



COBALT AS A CRITICAL RAW MATERIAL

# POLISH TECHNICAL REVIEW

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# COBALT AS A CRITICAL RAW MATERIAL

### KOBALT JAKO SUROWIEC KRYTYCZNY

**Summary:** In the paper, the basic information on cobalt – the element classified, for many years, in the group of critical raw materials has been presented. The current demand on the mentioned metal is higher and higher, caused mainly by the development of new technologies and also, by energetic transformation, including a development of electric cars' market. The subject connected with obtaining of critical raw materials is nowadays a key problem due to a high dependence of the EU countries on import. The obstructed access to raw materials may dramatically affect the break of the supply chain and, in consequence, affect negatively the manufacture of materials and equipment where cobalt is a component, increase the production costs and finally, limit technological development. The present article has a review character.

Keywords: cobalt, critical raw material, non-ferrous metals, recycling

#### Introduction

According to Organisation for Economic Cooperation and Development (OECD), the world demand on materials in 2060 will be equal to 167 billion Mg. Ensuring the access to the resources becomes a priority of all countries with the aim to enable their industrialization, digitalization and also, the passage to climatic neutrality via manufacture of low-emission technologies and products [1–2]. In 2020, in connection with the above fact, the European Commission submitted the list of 30 raw materials being recognized as the Critical Raw Materials (CRM), having a strategic importance for manufacturing industry of the Euro-

Tab. 1. List of critical raw materials, announced in 2020 [1–2]

**Streszczenie**: W artykule przedstawiono podstawowe informacje na temat kobaltu, pierwiastka, zaliczanego od wielu lat do surowców krytycznych. Obecnie zapotrzebowanie na ten metal jest coraz większe, głównie z powodu rozwoju nowych technologii a także transformację energetyczną, w tym rozwój rynku samochodów elektrycznych. Tematyka związana z pozyskiwaniem surowców krytycznych jest obecnie kluczowa, ze względu na wysokie uzależnienie krajów Uni Europejskiej od importu. Utrudniony dostęp do surowców może drastycznie wpłynąć na przerwanie łańcucha dostaw, a co za tym idzie, wpłynąć negatywnie na produkcję materiałow i urządzeń w którch kobalt stanowi składnik, podwyższyć jej koszta, a ostatecznie ograniczyć rozwój technologiczny. Artykuł ma charakter przeglądowy.

Słowa kluczowe: kobalt, surowiec krytyczny, metale nieżelazne, recykling

pean Union and for the improvement of our life quality. Critical materials are those ones, the possibilities of obtaining of which from primary as well as from the secondary sources are charged with a high risk, or there are big difficulties in their obtaining and the possibilities of their substituting are small. The list of critical raw materials, as announced in 2020, is given in Tab. 1 [1–2]:

Similarly as in 2014 and 2017, the group of critical raw materials included cobalt.

Cobalt is a silver-grey metal, having ferromagnetic properties. The mentioned metal is found at two different degrees of oxidation (+2, +3) and has one isotope, existing in a natural form (59Co). The temperature of Co melting is equal to 1493°C. It is

Critical raw materials 2020

Antimony, Beryllium, Borate, Cobalt, Coking coal, Fluorite, Gallium, Germanium, HREE LREE, Indium, Magnesium, Natural Graphite, Niobium, PGM, Rock phosphate, Silicon metal, Tungsten, Barite, Phosphorus, Scandium, Bismuth, Tantalum, Hafnium, Natural rubber, Vanadium, Strontium, Lithium, Bauxite, Titanium



Fig. 1. The examples of cobalt ores: cobaltite, cariolite and linneit [4L2] [5L3] [6L4]

estimated that the content of Co in the Earth's crust amounts to 15-30 ppm [3].

Cobalt is not present in nature independently in a metallic form. Most frequently, it is found in the form combined with copper, iron, nickel, in a form of arsenide e.g. cobaltite (CoFeAsS), skutterudite (CoAS) and sulphides e.g. cariolite (CuCo<sub>2</sub>S<sub>4</sub>) and linneit (Co, Ni)<sub>3</sub>S<sub>4</sub> [4].

Fig. 1 represents exemplary cobalt ores: cobaltite, cariolite and linneit [4–6].

### Cobalt deposits

The deposits of cobalt are found all over the world (Fig.2), mainly in African copper belt in the Democratic Republic of Congo (DRC). Cobalt is produced in technologies, accompanying production of basic metals, most frequently of copper (44%) or nickel (50%). In the EU countries, and especially in Belgium and Finland, the different products are made of cobalt (metal, oxide, and hydroxide) on the basis of imported primary raw materials, mainly from the Democratic Republic of Congo (DRC) and Zambia [7].

#### Production of cobalt

According to the data found below in Fig. 3, mining production of cobalt increased dramatically in 2009 and since the mentioned period it has been maintained at the constant level.



Fig. 3. Mining production of cobalt in the period of 2005-2015 [9]



Fig. 2. Cobalt deposits [8]



Fig. 4. Example of one of the mines where the extraction of cobalt is performed by "handicraft" method [27]

At present, almost 60% of the global cobalt production is implemented in the Democratic Republic of Congo. The cobalt-rich regions form there big mining complexes. The process of cobalt extraction in the DRC takes place in the mines differing from those ones which are functioning in the developed countries. The mines in the DRC create a group of small mining plants, having a typical handicraft character i.e. heavy mining machines are not employed. It is estimated that 15-30% of the cobalt obtained in the discussed country comes from the mines which do not meet the safety and hygiene standards and their functioning is not regulated by the state organizations of mining supervision. The United Nations informs that 40 thousand children are working in the mentioned type of manufacturing plants. A lack of real control of mining of the raw materials in Congo results from numerous ethnical conflicts, or still recurring epidemics of Ebola virus at the discussed territory [10].

During the recent years, a part of the companies, mainly of Chinese ones, has undertaken the implementation of the projects aimed at formalization of handicraft mining what is expected to lead to safety increase as well as observing the human rights in such work places. The formalization of the activities of the discussed above complexes, consisting of small handicraft mines is based upon the verification of the workers who want to obtain an access to the site of extracting the raw materials. The verification consists, *inter alia*, in control of soberness or monitoring of the age of the employed persons (the worker must have 18 years completed). Additionally, the periodical risk assessment and medical training are carried out [11].

### Metallurgy of Cobalt

The processes, employed in cobalt production are based on pyrometallurgical process of processing the cobalt-containing ores i.e. ores of copper and nickel (at high temperatures) and then, hydrometallurgical processes, with the aim to extract cobalt from the obtained alloys in a metallic form or in cobalt compounds. Manufacture of cobalt from sulphide ores of nickel is based upon melting of cobalt-containing sulphide concentrate of nickel (generated by flotation of sulphide ores) and production of nickel matte, containing considerable quantities of iron. Then, the obtained stone is subjected to converting process, in consequence of which the iron-poor matte is produced which is subjected to hydrometallurgical processing with the aim to recover cobalt from nickel. From the concentrate, obtained by the mentioned method, we obtained nickel and cobalt in a pure form [3].

Initially, the manufacture of nickel from laterite ores was not integrated with production of cobalt and the iron-nickel alloy was a final product of the described process. In the fifties of the 20<sup>th</sup> century in Cuba, there were employed the first integrated methods of nickel metallurgy, allowing the separation of cobalt from laterite ores. The mentioned method was based on the hydrometallurgical high-pressure leaching of cobalt from laterite ore, using sulphuric acid (VI) and successive its recovery from water solutions. At present, the discussed technology is universally employed [3].

Production of cobalt as copper-accompanying metal is practised, first of all, in relation to the deposits in Africa. The extracted ore is usually subjected to leaching and copper is separated from cobalt by the application of extraction with solvent. In effect, water solution of cobalt is obtained and it is precipitated using magnesium oxide to cobalt hydroxide. The cobalt hydroxide may become a commercial product, or may be later processes by electrolytic methods to obtain metallic cobalt [3].

#### Application of cobalt

Due to its hard fusibility and resistance to corrosion, cobalt is used in many applications such as super alloys, additives to highly-cutting steels, machining tools and, also, in electronics e.g. in lithium-ion batteries. The discussed metal has also magnetic properties; therefore, it is employed in production of permanent magnets (samarium-cobalt, Sam-Co); alnico – compound of aluminium, nickel and cobalt).

#### Superalloys based on cobalt

From among super alloys, we can distinguish super alloys based upon the nickel, iron and cobalt. Owing to additives of carbon and other metals such as wolfram and molybdenum, the alloys which are based on cobalt preserve their properties at the increased temperatures better than the alloys based on

#### Tab. 2. The composition of the selected cobalt alloys [13–15]

nickel. Additionally, cobalt super alloys contain often the addition of chromium (Cr); owing to this fact, such alloy is exceptionally resistant to corrosion. The elements made from cobalt super alloys are less resistant to cracking as compared to nickel alloys [12].

Table 2 shows the example of the composition of the selected cobalt alloys and their application.

