domain.[18] Since modern electrical energy conversion exposes laminated magnetic circuits to higher frequencies and larger excitation fields, novel models for core-loss prediction are pro-

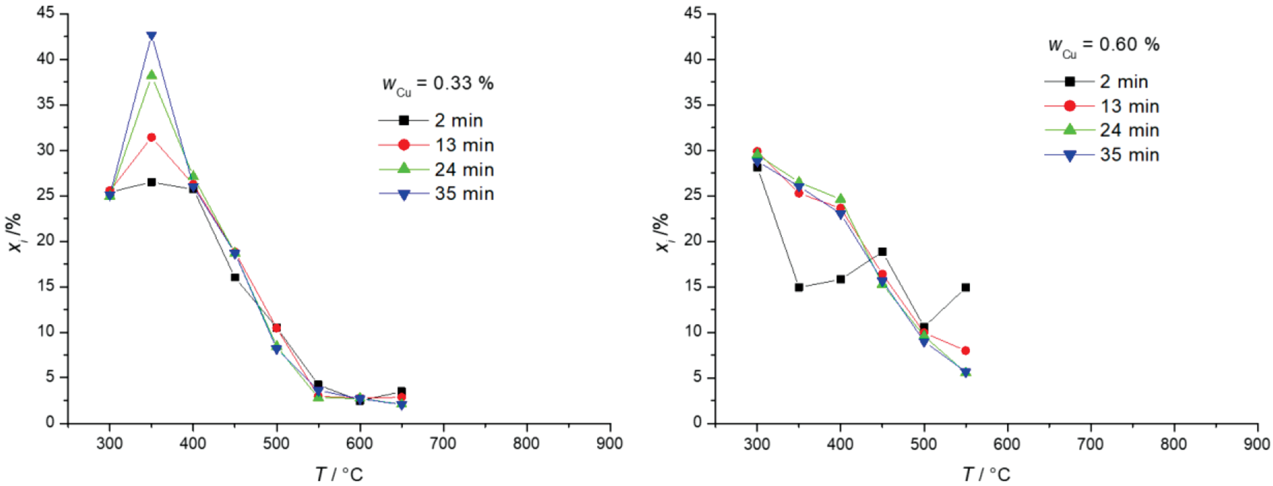

Figure 3: Carbon surface segregation in Fe-1.8 *w*/%Si-0.5 *w*/%Al alloys with different contents of Cu (0.3 *w*/%Cu and 0.6 *w*/%Cu, respectively) after annealing in vacuum

posed.[18–22] Numerous parameters, including the chemical composition and the production technology, influence the magnetic properties of the electrical steel.[23] Increased core loss could also result from the increased cumulative (total) value of the impurity elements in the steel. Impurities can have a synergy effect on the deterioration of the magnetic properties. Their elevated contents mean a larger number of inclusions and precipitates.[7,24–26]

An investigation of four different grades of fully-finished, non-oriented, electrical steels manufactured of secondary Fe-Si-Al alloys, with the silicon content from 1.0 to 2.5 *w*/% has shown that the slope of the total magnetic losses curve with increasing frequency is determined by the rate of magnetic domains mobility. The higher is the density of magnetic domains, the smaller is their mobility. The complexity of the magnetic domains and their surroundings is decreasing with increasing content of alloying elements. The rate of magnetic domains mobility is affected by the number of non-metallic inclusions and precipitates, which hinder the movement of magnetic domains and eddy currents, which produce losses and limit the mobility of domain walls.[27]

4.3 Magnetic aging

A necessary condition for obtaining high-quality electrical steel is a minimal amount of magnetic ag-

Figure 4: Instrument for measurements of magnetic properties of soft-magnetic silicon steels[9] – MPG 200D Brockhaus Messtechnik

Figure 5: Epstein tester for measurements of magnetic properties of soft-magnetic silicon steels[9]

ing.[28,29] Therefore, the electrical steel must be free of carbon and contain only small quantities of nitrogen. A low carbon content is important because the carbon that is in the form of ε-carbide at temperatures above 150 °C is eliminated within the crystal grains, and with the nitrides acting as a barrier to the movement of Bloch walls, there is a substantial deterioration in the electromagnetic properties. This process is called magnetic aging. In terms of the magnetic aging of non-oriented electrical steel of different qualities in reference to Si content, at temperatures of 225 °C and 300 °C, the most significant impact comes from the carbon content in the steel. To prevent magnetic aging associated with the precipitation of carbides, predominantly Fe_3C and ε-carbide $Fe_{2.4}C$, the decarburization of the cold-rolled steel sheets is a very important processing step.[7] Various iron carbides precipitate and degrade the magnetic properties by interfering with magnetic domain-wall motion. The slow precipitation of carbides during service can cause a substantial increase in the core losses. A study on the magnetic aging[30] of Fe-Si-Al alloys produced using secondary metallurgy has shown that the extent of the magnetic aging was affected by the copper content. The results indicate that the proportion of magnetic aging decreases with increasing copper content in samples.[30] In general, the mobility and rotation of the magnetic domains are affected by the non-metallic inclusions and precipitates, which constitute barriers to the movement of the magnetic domains.