	Chemical composition, %	Properties	Application
Haynes 25™ Alloy L605 [13]	C- 0,05-0.15; Mn-1.0-2.0; P-0.4; S-0.03; Si-0.4; Cr-19.0-21.0; Ni-9.0-11.0; W-14.0-16.0; Co-balance	Temperature strength and oxi- dation resistant to 2000°F (ca. 982°C). Resistant to acids, body fluids and marine environments	Gas turbine engine components; Springs; Heart valves; High temperature ball bearings and bearing races
High-Performance Alloy6B [14]	C- 0.9-1.40; Mn-max. 2.0; Si- max. 2.0; Cr- 28.0-32.0; Ni-3.0; Mo-1.50; Fe 3.0; W-3.50-5.50; Co-balance	corrosion and wear resistant high hardness at elevated tem- peratures	Medical blades; Valve parts; Pump plungers; Knives; Bearings
NASA Co-W-RE Cast Cobalt Alloy [15]	C-0.40; Cr- 3.0; Co-67.6; Nb-1.0; Re-2.0; W-25; Zr-1.0	-	High-temperature space applications

#### Magnets

Permanent magnets are such materials which preserve their magnetic properties after exposition to magnetic field effect. They are found in a wide spectrum of materials, employed in many industrial and commercial applications. They include micro-engines and condensers used in computers, audio-visual equipment (loudspeakers, video recorders etc), cars (electric windows, ABS, computers in dashboard etc) and electronic household appliance and equipment (dishwashers, washing machines, air conditioners etc.). Permanent magnets are also used as time engines in industrial robots, military and cosmic technologies and in clocks [16–17].

Permanent magnet materials are identified by the following principal magnetic properties [17]:

- Maximum value of energy product, BH<sub>max</sub> (MGO);
- Residual induction, B<sub>r</sub> (gauss);
- Coercive force, H<sub>c</sub> (oersteds).

#### Tab. 3. Parameters of AlNiCo magnets [17]

Types of permanent magnets according to their chemical composition:

- Alnico
- Ceramic
- Rare-Earth
- Iron-Chromium-Cobalt.

#### AlNiCo magnets

Alnico magnets are the alloy of aluminium (Al), nickel (Ni), cobalt (Co), copper (Cu) and iron (Fe). The mentioned magnets are the oldest permanent magnets. The development of Alnico alloys was commenced in the thirties of the 20<sup>th</sup> century. Alnico alloys are characterized by a high resistance to demagnetization (i.e. high coercion). In spite of the fact that other permanent magnets (e.g. Sm-Co) are more frequently applied, Alnico magnets are still employed in aircraft or car industry (in sensors).

			Chemical of	compositio	n		Magnetic properties				Working temp.
Original MMPA Class	AI	Ni	Co	Cu	Ti	Fe	BHmax	Br	Hc	Hi	т
	%						MGOe	Gauss	Oer	sted	°C
Alnico 1	12	21	5	3	-	balance	1.4	7200	470	480	No data
Alnico 2	10	19	13	3	-	balance	1.7	7500	560	580	450
Alnico 3	12	25	-	3	-	balance	1.35	7000	480	500	450
Alnico 5	8	14	24	3	-	balance	5.5	12800	640	640	525
Alnico 5 DG	8	14	24	3	-	balance	6.5	13300	670	670	525
Alnico 5-7	8	14	24	3	-	balance	7.5	13500	740	740	525
Alnico 6	8	16	24	3	1	balance	3.9	10500	780	800	525
Alnico 8	7	15	35	4	5	balance	5.3	8200	1650	1860	550
Alnico 8HC	8	14	38	3	8	balance	5.0	7200	1900	2170	550
Alnico 9	7	15	35	4	5	balance	9.0	10600	1500	1500	550

Table 3 shows the parameters of Alnico magnets according to the standards, defined by [17].

### SmCo magnets

Samarium-cobalt magnets (Sm-Co) are applied at high temperatures. They have working temperature reaching to 350°C [18]. Table 4 contains the most important parameters of Sm-Co magnets [18]. Sm-Co magnets have a high resistance to corrosion in wet, acid or alkaline environment. Due to the discussed properties, they may be employed in engines of traction railway locomotives and heavy industrial engines and generators [19]. Besides it, Sm-Co magnets are also used in medical implants and tools [18]. Fig. 5 represents the percentage participation of SmCo magnets in various applications [19].

Tab. 4. Parameters of Sm-Co m	nagnets [18]
-------------------------------	--------------

	-	Remanence Br	Normal coercivity HcB	Maximum energy pro- duckt BH <sub>max</sub>	Maximum operating temperature	
Goudsmit Grade code	Гуре	typical value (mT)	Typical value (kA/m)	Typical value (kJ/m³)	Tmax (°C)	
S20	SmCo5	950	756	175	250	
S22	SmCo5	1000	772	191	250	
S24	Sm2Co17	1040	796	199	350	
S26	Sm2Co17	1060	820	215	350	
S28	Sm2Co17	1100	820	231	350	
S30	Sm2Co17	1120	828	247	350	
S32	Sm2Co17	1150	796	239	350	



Fig. 5. Application of Sm-Co magnets [19]

### Rechargeable batteries

Lithium-ion cells belong to the most popular rechargeable batteries [26]. They are characterized by a high density of the stored energy in comparison to the remaining types of rechargeable batteries [21]. Lithium-ion cell consists of five components: anode, cathode, electrolyte, polymer separator and nickel-covered steel or aluminium casing. Cathode is made of oxides of transitory metals (most frequently cobalt, manganese, nickel). The electrode material is laid on copper and aluminium foils. The electrodes are separated with the separator in a form of polyethylene or polypropylene. The electrode materials are immersed in electrolyte solution which enables movement of lithium ions between the electrodes [26]. Cathodes are - most frequently -

Tab. 5. The approximated composition of lithium-ion batteries (LCO, NMC)

Marking	Cathode	Anode	Application	Approximate content
LCO	LiCoO <sub>2</sub>	graphite	Mobile telephones, tablets, laptops, cameras	Li 2,7%; Co 22,8%; Ni 0,2%; Cu 8%
NMC	LiNi <sub>x</sub> Mn <sub>y</sub> Co <sub>2</sub> O <sub>2</sub>	graphite	Electric cars, medical equip- ment, scooters, electric bicycles, electric tools	Ni 39 g/kg; Co 39 g/kg; Mn 36 g/kg; Li 14 g/kg

cobalt spinels (LiCoO<sub>2</sub>; LCO), manganese spinels (LiMn<sub>2</sub>O<sub>4</sub>; LMO) intercalated with lithium ions. On the other hand, graphite is employed as anode material [21]. Table 5 shows the examples of the employed cathode materials and the approximated composition of lithium-ion batteries [2, 20].

### Demand on cobalt

Development of lithium-ion batteries (LIB) has experienced a tremendous growth during the recent years; it is connected with the increasing production of mobile devices and with the constantly growing demand in the sector of electric vehicles (EV). It is estimated that the demand on lithium-ion batteries will be increasing annually above 30% during the coming 10 years. Manufacture of lithium-ion batteries employs mainly such raw materials as copper, graphite (natural or synthetic), cobalt, lithium, manganese, aluminium and nickel [1, 22]. The EU countries (EU-27, without Great Britain) produce at present only 1% of all raw materials, necessary for manufacture of batteries. Asia, as being represented by China, Japan and the South Korea, supplies 86% of the processed materials and components for production of lithium-ion batteries all over the world. The Peoples' Republic of China accounts for 66% of global production of lithium-ion

batteries. A full dependence of the EU countries on import of cells and batteries creates a serious threat to the raw material deficit and potentially high costs connected with the application of technologies of lithium-ion batteries. Due to a high demand on cobalt and a strong dependence on the supplies from other countries, there is a high necessity of recycling of cobalt from various applications [1–2].

### The methods of recycling

The technologies of recycling of the raw materials, containing critical metals, are based on the pyrometallurgical and hydrometallurgical processes [23].

Table 6 shows the examples of the employed processes of recycling the rechargeable lithium-ion batteries [2; 24].

Table 7 presents the advantages and disadvantages of the processes, employed in recycling of lithium-ion batteries, with the consideration of mechanical processes [25].

The high temperature processes are characterized by a high charge to the environment in connection with the generation of dusts and gases and high energy consumption [23]. The hydrometallurgical processes are characterized by lower energy consumption and a high selectivity of the recovered metals. The

Company	Recovered metals	Proces	Country
Glencore Recycling	Ni, Co, Cu	thermal preparation + pyrometallurgy + hydrometallurgy	Canada
AkkuSer	Ni, Co, Cu, Fe	mechanical process	Finland
Recupyl	Ni, Co, Cu, Fe, Li	mechanical process + hydrometallurgy	France
Umicore	Ni, Co, Cu, Fe	pyrometallurgy + hydrometallurgy	Belgium
Accurec	Ni, Co, Cu	thermal preparation + pyrometallurgy + hydrometallurgy	Germany
Kyoei Seiko	Ni, Co, Fe	pyrometallurgy	Japan
JX Nippon	Ni, Co, Cu, Fe, Mn, Li	thermal preparation + mechanical process + hydrometallurgy	Japan
Dowa	Ni, Co, Cu	thermal preparation + pyrometallurgy + hydrometallurgy	Japan
GEM	Ni, Co, Cu, Fe, Mn, Li	mechanical process + hydrometallurgy	China
Brunp	Ni, Co, Cu, Fe, Mn	mechanical process + hydrometallurgy	China
Telerecycle	Ni, Co, Cu, Fe, Li	mechanical process + hydrometallurgy	China
Kobar	Ni, Co, Cu, Fe	mechanical process + hydrometallurgy	Korea
Retriev	Ni, Co, Cu, Fe	mechanical process (in liquid) + hydrometallurgy	United States of America
Batrec	Ni, Co, Cu	mechanical process	Switzerland

Tab. 6. Examples of recycling technologies of lithium-ion batteries [2, 24]

Tab. 7. Advantages and drawbacks of hydrometallurgical, pyrometallurgical and mechanical processes [25]

Process	Advantages	Disadvantages
Hydrometallurgy	High recovery rate High purity product Low Energy consumption Less waste gas High Selectivity	More wastewater Long process
Pyrometallurgy      Simple operation and short flow        No requirement for categories and the size of inputs      High efficiency		Li and Mn are not recovered High energy consumption Low recovery efficiency More waste gas and the cost of waste gas treatment
Physical recycling process	Short recovery route Low energy consumption Environmental friendly High recovery rate	High operational and equipment requirements Incomplete recovery

main disadvantage of the discussed process consists, however, in generation of waste in a form of sewages, containing the dangerous substances [23].

#### Summing up

In spite of the existing and functioning plants which process the cobalt-containing waste, the EU countries are still dependent on the supplies of the discussed raw material from other countries. The mentioned metal has been recently widely employed in lithium-cobalt batteries. Cobalt is a metal, affecting the development of the newest technologies; additionally, in certain applications, it cannot be replaced by another raw material.

The Research Network Łukasiewicz – Institute of Non-Ferrous Metals has conducted, for many years, the studies on the technologies of waste management in Poland; their solutions were introduced in many enterprises. The Research Network Łukasiewicz is the third – in respect of size – research network in Europe. It supplies attractive, complete and competitive technological solutions. It offers the unique system of "Challenging" to the business; owing to the mentioned system, is undertakes the business challenges during not longer than 15 days and submits, free-of-charge, the direction of solving problem and the ideas of the research project to the entrepreneur [28].

#### References

- Study on the EU's list of Critical Raw Materials (2020) Final Report "European Commission, Study on the EU's list of Critical Raw Materials - Final Report (2020)" ISBN 978-92-76-21049-8 doi: 10.2873/11619
- [2] M. Potempa, surowce krytyczne w nowych technologiach artykuł przeglądowy, Rudy i Metale Nieżelazne, Rocznik 2020 – zeszyt 11-12
- [3] S. Roberts, G. Gunn, Cobalt, Critical Metals Handbook, First Edition. Edited by Gus Gunn, pp 123-149; Nottingham, UK 2014
- Kobaltyt, Available online: https://flosferriminerals.com/803-medium\_ default/kobaltyt-hnusta-mutnik-slovakia.jpg (accessed on 10 June 2021)
- Kariolit, Available online: https://kopalniawiedzy.pl/media/lib/73/skutterudite-c94b4f6ee1c403cac8a1329ee8b51747.jpg (accessed on 10 June 2021)

- [6] Linneit, Available online: https://www.mineralienatlas.de/VIEWmaxFULL.php/param/1111939054-Linneit-Kubooktaeder-bis-4-mm-Linnaeit.jpg (accessed on 10 June 2021)
- [7] Study on the review of the list of Critical Raw Materials, Critical Raw Materials Factsheets, Luxembourg: Publications Office of the European Union, 2017.
- [8] Map of cobalt resources, Available online: https://www.firstcobalt. com/\_resources/images/geology-of-cobalt-Figure-4.jpg (accessed on 10 June 2021)
- [9] Evi-Petavratzi, Gus Gunn, Carolin Kresse, "Commodity review: Cobalt", British Geological Survey, 2019
- [10] Available online: https://m.ekonsument.pl/a67058\_ludzie\_ktorzy\_umieraja\_za\_twojego\_smartfona.html (accessed on 10 June 2021)
- [11] F. Calvão, C. Erica, A. Mcdonald, M. Bolay "Cobalt mining and the corporate outsourcing of responsibility in the Democratic Republic of Congo", The Extractive Industries and Society 2021
- [12] N. Mohammed Dawood and A. Mishaal Salim, "A Review on Characterization, Classifications, and Applications of Super Alloys", JUBES, vol. 29, no. 1, pp. 53 - 62, Apr. 2021.
- [13] Haynes 25 Alloy L605, Available online: https://www.magellanmetals. com/haynes-25 (accessed on 10 June 2021)
- [14] Cobalt alloy 6B, Available online: https://www.magellanmetals.com/ cobaltalloy-6b (accessed on 10 June 2021)
- [15] Co-W-RE Cast Cobalt Alloy; Available online: http://www.matweb. com/search/datasheet.aspx?matguid=f30c81ec85a14bd39653e-2aa226630bd&ckck=1 (accessed on 10 June 2021)
- [16] Z. Zheng, J.E. Greedan, Rare Earth Elements and Materials; Encyclopedia of Physical Science and Technology (Third Edition), 2003 Pages 1-22
- [17] Standard specifications for permanent magnet materials; MMPA STAN-DARD No. 0100-00; Magnetic Materials Producers Association; ILLINOIS
- [18] SmCo magnets; Available online: https://www.goudsmitmagnets.com/ pl/solutions/permanent-magnets-tapes-sheets/permanent-magnets/ samarium-cobalt.html (accessed on 10 June 2021)
- [19] X. Zhou, A. Huang, B. Cui, J.W. Sutherland, Techno-economic Assessment of a Novel SmCo Permanent Magnet Manufacturing Method, Procedia CIRP, Volume 98, 2021, Pages 127-132
- [20] R. Danino-Perraud, "The Recycling of Lithium-ion Batteries: A Strategic Pillar for the European Battery Allience", Études de l'Ifri, Ifri, March 2020
- [21] M. Świętosławski, Kierunki rozwoju akumulatorów litowych, Przemysł Chemiczny 95/1, 2016
- [22] A. Holzer; S.Windisch-Kern; C. Ponak; H. Raupenstrauch, A Novel Pyrometallurgical Recycling Process for Lithium-Ion Batteries and Its Application to the Recycling of LCO and LFP. Metals 2021, 11, 149. https:// doi.org/10.3390/met11010149

- [23] N. Um, Hydrometallurgical Recovery Process of Rare Earth Elements from Waste: Main Application of Acid Leaching with Devised T-T Diagram, Rare Earth Element, DOI: 10.5772/intechopen.68302
- [24] P.Tecchio; F Ardente; M. Marwede; C. Christian; G. Dimitrova; F. Mathieux, Analysis of material efficiency aspects of personal computers product group, EUR 28394 EN, Publications Office of the European Union, Luxembourg, 2018, ISBN 978-92-79-64943-1, doi:10.2788/89220, JRC105156.
- [25] LF. Zhou; D. Yang; T. Du; H. Gong; WB. Luo, The Current Process for the Recycling of Spent Lithium Ion Batteries. Front Chem. 2020;8:578044. Published 2020 Dec 3. doi:10.3389/fchem.2020.578044
- [26] M. Bakierska; A. Chojnacka, Akumulatory litowe jako współczesne systemy magazynowania energii, Wiadomości Chemiczne 68, 2016
- [27] Cobalt mining, Available online: https://content.fortune.com/wp-content/uploads/2018/08/sebastianmeyer\_cobalt\_100-e1535035752615. jpg?resize=1200,600 (accessed on 10 June 2021)
- [28] www.lukasiewicz.gov.pl/biznes.

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# THE HISTORY OF INTRODUCING THE SILICONES IN POLAND

### HISTORIA WPROWADZENIA SILIKONÓW W POLSCE

**Summary:** The present elaboration presents a fragment of the history of developing and implementation of the manufacturing technology and application of silicones in Poland. At present, the silicones are already well known in Poland not only for plumbers or producers of windows, restorers of monuments and physicians but also for the users of cosmetics or baby pacifiers. Market-oriented economy and prices decide on the choice of the supplier. Undertaking the studies and a small-scale production about 70 years ago has contributed to their wide utilization and benefits for the economy.

**Keywords**: silicon, oil, fluid, rubber, resin, hydrolytic poly-condensation, monomer polymerization

Streszczenie: Artykuł przedstawia fragment historii opracowań i wdrożeń technologii produkcji oraz zastosowań silikonów w Polsce. Obecnie silikony są już dobrze znane w Polsce, nie tylko hydraulikom czy producentom okien, konserwatorom zabytków i lekarzom, ale też użytkownikom kosmetyków czy smoczków. Wolny rynek i ceny decydują o wyborze dostawcy. Podjęcie przed blisko 70. laty badań, niewielkiej produkcji przyczyniły się do ich o szerokiego wykorzystania i korzyści dla gospodarki.

**Słowa kluczowe**: silikon, olej, kauczuk, żywica, polikondensacja hydrolityczna, polimeryzacja monomerów

### What are the silicones?

Silicones are organic polysiloxanes – high molecular siliconorganic compounds, composed of silicon atoms, linked with oxygen bridges, similarly as in non-organic silicates, but containing, additionally, organic groups, connected with the silicon atoms, such as, for example:

We can distinguish three varieties of silicones: oils (fluids), rubbers and resins. Oils have a structure of simple or branched siloxane chains with a molecular weight in the range of several – few thousand g/mol. Rubbers – elastomers have the structure of long siloxane chains with the molecular weight from few thousand to several hundred thousand g/mol. Resins have the structure of siloxane networks with the molecular weight amounting to few hundred – several thousand g/mol.

From the mentioned three basic varieties, the new type of silicones such as emulsions, pastes, greases, gums, glues, fitters, varnishes, paints, hydrophobic, antifoaming and anti-adhesive agents and others, have been developed and are still produced. From among plastics, the silicones are distinguished by the following specific properties:

- thermal and oxidative resistance in a wide spectrum of temperatures from -60°C to 250°C,
- the resistance to the effect of atmospheric factors, UV radiation and other,
- good dielectric properties,
- hydrophobic capabilities,
- a small variation of physical properties in a wide temperature spectrum.