To clearly determine the role of copper precipitates on the magnetic behavior, final magnetic properties and magnetic aging of non-oriented electrical steels, further studies are needed.

5 POTENTIAL RESEARCH TOPICS ON THE SUSTAINABILITY OF METALS

Recently, Raabe has proposed several novel directions for a potential basic research on the direct sustainability of metals,[1] some of them being: (i) efficiency of metallurgical processes, (ii) recycling of well-sorted scrap, (iii) the nanoscrap recycling from complex modern products (microelectronics and catalysts), (iv) a deep understanding of all impurity and contaminant effects in the entire spectrum of alloys, scrap, reductants, by-products, waste products and minerals, (v) multi-element and high-quality recycling, (vi) material- and recycling-oriented product design, etc. Based on this, metallurgical sustainability can be expected to break down into a large spectrum of different disciplines and branches.[1]

6 SUMMARY

Ferrous scrap is the most recycled material in the world, and through its utilization CO_2 emissions can be greatly reduced. In order to follow the set goals of the EU and achieve climate neutrality by 2050, silicon electrical steels produced with sustainable circular-economy approaches must be recognized as a strategic material for the EU. Therefore, the soft-magnetic silicon steels are strategic not only in the economic, but also in the social sense. Sustainable technologies based on the secondary metallurgy that recycles steel scrap have many limitations. Impurities that cannot be effectively removed from the steel melt increase the complexity of the material and its behavior. One of the most contaminating element in secondary Fe-Si-Al alloy is copper (Cu). The identified challenges concerning the unexplained mechanisms and efficiency of some metallurgical processes related to the presence of Cu in Fe-Si-Al steels (e.g., decarburization, magnetic domains mobility, magnetic aging, etc.) may motivate further (inter)disciplinary research, basic or applied.

6 REFERENCES

[1] D. Raabe, Materials Science behind Sustainable Metals and Alloys, Chemical Reviews 123 (**2023**), 2436–2608, doi:10.1021/acs.chemrev.2c00799

[2] https://commission.europa.eu/strategy-and-policy/priorities-2019-2024/european-green-deal_en, 8. 12. 2023

[3] https://www.mpie.de/4673836/magnetic-materials, 8. 12. 2023

[4] B. D. Cullity, C. D. Graham, Introduction to Magnetic Materials, 2nd ed., Wiley – IEEE Press, New Jersey 2008, 317–325

[5] F. Fiorillo, G. Bertotti, C. Appino, M. Pasquale, Soft Magnetic Materials, in Wiley Encyclopedia of Electrical and Electronics Engineering, (**2016**), 1–42, doi:10.1002/047134608X.W4504.pub2

[6] A. Krings, A. Boglietti, A. Cavagnino and S. Sprague, Soft Magnetic Material Status and Trends in Electric Machines, IEEE Trans Industrial Electronics, 64 (**2017**) 3, 2405–2414, doi:10.1109/TIE.2016.2613844

[7] D. Steiner Petrovič, Non-oriented electrical steel sheets - a review, Mater. Tehnol., 44 (**2010**) 6, 317–325

[8] A. El-Refaie, M. Osama, High Specific Power Electrical Machines: A System Perspective, CES TEMS 3 (**2019**), 88–93, doi:10.1109/ICEMS.2017.8055931

[9] https://sij.acroni.si/en/, 8. 12. 2023

[10] D. Panasiuk, I. Daigo, T. Hoshino, H. Hayashi, E. Yamasue, D.H. Tran, B. Sprecher, F. Shi, V. Shatokha, International comparison of impurities mixing and accumulation in steel scrap, Journal of Industrial Ecology, 26 (**2022**) 1040–1050, doi:10.1111/jiec.13246

[11] T. B. Massalski, Binary Alloy Phase Diagrams, ASM, Materials Park, Ohio 1991, 1772

[12] Steel Statistical Yearbook 2018; The World Steel Association: Brussels, Belgium, 2019; Available online: https://www.worldsteel.org/en/dam/jcr:e5a8eda5-4b46-4892-856b-00908b5ab492/SSY_2018.pdf, 8. 12. 2023