Silicones are mainly obtained in the processes of hydrolytic poly-condensation or polymerisation of monomers, as e.g.:

$$\begin{array}{c} \mathsf{CH}_3 \ \mathsf{CH}_3 \ \mathsf{CH}_3 \ \mathsf{CH}_3 \\ | \ | \ | \ | \\ \mathsf{H}_2 \\ \mathsf{CH}_3)_2 \mathsf{SiCl}_2 + \mathsf{n} \ \mathsf{H}_2 \mathsf{O} = \mathsf{HO}\text{-}\mathsf{Si}\text{-}\mathsf{O}\text{-}\mathsf{Si}\text{-}\mathsf{O}\text{-}\mathsf{Si}\text{-}\mathsf{O}\mathsf{H} + \mathsf{4} \ \mathsf{HCl} \\ | \ | \ | \\ \mathsf{CH}_3 \ \mathsf{CH}_3 \ \mathsf{CH}_3 \ \mathsf{CH}_3 \end{array}$$

Methyl- and phenylchlorosilanes and their derivatives: alkoxy-, acetoxy- and vinylsilanes are most frequently used monomers but many reactive organic compounds are employed, as well. The most important raw material, used in synthesis of monomers is metallic silicon, non-occurring in a natural form, but synthesized in metallurgical processes from silicate and carbon in the following reaction:

The first trials to synthesize organic polysiloxanes at a laboratory scale were conducted in the thirties of the 20<sup>th</sup> c., mainly in the reaction of siliconetetrachloride with the magnesiumorganic compounds and then, hydrolytic polycondensation of the obtained magnesium-organic compounds. The attempts to utilize the mentioned method in production of silicones were undertaken at the beginning of the forties by General Electric and Corning Glass in the USA, as performed by the teams, guided by W. Patnode and E.G. Rochow. After certain time, E.G. Rochow invented more economic method of direct synthesis of methylchlorsilanes in reaction of powdered metallic silicon with methyl chloride, with the participation of copper powder as a catalyst. The reaction occurred at the temperature of 260-300oC in fluidal phase, in the special reactors. There was obtained a liquid mixture of the products which were separated by a fractionated distillation method in the columns with a very high efficiency. Four methylchlorosilanes were separated: CH<sub>2</sub>SiCl<sub>2</sub>, (CH<sub>2</sub>)<sub>2</sub>SiCl<sub>2</sub>, CH\_SiHCl\_ and (CH\_)\_SiCl. There was always developed the method for synthesis of phenylchlorosilicones in reaction of powdered silicon with chlorobenzene at temperature of ca. 450°C, also with copper or silver catalyst.

Production of the silicones started in 1943 in the USA, and in the fifties, it was also commenced in Great Britain, France and Federal Republic of Germany, on American licences and, also, on the own developments, in the Soviet Union and Democratic Republic of Germany. In the Eastern Europe, the work on the production technology of monomers and certain varieties of the silicones was conducted at the mentioned time – apart from the Soviet Union and DDR – at the Academy of Sciences in Prague. The first American publications, patents and the first monographs appeared: American – by E.G. Rochow, – Russian by K.A. Andrianov and Czech – by Prof. Bażant and Chvalovski; the recent one was translated into Polish in 1955 [1].

#### The beginning of the research studies in Poland

In 1952, the Ministry of Chemical Industry instructed the former Institute of Plastic Materials (ITS) in Warsaw to develop the technology of synthesizing the silicones, including varnish resins for production of heat-resisting silicone paint, necessary for painting the nozzles of military aeroplanes MIG on the Soviet licence. There was created a team, consisting of several persons, guided by L. Zakrzewski. The author of the present paper received a task to develop the synthesis of methylchlorosilanes.

After having familiarized with the available abstracts in *Chemical Abstracts* and assembling the appropriate apparatus, we brought the samples of ferrosilicon with Si content equal to 90–95%, and the samples of copper powder and methyl chloride. After grinding the ferrosilicon and collecting the sieved powder with grains below 0.1 mm, we conducted the trials to synthesize methylchlorosilanes in pipe reactors, heated in electric furnaces at temperature of 260-320°C, passing the methyl chloride vapours over the mixture of silica with the addition of cooper and collecting the condensed mixture of methylchlorosilanes which were separated by a fractionated distillation [2, 3, 4, 5]. After con-

ducting ca. 150 trials during ten months, which lasted 100 h each of them, we commenced (in 1954) the trials of synthesis at a semi-technical scale in a metal reactor, containing 50 kg of silicon and equipped with the agitator and electric heating system. The obtained methylchlorosilanes were separated on a glass column with 50 mm diameter and height of ca. 6 m, filled with glass helices, with the electronically operated head.

In parallel, the team of scientists conducted the studies on synthesis of phenylsilicone monomers in reaction of silicontetrachloride and bromobenzene by Grignard method and later on, the attempts to synthesize silicone oils and resins from the obtained monomers. They commenced a series of publications on silicones and their applications [6,7,8,9,10] while conducting the consultations with the representatives of western companies, which offered the samples of the silicones and the prospects. In 1958, the author of the present paper participated in the conference on silicoorganic compounds held in Leningrad and delivered a lecture concerning Polish trials to synthesize methylchlorosilanes [11].

I developed the monograph concerning the applications of silicones which was published in 1964 [12]. We got information on the studies on siliconorganic chemistry, as being conducted in the fifties by the teams of Z. Lasocki at the Łódź University of Technology and of W. Rodziewicz at the Gdańsk University of Technology. When possessing the above information, we organized symposium of the experts in siliconorganic matters at our Institute (in 1965). During the mentioned seminar, the research achievements and the possibilities of cooperation as well as financing of the studies were discussed. When perceiving the visible benefits resulting from such meetings, the participants of the conference agreed to organize the symposia of Polish "siliconorganic chemists" every three years. During the next meetings, the successive teams joined the debates; their number was increased, inter alia, by the scientists from the University of Poznań, Gdańsk Medical University and the Centre of Molecular and Macromolecular Studies in Łódź.

#### Preparations to production

As we could not continue production of methylchlorosilanes at the Institute (threat to the environment), we obtained – in 1958 – a decision of the Ministry of Chemical Industry on the transfer of further semi-technical trials and construction of technical installation to the Chemical Works in Nowa Sarzyna. In 1962, there was created a small Department of Silicones at the mentioned plant. It was equipped with the reactor for synthesis of methylchlorosilanes, transported from our Institute. The installation for their fractionated distillation and some sets of the equipment for semi-technical production of silicone fluids and resins were assembled. Production of methylchlorosilanes (with my participation) was commenced and the first attempts to synthesize silicone oils and resins were carried out.

Manufacturing capacity of monomers did not exceed 3t/ year what was insufficient quantity for a profitable production of the silicones. In 1964, the Chemical Works did not have the

sufficient financial means for the project and building of technical installation and eventual purchase of licence, so they gave up the continuation of production and transmitted the mentioned object to the Institute. It became transformed into Experimental Station of Silicones (Polish: ZDS) which was subordinate to the management of the Institute. We utilized the new possibilities of importing the monomers from the Soviet Union, and later on, from DDR and we introduced practically the technologies of synthesising different varieties of silicones for the successive 30 years. The mentioned silicones were developed at our Institute in a laboratory scale. We cooperated with the employees of the Experimental Station in the field of the production improvement and in marketing.

The beginnings were very difficult as it was a period of the so-called cold war and the heavy and military industries were mainly financed at the cost of chemical industry, including industry of plastic materials, and the research work. There was a system of limited distribution of foreign currency for import of raw materials and equipment for possible investments. In the sixties and the seventies, there were considerable restrictions in contacts with the western scientists and companies and the passports for common citizens were practically unavailable.

Apart from introduction and development of the successive varieties of the silicones, the important task of ZDS included the development of the methods for attestation and then, technical conditions and elaboration of the prospects and guidelines to be employed as well as claims of 15 trade names at the Patent Office. After overcoming the synthesis of methylchlorosilanes, we were occupied in development of the successive production technologies of silicones and the attempts of their introduction; it was carried out at the Experimental Station (ZDS).

### Methylsilicone oils

The first silicone oils, as developed at the Institute in the period of 1954–1956 by M. Tomaszewicz [13] were methylsilicone oils; they were obtained by the hydrolysis of dimethyldichlorosilane with water and after the separation from hydrochloric acid, by condensation of the resulting dimethylsiloxanoles with hexamethyldisiloxane ("MM") which served for termination of dimethylsiloxane chain. The length of the resulting siloxane chain and the relating viscosity of the oil were regulated by the choice of the quantity of the added MM.

After implementation of the described above technology at the Experimental Plant in 1964, there were introduced oils, named "*Silol*"; they had five viscosity values in the range of 20-1000 MPa·s. We developed also the method for obtaining water emulsions of the mentioned oils. In 1965, we introduced four emulsions *Aquasil* to manufacture.

The successive products, obtained from methylsilicone oils and colloidal silicate and introduced at ZDS were two silicone pastes – "Silpasta". We utilized methyl dichlorosilane  $CH_3SiHCl_2$ , obtained during the synthesis of methylchlorosilanes, and dimethyldichlorosilane and we developed the method for synthesis of methyl hydrogensilicone oils [14, 15, 16]. The mentioned method was later employed in obtaining of the agents for waterproof impregnation of fabric – *Siltex*.

In 1973, the management of Chemical Works Sarzyna made the contacts with the French company Rhone-Poulenc; within the frames of the mentioned cooperation, there was undertaken production of methylsilicone oils, water emulsions and pastes produced from semi-products, imported from France. The Department of Silicones was organized in the Chemical Works and in 1975, it commenced a technical-scale production of methylsilicone oils and their processing into emulsions and pastes; their semi-technical production in the Experimental Station was stopped.

### Methylsilicone resins

The first methylsilicone resin was obtained in 1955 at the Institute of Plastic Materials (ITS). It was carried out by dropping methyltrichlorosilane to water, separation of solid resin from hydrochloric acid by filtration and washing out with water. Then, the powdered resin was dissolved at hot in a ca. 40-% solution of potassium or sodium hydroxide. The obtained solutions of methyl sodium and potassium siliconates were used (after appropriate dilution with water, most frequently to 5%) as the agents for hydrophobicity of different materials (it will be discribed later on in the present paper).

Potassium methylsiliconate, called *Ahydrosil-K*, was implemented at the semi-technical scale in 1965, in the Experimental Station. Our main task was however, to develop varnish methyl silicone resin for manufacture of heat-resistant paint. The attempts to synthesize the mentioned resin were conducted by hydrolytic polycondensation of methyltrichlorosilane with a small (up to 10%) addition of dimethyl dichlorosilane, in the solutions of various solvents, using water with the addition of different alcohols. Owing to the application of alcohol, we were successful in mediating the process of polycondensation of monomers; apart from hydrolysis of Si–Cl bonds at the border of organic phase and water, the alcoholysis in organic phase had place in the solution of monomers of the most reactive Si–Cl bonds in methyl trichlorosilane:

### MeSiCl<sub>3</sub>+ROH = MeSi(OR)Cl<sub>2</sub> + HCl

After many trials, we chose toluene as solvent and cyclohexanol with a very limited (ca. 3%) water-solubility as a moderator of hydrolysis; owing to this fact, in spite of a small addition of dimethylchlorosilane (up to the methyl groups content, expressed by ratio of 1.09 mole of  $CH_3$  per 1 mole of Si), the cyclohexyl groups –  $OC_6H11$  were additionally built-in the structure of resin. Their quantity was – according to the results of elementary analysis and NMR – equal to 0.08-0.13%. Moreover, thanks to the better solubility of cyclohexanol in toluene than in water, its losses in the process of water phase separation and washing of the obtained resin solution were limited. In effect of conducting many trials, we established the recipe and the method for synthesis of varnish methylsilicone resin and we applied for the patent rights for the

mentioned method. We prepared also the respective publications [17, 18, 19]. In 1964, the discussed above resin was introduced to *a semi-technical production under the name Silak M10* at the Experimental Station.

In the years 1962–1965 we carried out also the trials to synthesize methylphenyl- silicone resins, utilizing dimethyldichlorosilane, methyltrichlorosilane and phenyltrichlorosilane as monomers. Toluene or xylene, usually at the 50-% concentration was used as solvent. The mentioned above monomers were mixed at various concentrations and we conducted their hydrolysis, using water with the addition of butanol or cyclohexanol, at the temperature of  $15-20^{\circ}$ C [20]. After separation of the lower phase of acid and washing out with water, the obtained solutions of siloxanes were subjected to polycondensation by heating up in thermostat and mixture with the addition of 10-% alcohol solution of KOH as catalyst. The described process was controlled by the increase of viscosity and interrupted after reaching the defined viscosity, by the neutralization with alcohol solution of HCI [21].

The properties of the obtained varnish films were examined by pouring the steel sheets with the toluene solutions of resins. Then, we tested the time of drying at temperature of 150°C and time of hardening at 250°C. We measured the thickness of the obtained films, their hardness and resistance to bending as well as dielectric properties. We observed also their thermal resistance when heating the coatings at temperature of 250°C. We developed seven varieties of methyl phenyl silicone resins [22, 23, 24]. All the resins has a good heat resistance and adhesion to metals (excluding copper, its alloys and zinc) as well as wood and minerals but a different flexibility and rate of hardening. They were introduced to production at ZDS gradually with the demand in the period of 1969–1975.

After having found the suitability of our methylsilicone resin – *Silak M10*, the industry of paints and lacquers informed about a big demand on these products what caused the necessity of assembling a separate technical installation. As we had neither means nor the place in the ZDS in the discussed period, we adopted the offer of the Chemical Works (in 1975) which possessed the non-utilized object at that time. After assembling the necessary installation at the capacity of ca.300 t/year, the manufacture of modified methylsilicone resins, named *Silak M101* was commenced. The described object was attached to the Department of Silicones in the Chemical Works.

The necessity to employ the solvents was a certain shortcoming of our resins in comparison to e.g. polyester and alkyd resins, used in the industry of paints and varnishes. We tried to seek the possibilities of obtaining more "organic" resins, i.e. not containing the solvents; therefore, we undertook in our Institute of Industrial Chemistry – IChP (orygenized from ITS) in cooperation with the team of B.Marciniec from the Poznań University – the attempt– to synthesize butylchlorosilanes and to apply these monomers in obtaining the liquid, non-solvent containg silicone resins. We utilized trichlorosilane HSiCl<sub>3</sub>, produced at the Nitrogen Plant in Tarnów and butylenes from the Petrochemical Works in Płock and we conducted the trials to synthesize butylchlorosilanes by addition of technical butylene (mixture of n-butene and izo-butene) to trichlorosilane, with the use of platinum catalyst in reaction, e.g.:

### CH<sub>3</sub>CH<sub>2</sub>CH=CH<sub>2</sub>+HSiCl<sub>3</sub> = CH<sub>3</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>SiCl<sub>3</sub>

We tested also the possibilities of synthesizing butylmethyldichlorosilane when replacing trichlorosilane by methyl dichlorosilane CH<sub>2</sub>SiHCl<sub>2</sub>. The conducted reactions consisted in passing the gaseous butylene through the heated solutions of chlorosilanes with the addition of catalyst and cooling down the products and then, separation and purification of the obtained n-butyl and izo-butyl monomers by distillation [25]. After elaboration of the method and performing some trials at the semitechnical scale, we utilized the obtained butylsilane monomers for development of the synthesis of few varieties of butylmethyl-, butylphenyl- and butylmethylphenyl silicone resins [26]. According to the expectations, the obtained series of the new resins, with a lower content of solvents, and of the solvent-free ones was characterized by a good miscibility with varnish alkyd and polyester resins, at a smaller hardness as compared to methylphenylsilicone resins [27]. The samples of several varieties of the new resins were forwarded to Manufacturing Plant of Paints and Varnishes in Radom with the aim to try them; we offered also the mentioned products to Włocławek Factory but they were not interested in undertaking the task of elaborating newer paints, having better thermal resistance and other properties but being undoubtedly more expensive. It was also connected with undertaking marketing activity. Also, The Tarnowskie Works stopped producing trichlorosilane at the discussed period.

In 1997, we commenced the trails aiming at synthesis of methylphenylvinyl silicone resins by the method, proven in synthesis of methylphenyl silicone resins. We utilized there vinyltrichlorosilane, produced at the Unisil Enterprise in Tarnów. We developed the synthesis of few varieties of resins, differing in composition and properties [28]. The mentioned resins, as containing vinyl substituent capable of copolymerization, were utilized at our Institute in development of the method for emulsion copolymerisation with butyl acrylate, methyl acrylate, vinyl acetate or styrene. The copolymerization was catalyzed by peroxides. There were developed various siliconorganic dispersions for obtaining the paints; they were covered with the patent rights and published [29, 30].

In the years 1989–1990, we utilized the obtained methylphenylvinylsilicone resins and we were successful in their quick cross-linking after addition of polymethylhydroxysiloxane and platinum catalyst at temperature of 80–100°C. We obtained transparent soft gels with the dielectric and mechanical properties, favourable for electronics [31]. The mentioned resins were employed in the preparation of two-component fillings for microelectronic devices. They were successfully tested at the Tele and Radio Research Institute, Centre of Semiconductors - "Unitra CEMI" and at the National Institute of Telecommunications in Warsaw. The aim was to protect the semiconductor structures on the printed circuits, with the application of dropping dosim-

eters. I had the pleasure to submit our achievements in this respect during the conference in Edinburgh in 1990 [32].

In the years 2009-2010, we conducted the trails aiming at obtaining the solvent-free silicone resins, containing substituents capable of cross-linking via polymerization with the participation of peroxides. The mentioned experiments were conducted in cooperation with the research laboratory at the "Polish Silicones" Plant in Nowa Sarzyna (established in 2000, from Experimental Station and Department of Silicones in Chemical Plant)). We synthesized resins, containing additionally - apart from methyl and phenyl groups - vinyl and metacryloxypropyl groups, capable of polymerisation. They are obtained in two typical reactions: 1. hydrolytical polycondensation of solvent solution organochloro- and organoalcoksy-silanes and 2. thermal and catalytic policondensation of polyorganosiloxanoles. The obtained several ten types of resins did not contain solvents and created polymers after addition of peroxide at temperature of 100-120°C during some tens of seconds. [33].

#### Silicone rubbers

When conducting the trials of obtaining the oils with viscosities above 1000 MPa·s, we commenced (in 1967) the synthesis of liquid methylsilicone cross-linking rubbers without heating and of two-component tuber pastes [34]. There were developed the methods for synthesis of elastomers with the viscosities from few to several thousand MPa·s, with the chains terminated with silanol groups (-OH). The methods of their cross-linking, with the utilization of different metal-organic compounds and their grinding with silica and other minerals and pigment with the aim to obtain paste were tested and improved. The mentioned processes were carried out without heating, receiving the gums with a required consistency and destination. After improvement of the methods for preparation and choice of the first recipes at the Institute, the further work was conducted by the employees of the Experimental Station.

There were conducted the varieties of elastomers as fillers and sealants for electronic purposes as well as varieties of pastes for gypsum forms for moulding of different products made of wax, paraffin, epoxy or polyester resins, or of gypsum. The Experimental Station (ZDS) developed the production of the mentioned rubbers guite guickly; the discussed materials found many users. In parallel, J. Maciejewski and his team worked at the Institute in the synthesis of methylvinylsilicone rubbers with higher molecular weight [35] and on the cross-linking putties, as affected by the air humidity. The varieties of methylsilicone rubbers, containing acetoxyl groups and cross-linking due to the humidity in the air were introduced in the nineties at ZDS in a new building, after assembling the line for packing into containers. They were mainly applied as hydraulic sealant putties. High molecular methylvinylsilicone rubbers were implemented as late as in 2003, on the Chinese licence at "Polish Silicones" Plant which was established in 2002. The manufacture of the mentioned rubbers was commenced at the scale of ca. 400 per year. In the successive years, there were assembled the respective installations in the mentioned plant and the processing of the discussed rubbers into silicone gums was undertaken. It commenced the new stage of development of silicones what exceeded the frames of my paper about the beginnings of production and application of silicones in Poland.