[13] A. Martinelli Miranda, P. S. Assis, G. A. Brooks, M.A. Rhamdhani, A. Fontana, A. King, G. Sanders, G. P. Da Costa Moreira, Monitoring of less-common residual elements in scrap feeds for EAF steelmaking, Ironmaking and Steelmaking 46 (**2019**), 598–608, doi:10.1080/03019233.2019.1601851

[14] D. Steiner Petrovič, Kinetics of Arsenic Surface Segregation in Scrap-Based Silicon Electrical Steel. Metals, 11 (**2021**) 1, doi:10.3390/met11010001

[15] N. Birks, Decarburization, The Iron and Steel Institute, London 1970, 1–11

[16] D. Steiner Petrovič, M. Jenko, V. Doleček, The influence of Copper on the decarburization and recrystallization of Fe-Si-Al alloys, Mater. Tehnol., 40 (**2006**) 1, 13–16

[17] L. Wen, L. Ai, L. Hong, Y. Zhou, G. Zhu, C. Sun, Diffusion Behavior of Carbon and Silicon in the Process of Preparing Silicon Steel Using Solid-State Decarburization, Processes 11 (**2023**), 3176, doi:10.3390/pr11113176

[18] B. Ducharne, G. Sebald, Fractional derivatives for the core losses prediction: State of the art and beyond, Journal of Magnetism and Magnetic Materials, 563 (**2022**) 169961, doi:10.1016/j.jmmm.2022.169961

[19] J. Füzer, S. Dobák, I. Petryshynets, P. Kollár, F. Kováč, J. Slota, Correlation between Cutting Clearance, Deformation Texture, and Magnetic Loss Prediction in Non-Oriented Electrical Steels, Materials, 14 (**2021**), 6893, doi:10.3390/ma14226893

[20] G. Novak, J. Kokošar, M. Bricelj, M. Bizjak, D. Steiner Petrovič, A. Nagode, Improved Model Based on the Modified Steinmetz Equation for Predicting the Magnetic Losses in Non-Oriented Electrical Steels That is Valid for Elevated Temperatures and Frequencies, IEEE Transactions on Magnetics, 53 (**2017**) 10, 1–5, doi: 10.1109/TMAG.2017.2726500

[21] G. Novak, J. Kokošar, A. Nagode and D. Steiner Petrovič, Core-Loss Prediction for Non-Oriented Electrical Steels Based on the Steinmetz Equation Using Fixed Coefficients With a Wide Frequency Range of Validity, IEEE Transactions on Magnetics, 51 (**2015**) 4, 1–7, doi:10.1109/TMAG.2014.2354317

[22] G. Novak, J. Kokosar, A. Nagode, D. Steiner Petrovic, Correlation between the excess losses and the relative permeability in fully finished non-oriented electrical steels, Mater. Tehnol., 48 (**2014**) 6, 997–1001

[23] M. Dems, Z. Gmyrek, K. Komeza, The Influence of Cutting Technology on Magnetic Properties of Non-Oriented Electrical Steel—Review State of the Art, Energies 16 (**2023**), 4299, doi:10.3390/en16114299

[24] D.Steiner Petrovič, M. Jenko, A HRAES study of the morphology of non-metallic inclusions in non-oriented electrical steel containing Cu and Se, Vacuum, 71 (**2003**) 1–2, 33–40, doi:10.1016/S0042-207X(02)00710-8

[25] M. Wu, Y. Zeng, Effect of copper precipitates on the stability of microstructures and magnetic properties of non-oriented electrical steels, Journal of Magnetism and Magnetic Materials, 391 (**2015**) 96–100, doi:10.1016/j.jmmm.2015.04.085

[26] D. Steiner Petrovič, M. Jenko, A. Jaklič, A. Čop, Correlation of Titanium content and core loss in non-oriented electrical steel sheets, Metalurgija, 49 (**2010**) 1, 37–40

[27] G. Novak, Designing of electromagnetic properties of non-oriented electrical steel, PhD Thesis, University of Ljubljana, Faculty of Natural Sciences, Ljubljana 2015, 1–172

[28] A. J. Moses, Energy efficient electrical steels: Magnetic performance prediction and optimization, Scripta Materialia, 67 (**2012**) 6, 560–565, doi:10.1016/j.scriptamat.2012.02.027

[29] M. F. de Campos, M. Emura, F. J. G. Landgraf, Consequences of magnetic aging for iron losses in electrical steels, Journal of Magnetism and Magnetic Materials, 304 (**2006**), 2, e593–e595, doi:10.1016/j.jmmm.2006.02.185

[30] A. Kofol, Magnetic aging of non-oriented electrical steels, MSc Thesis, University of Ljubljana, Faculty of Natural Sciences, Ljubljana 2016, 1–130