### Auxiliary silicone aids Hydrophobic agents

Many varieties of silicones reveal hydrophobic properties what was utilized in a different way, e.g. in hydrophobization of surface or in water-resistant impregnation. We utilized methylsilicone and methylhydrosilicone oils and methylsilicone resins and we developed the products in a form of oil and resin solutions in volatile solvents or in a form of water solutions for sprinkling, pouring or coating with paint. We developed also various agents for waterproof impregnation of building materials, different porous materials, fabric, etc. Potassium methylsiliconate Ahydrosil K, as described above was a cheap and universally applied agent, especially used in construction industry. Silicone compositions, employed in hydrophobization of radio-technical ceramics were also successful and improved dielectric properties [36]. A lot of work was dedicated to elaboration and implementation trials of silicone agents for impregnation of textiles (Siltex). The mentioned processes were carried out at the Department of Silicones in the Chemical Works in Sarzyna and will be discussed later on in the present paper.

#### Anti-adhesive agents

Most of oils and many rubbers and also some silicone resins are characterized by a low surface tension and anti-adhesive properties - lack of adhesion to many materials. We described the suggestions to utilize this phenomenon in many prospects and publications. We recommended oils, pastes and oil emulsions for greasing or sprinkling moulds in the rubber and plastics industry. We developed also the special anti-adhesive agents in volatile solvents intended for plastics, rubber [37, 38], and welding industry [39]. They are successfully employed in shipyard sector. The anti-adhesive agents are very economic in use, as e.g. the emulsions for moulds are diluted to 1-2% concentration before sprinkling and they abbreviate the technological processes and decrease the losses. Similarly, the agents in aerosols occurred to be very economic and convenient. Owing to the mentioned advantages, the demand on the described means was growing. For example, after starting up the production of methylsilicone emulsions for tyres-manufacturing sector, the demand on the mentioned products was equal to ca. 200t/year in the nineties.

Methylphenylsilicone varnishes as anti-adhesive means were widely applied in coating of bakery moulds [40] what will be discussed later on in the present paper.

#### Anti-foaming agents

As early as in the seventies we were asked to develop the replacers of imported silicone antifoaming agents, especially by

the users of papermaking, textile and sugar industries. We elaborated the method of synthesis of branched methylsilicone oils and paste with colloidal silica and water emulsions, having the capability of foam extinguishing. The implementation trials were conducted at the Department of Silicones in the Chemical Works Sarzyna, managed by T. Romańczuk. We participated also in the attempts of application in the paper and textile industry factories. The discussed method was covered with the patent rights in Poland and in the Soviet Union [41, 42].

#### Products for health protection

Physicians and pharmacists revealed interest in silicones as early as at the beginning of our trials at ITS. We sent the samples of methylsilicone oil for treatment of burns as well as the results of producing the protective ointments. Our methylsilicone oil emulsions were utilized in hydrophobization of the bottles for storage of preserved blood and ampoules for antibiotics. We collected literature information and made interesting contacts with the physicians; in effect, there were developed publications [43, 44] and co-authorship in monograph on the application of plastic materials in medicine [45].

After commencement of production of methylsilicone oils at ZDS, we obtained the consent of the Institute of Hygiene for admission of specially refined methylsilicone oils to application in medicine; they were found in the list of the products, distributed to hospitals and pharmacies. Moreover, one of cream-manufacturing plants in Warsaw undertook production of protective ointment made from our oil.

#### **Biocidal products**

In the monographs and publication on silicones, as well as in company prospects since the fifties, the opinion was repeated that silicones, and in particular, polydimethylsiloxane -containing varieties, being widely used in cosmetics and also, in medicine, are not subjected to degradation. When discussing with the organic silicone specialists from the Gdańsk Medical University, we recognized as being purposeful to undertake the studies on resistance of different varieties of polysiloxanes to the effect of certain species of common bacteria. We prepared some samples of cyclic and linear polysiloxanes with different organic substituents nearby silicone and with different molecular weight. In 1993 and 1994, the attempts to examine the effect of 6 species of common heterotrophic bacteria on the mentioned above compounds were conducted in Gdańsk Medical University under the guidance of J. Łukasiak. The analytical control of the possible changes in the composition of the samples was carried out. It was found that the most of the examined polysiloxanes was subjected to a differentiated biodegradation, dependent on their composition, structure and molecular weight and also on the species of bacteria. The results of the mentioned studies were published and submitted during several conferences [46, 47].

Our report presented during the international symposium IUPAC in Stockholm in 1997 on the degradable polymers, as

published in post-symposium materials [48] caused invitation to write a chapter on the biodegradation of polysiloxanes in volume 9 of Encyclopaedia "Biopolymers", to be published by Wiley-VCH Verlag Co. The mentioned task was perfectly implemented by J. Łukasiak [49].

During the described above tests of microbiological degradation of different polysiloxanes, we observed the evident resistance of siloxanes with the 3-chloropropyl substituents nearby Si atom to the activity of various bacteria. In 1995, we undertook the trials to synthesize some chloropropylsiloxanes and sent them to Gdańsk Medical University where the team of J. Łukasiak examined and stated their bacteriostatic effect. We developed the guidelines for synthesis of the mentioned biocides and prepared the patent application [50].

Other biocides, based upon ammonium salts of silsesquioxanes were developed in 2002–2003 by the team of J. Chojnowski at the Centre of Molecular and Macromolecular Studies in Łódź in cooperation with my team and the team of J. Łukasiak from Gdansk Medical University. Two varieties the mentioned salt were developed: one with the utilization of 3-chloropropylsilsesquioxane and n-octyl dimethylamine and the other one, with 3-aminopropylsilsesquiosane and butyl bromide or amyl bromide [51, 52].

#### Development of the silicon applications

Seventy years ago, apart from developing the technology of synthesis of basic silicone varieties and preparation of their manufacture, our important task included transfer of information about the possibilities of applications and cooperation with the users. The results of our elaborations were submitted immediately after their termination in the patent descriptions and publications. We published the summary up of our achievements, too [53, 54, 55]. In 2002, myself and my co-workers published the second monograph about silicones in which we described also certain Polish applications.

In 2002, the second monograph about silicones was published; we described therein certain Polish applications.

At our Institute, we conducted the informational activity on the applications of silicones and sent the samples from the first syntheses of silicone oils and resins to the interested units. For example, the first applications of the oils obtained in 1954 were the trials of their utilization in treatment of burns and bedsores in the hospital in Warsaw-Prague district and sealing with silicone of bottles for preserved blood and ampoules for antibiotics in the Tarchomińskie Pharmaceutical Works. We were also involved in cooperation, with the interested users, in the field of seeking for the ways of utilizing our silicones. So, after commencement of production of potassium methylsiliconate, the Station of Filters in Warsaw used the mentioned product in hydrophobization of the concrete-made objects and the Air Force Institute of Technology utilized it for protection of runways at the airports from icing. Potassium methylsiliconate was also applied in hydrophobization of the facade of our Parlament building in Warsaw. The mentioned siliconate, as diluted with water to the concentration

of ca. 5%, penetrates the porous surfaces of building materials at the depth of few millimetres and improves the heat stability of the construction and resistance to dirt. The mentioned properties are maintained at least for several years. After the start of methylsilicone resin *Silak M10* production at the Experimental Plant (ZDS) we utilized its solutions in volatile solvents for hydrophobization, especially of porous construction materials, gypsum and for building, as well.

When cooperating with the Factory of Paints and Varnishes (RAFIL) in Radom and Gdańsk, we utilized later improved *Silak M101* in production of heat-resistant and anticorrosive paints with aluminium or zinc fillers, or those with titanium white powder. We also cooperated with Radom Factory in respect of development and production of multicoloured silicone facade paints [57, 58, 59, 60].

Methylphenylsilicone resin Silak O (beside Silak M101) was mainly applied in production of highly heat resistant (in the range of 350-500°C, depending on the pigment) enamel, also with the resistance to chemicals. The mentioned enamel was destined for steel constructions and chemical apparatuses and, in composition with phthalic paints - in shipyard industry and in heating. Silak O was also employed as sealing material for connecting glass and metal elements in electric bulb holders and in mercury lamps. Another methylphenylsilicone resin, Silak 26, was used in anti-adhesive coatings, e.g. for varnishing of moulds for plastic castings, especially those made from epoxy resins. Based upon the knowledge obtained from German silicoorganic specialists, we suggested - to Polish experts of food industry - the application of our resins: Silak 26 and Silak 0 in varnishing of pans, saucepans and similar vessels for frying and baking without fat (Teflon was not produced yet at the discussed time) [40]. It brought considerable profits owing to the possibility of repeated baking in the lacquered moulds. In one of the waffles-producing plant, everyday covering of moulds with fat was eliminated and replaced by their cleaning and varnishing every few months.

*Silak 30*, similarly as *Silak 26*, was tested in hydrophobic and adhesive coatings and also, as electro-insulating varnish in electrical engineering e.g. in production of bulbs.

The application of silicone pastes in coating of porcelain insulators of a high voltage with the aim to protect from dirt with soot, concrete and other electro-conducting dusts, causing failures and cracking of insulators during rain or fog occurred to be very advantageous. Coating of insulators every few months with a new layer of hydrophobic paste with dielectric properties protected effectively the energetic sub-stations and lines from expensive failures and breaks in the supplies of the electric current. In Poland, the discussed application was implemented at the end of the seventies of the 20<sup>th</sup> century. The demand on *Silpasta E* being produced at the Department of Silicones in Sarzyna Chemical Plant was increased by few tons per year.

We cooperated also for some years with the Wrocław Department of the Institute of Electrotechnic and Plastic Materials Manufacturing Plant (IZO-ERG) in Gliwice. In effect, we were successful in applying *Silak 40* in the Elmor enterprise in Gdansk for electroinsulated impregnation of motor windings for ship lifts (instead of the formerly used varnish of Dow Corning company), for windings in dry transformers in Mikołowo, and also, in Elkal company in Łodź for heaters of reactors in polyester production. The development of production technology of glass-silicone oilcloths, silicone bands and shirts, with the use of Silak 50 was the important common achievement. We managed to develop a special catalyst, abbreviating the time of drying of glass textile, coated with Silak up to 3 minutes at temperature of 150°C in coating machine for production of oilcloths what enabled to undertake their production. Coating of glass bands and shirts was conducted while immersing their segments of ca. 1 m length in the solution with Silak 50 with catalyst and then, drying them in a drying machine. The obtained electro-insulating materials were characterized by a good elasticity, good dielectric properties and a perfect thermal resistance. They could withstand up to 1000 hours of heating at 250°C and met the requirements of standard of electro-insulating materials of the highest class H. The discussed materials were widely employed in construction of electric and electronic machines and devices [61]. We tried to utilize more reactive methylphenylsilicone resins: Silak 15, 20 and 31N and we developed (in cooperation with the specialists from ERG enterprise in Gliwice) the production technology of glasssilicone plates of 2-10 mm thickness and also, of the pipes rolled up under lower pressure [62].

Building and conservation of monuments belonged to the more important domains where the silicones were applied. We were also involved in this undertaking. Apart from the described above attempts of employing silicones, the solutions of methylsilicone resin in volatile solvents were the successive hydrophobic agents which were developed. They were used in 5-10% concentration for water proofing impregnation of porous materials (such as e.g. cellular concrete or plaster) and for coating of asbestos-concrete plates etc. [63, 64, 65, 66]. They were also successfully applied in waterproofing of radio engineering ceramic elements.

We developed - for the needs of conservation of monuments - the composition, containing solution of Silak M101 in petrol or inflammable volatile solvent, with the addition of a special, mechanically fortifying agent [67]. Multi-coloured silicon façade paints, produced in the Radom Factory of Paints and Varnishes, as being obtained from our Silak M101 were in 1974-1976 tested in Warsaw and received a very good evaluation of the performers and inhabitants. They were widely employed in the housing, industrial and monumental building. The mentioned paints were distinguished by non-wettability for water and good vapour permeability (in the contrary to alkyd or acryl paints). They improved thermo-insulating properties of the walls and were characterized by a perfect stability. We could observe, later on, well-preserved, clean walls, as painted 20 years ago as well as the historic relics and monuments. In the eighties, we developed cooperation with the specialists in construction and conservation of monuments [68, 69. 70] by elaborating and implementing the production of the new hydrophobizing-strengthening agents for building materials, especially for limestone, sandstone, bricks and plasters. Additionally, we developed putties and silicone glue for stones

[71] and sent the samples to be tested by the conservators. We consulted the application of the mentioned agents in Powązki cemetery and Wilanowski Palace in Warsaw, at Wawel in Cracow, at the market place in Zamość, in Toruń and in many other sites. We participated also in different exhibitions, conferences and congresses concerning the preservation of national and foreign historical monuments. I had the pleasure to participate twice (with M. Zielecka) in the conservatory work in Egypt, together with the archaeologists from the University of Warsaw and it was the most interesting form of my cooperation with the conservators. The task implemented in the years 1986 and 1987 included the strengthening waterproofing of the walls of the 4 thousand years-old grave mastaba Nefermaat in Meidum near Cairo, which was made from a mud brick, dried at the sun. To this end, our product, *Ahydrosil*, was employed.

In the eighties, we came back to the tests aimed at improving of the recipe for hydrophobic finish of textiles. After their implementation at the Department of Silicones in Sarzyna, we joined a wide application activity in several factories of the textile industry and cooperated with the specialists from some research centres [72, 73, 74, 75]. It was the interesting, few-year lasting activity, staying in the competition with the more abundant offer of experienced western companies. In effect, in spite of quite good hydrophobic effects and meeting the other suitability requirements in certain application trials, we were not able to break the monopoly of the western companies. Only singe factories of wool industry as well as mineral wool-producing enterprises employed our agents for waterproofing purposes.

### **Final remarks**

After termination of Poland's isolation in 1990 and opening of the access to the world markets, there was also changed the access of the users to a wide offer of silicones produced not only in the USA and in Europe but also in Japan, China, the South Korea, Brazil and other countries. Those interested in utilization of silicones may take advantage of advises and offers of many companies and specialists and chose in the abundant assortment of the offered silicones. Our role consisting in information and implementation of silicones lost its primary meaning.

At the end of this present publication, I would like to mention that it describes only a certain fragment of the beginnings of development and implementation of production technology and application of silicones in Poland. Apart from my team and the cooperating persons at the Institute of Plastic Materials (since 1970, at Institute of Industrial Chemistry), whose names are found in the enclosed bibliography, the research and implementation work was conducted by the employees of the Experimental Station of Silicones (ZDS) and Department of Silicones at Chemical Works in Nowa Sarzyna, transformed (in 2000) into "Polish Silicones" enterprise. The teams of siliconorganic chemists were developed at the University of Technology in Łódź and Gdańsk, Poznań University, Centre for Molecular and Macromolecular Studies in Łódź and Gdańsk Medical Academy. The scientists from these centers have been conducting their own

studies and also joined the development of production technology of silicones implemented in ZDS Nowa Sarzyna. Apart from the international siliconorganic symposia, being held every three years, there were organized, since 1956 to 2005, the symposia of Polish siliconorganic specialists (also every three years). During the mentioned meetings, the results of the research work and the new projects and tasks were submitted. In the eighties, the enterprise "Unisil" was established at the Nitrogen Industry Works in Tarnów. It was managed by G. Oczkowicz. The mentioned plant produced ethyl silicates from silicontetrachloride and ethanol at a semi-technical scale; they were destined for adhesive of moulds for casting industry. In the successive years, the team guided by B. Marciniec form the University in Poznań implemented the technology of carbofunctional silanes, with production in "Unisil" and utilization of trichlorosilane produced in the Factory in Tarnów. At the same time, "Topsil" company, situated near Warsaw and managed by T. Padee, commenced production of different silicone rubber products from the imported rubbers which is successfully continued until now.

The "Polish Silicones" enterprise, managed by A.Miazga, developed independently production of hydrophobic and silicone facade paints for building and conservation of monuments and of two-component cross-linking rubbers without heating and of rubber sealants (putties), cross-linking as affected by humidity. In the years 2001–2002, the mentioned factory started production of methylvinylsilicone rubbers (on the Chinese licence) in the quantity of ca. 400 t per year and later on, their processing into silicone rubbers. The discussed enterprise is still the main producer of silicones in Poland.

At present, the silicones are well known in our country not only for the plumbers or producers of windows, conservators of monuments and physicians but also users of cosmetics or baby pacifiers. A free market and prices decide on the choice of supplier. Undertaking the studies and a small production almost 70 years ago, the participation of our scientists and Polish silicoorganic specialists in development of technologies and applications and constant informing about the advantages of silicones have, in our opinion, contributed to their wide application and benefits for economy.

#### References

- Bażant V., Chvalovsky V., Radhousky J.: Silikony, (Silicones) WNT Warszawa 1955
- Rościszewski P., Sobiczewski Z.: Synteza bezpośrednia metylochlorosilanów (Direct synthesis of methylchlorsilanes), Przem. Chem. 1954,33,515
- [3] Sobiczewski Z., Rościszewski P.: Rozdzielanie metylochlorosilanów i wyodrębnianie monomerów (Separation of methylchlorsilanes and monomers isolation), Przem. Chem. 1954, 33, 570
- [4] Rościszewski P.: Synteza metylochlorosilanów (Synthesis of methylchlorsilanes), Tworzywa Guma Lakiery 1959, 4, 670
- [5] Rościszewski P., Zakrzewski L., Zieliński W.: Sposób prowadzenia syntezy bezpośredniej metylochlorosilanów (Procedure of direct synthesis of methylchlorsilanes), Pat. polski 44 883 (1959)
- [6] Rościszewski P.: Silikony tworzywa przyszłości (Silicones materials for future), Przem. Chem. 1954, 33, 153

- [7] Rościszewski P.: Silikony (Silicones), Młody Technik 1957, nr. 2, 18
- [8] Rościszewski P.: Impregnacja wodoodporna tkanin preparatami silikonowymi (Textile water proofing with silicone agents), Przem. Chem. 1957, 36, 567
- [9] Rościszewski P.: Nowe kierunki rozwoju chemii związków krzemoorganicznych (Some new directions of the silicone compounds chemistry), Przem. Chem. 1959, 38, 272
- [10] Rościszewski P.: Nowe metody produkcji monomerów silikonowych, (Some new methods of silicone monomers production), Tworzywa Guma Lakiery 1960, 5,16
- [11] Rościszewski P: Issledowanije priamowo sinteza metiłchłorsilanow (Investigation of direct synthesis of mthylchlorsilanes), Trudy Konf. Chimija i Prakticzeskije Primienije Kremnijorganiczeskich Sojedinienii, Leningrad 1961, wyp. 6, 42
- [12] Rościszewski P.: Zastosowanie silikonów (Aplication of silicones), WNT, Warszawa 1964
- [13] Tomaszewicz M., Zakrzewski L.: Oleje metylosilikonowe, (Methyl silicone fluids), Tworzywa Guma Lakiery 1958, 3, 83
- [14] Rościszewski P., Zawistowska D., Bartosiak K.: Silikonowy środek do impregnacji wodoodpornej (Silicone agent for waterpoof preparing), Pat. polski 71 486 (1970)
- [15] Rościszewski P., Zawistowska D., Wojnowski W., Bartosiak K.: Silikonowy, niewymagający wygrzewania środek do impregnacji wodoodpornej (Silicone air drying water proofing agent), Pat. polski 74 205 (1971)
- [16] Rościszewski P., Witlib R., Książek M.: Sposób wytwarzania olejów alkilowodorosilikonowych (Results for alkylhydrogen fluids obtaining), Pat. polski 92673 (1975)
- [17] Rościszewski P., Maciejewski J.: Sposób otrzymywania żywicy metylosilikonowej (Procedure of methylsilicon resins obtaining), Pat. polski 49 957 (1964)
- [18] Rościszewski P.: Untersuchungen über den Einfluss von organischen Gruppen in Siliconharzen auf ihre Eigenschaften (Studing of influence organic radicals on silicon resins properties), Chimie et Industrie 1964, 12, 251
- [19] Rościszewski P., Maciejewski J.: Methylsilicon resin with R/Si ratio 1:1, Abstr. of Intern. Symp. on Organosilicon Chemistry, Prague 1965, s.176-8020
- [20] Rościszewski P., Kocoń Z., Kohman Z.: Influence of some alkoxy radicals in methylphenyl silicon resin on their dynamic mechanical properties, Abstr. of Intern. Symp. on Organosilicon Chemistry, Prague 1965, s.181-5
- [21] Rościszewski P., Kocoń Z.: Żywice metylofenylosilikonowe, cz.
  1. Synteza (Methyl silicon resins. Part 1. Synthesis), Polimery Tworzywa Wielkocząsteczkowe 1966, 11, 123
- [22] Rościszewski P., Kozłowski A., Dul J., Strawski B., Daniszewska K.: Sposób otrzymywania stabilnych w czasie przechowywania żywic metylofenylosilikonowych (Procedure for obtaing storing stable methylphenyl silicone resins), Pat. polski 79 845(1973)
- [23] Rościszewski P., Kozłowski A.: Sposób wytwarzania żywic silikonowych (Procedure of silicon resins obtaining), Pat. polski 68 780 (1969)
- [24] Rościszewski P.: Sposób wytwarzania szybkoschnącego, elastycznego lakieru silikonowego (Procedure of obtaining of rapid drying, elastic silicone varnish), Pat. polski 59 528 (1956)
- [25] Marciniec B., Foltynowicz Z., Perkowski J., Rościszewski P., Wachowski L.: Sposób otrzymywania butylo(metylo)chlorosilanów (Procedure for butyl methylchlorsilanes obtaining), Pat. polski. 133 266 (1982)
- [26] Rościszewski P., Strojny T., Siemieńska J., Iwańska S., Kosińska W.: Synthesis and investigation of some physical and thermome-

chanical properties of butylmethylphenylsilicon resins, Abstr. of 6-th Intern. Symp. on Organosilicon Chem., Budapest 1981, s. 151

- [27] Rościszewski P., Zielecka M., Sołtysiak J., Leszczyńska I.: Butylmethylpolysiloxanes synthesis, analysis and properties, 10th Intern. Symp. on Organosil. Chem., Poznań 1993, Abstr. 247 103
- [28] Rościszewski P., Siek Z., Makosik A.: Sposób wytwarzania małocząsteczkowych, reaktywnych żywic silikonowych (Procedure for low molecular reactive silicone resins obtaining), Pat. polski 148 395 (1985)
- [29] Kozakiewicz. J., Rościszewski P., Skarżyński J., Kołdoński D.: Sposób wytwarzania błonotwórczych wodnych dyspersji silikonowo-winylowych (Procedure for film forming silicon-vinyl aquous dispersion obtaining), Pat. polski 192 183 (1999)
- [30] Kozakiewicz. J., Rościszewski P., Rokicki G., Kołdoński G., Skarżyński J., Koncka-Holand A.: Aqueous dispersions of siloxane--acrylic/styrene copolymers for use in coating, preliminary investigations, Coatings Trans. 2001, 84, B4, 301
- [31] Rościszewski P., Zielecka M.: Sposób otrzymywania silikonowego żelu elektroizolacyjngo, (Procedure for silicone electroinsulating glue obtaining), Pat. polski 164 929 (1990)
- [32] Rościszewski P., Zielecka M.: Solventless silicone resins for electronics, 9th Int.Symp. on Organosilicon Chem. Edinburg 1990. Abstr. 6, 11
- [33] Rościszewski P., Sołtysiak J., Łubkowska M., Walendziak Z.: Nowe reaktywne żywice silikonowe (New reactive silicone resins), Polimery 2015,61,4
- [34] Maciejewski J.: Zastosowanie silikonowych past kauczukowych do klejenia i uszczelniania, (Silicone elastomeric compouds for bonding and sealin aplication) Polimery 1968.13,nr.9 i 10
- [35] Maciejewski J., Sadowska W., Michalski M.: Próby otrzymywania kauczuków silikonowych, (Test of silicone elastomers obtaining-Polimery 1982,27, 24
- [36] Bancer S., Rościszewski P.: Sposób wytwarzania warstwy o dużej rezystencji na izolatorze szklanym lub ceramicznym,(Procedure for film with high resistance forming on glass or ceramic insulator Pat. polski 82 310 (1976)
- [37] Rościszewski P., Maciejewski J.: Silikonowe środki przeciwprzyczepne dla przemysłu gumowego, tworzyw sztucznych i metali (Silicon abhesive agents for rubber and plastic industry and for metal industry), Polimery 1968, 13, 508
- [38] Rościszewski P., Maciejewski J.: Sposób wytwarzania silikonowych środków do wyrobu papierów antyadhezyjnych (Procedure for production of silicone agents abhesive papers), Pat. polski 124 078 (1967)
- [39] Rościszewski P., Maciejewski J.: Środek przeciwprzyczepny dla spawalnictwa (Abhesive agent for welding), Pat. polski 60 398 (1967)
- [40] Rościszewski P.: Silikony rewelacyjne nowe rodzaje tworzyw sztucznych (Silicones a famous new kind of plastics), Przegląd Piekarniczy i Cukierniczy 1968, 2, 44
- [41] Rościszewski P., Jagielska E., Fusiek S.: Sposób wytwarzania silikonowych środków przeciw pienieniu (Procedure for antifoam agents obtaining), Pat. polski 96 102 (1975)
- [42] Jagielska E.,Rościszewski P.,Fusiek S.: Sposob połuczenija poliorganosiłoksanow, (Method of polyorganosiloxanes obtaining), Opisanije Izobretenija 791 252 (19.08.76)
- [43] Rościszewski P.: Silikony w farmacji i medycynie (Silicones in Pharmacy and Medicine) Biuletyn Inst.Farm, 1956,Nr.3
- [44] Rościszewski P.: Silikony polskiej produkcji dla medycyny (Polish silicones for medicine), Polimery w medycynie 1971, 1, 67
- [45] Praca zbiorowa Tworzywa sztuczne w medycynie (Plastics in medicine), WNT Warszawa 1970

- [46] Rościszewski P., Iwańska S., Łukasiak J., Galiński J., Dorosz A., Wiśniewska K., Szponar M.: Biodeganaretion of Polyorganosiloxanes, Industr. Chem. Res. Int. Annual Rep. 1994, 94,59
- [47 Rościszewski P., Iwańska S., Sołtysiak J., Cholińska M., Łukasiak J., Galiński J., Dorosz A., Wiśniewska K., Szponar M.: Preliminary studies of biodegration of some polyalkylsiloxanes, Inter. Conf. "Silicone Containing Polymers", Canterbury 1994, Abstr.P25
- [48 Rościszewski P., Łukasiak J., Galiński J., Dorosz A., Szponar M.: Biodegration of polyorganosiloxanes, Macromol. Symp. Stokholm 1998, 130, 337
- [49] Łukasiak J., Galiński J., Rościszewski P.: Biodegradation of Silicones (Organosiloxanes), Encyclopedia Biopolymers, Wiley--VCH Verlag GmbH, Muenster 200250
- [50] Rościszewski P., Łukasiak J., Galiński J., Szponar M.: Sposób wytwarzania polialkilosiloksanów o własnościach bakteriostatycznych (Procedure for bakteria-static polyalkylsiloxanes obtaing), Pat. polski 181 411 (1995)
- [51] Chojnowski J., Fortuniak W., Rościszewski P., Łukasiak J., Warel W., Hałas R., Kamysz W.: Środki krzemoorganiczne hamujące rozwój szczepów, bakterii, grzybów i innych drobnoustrojów oraz sposób ich wytwarzania (Siliconorganic agents for preserving of bacteria races, mushroom and microorganism developnes and procedure for their obtaining), Pat. polski 204 957 (2005)
- [52] Chojnowski J., Fortuniak W., Rościszewski P., Werel W., Łukasiak J., Kamysz W.: Polysilsesquioxanes and Oligosilsesquioxanes Substituted by Alkylammonium Salts as Antibacterial Biocydes, J. Inorg. a. Organometal. Polym. a.Mater. 16, 219, (2006)
- [53] Rościszewski P., Zielecka M.: Silikony nadal tworzywami przyszłości (Silicones are stil plastics for future), Polimery 1992,37,499
- [54] Rościszewski P., Maciejewski J., Zielecka M., Miazga A., Pączka J., Porcja.I.:Udział prac badawczych i produkcji doświadczalnej IChP w rozpowszechnianiu zastosowań silikonów w Polsce, (Participation of investigation and pilot plant production on silicone application in Poland), Polimery 1997,42,288
- [55] Rościszewski P.:Żywice silikonowe- 6 0 lat badań w Instytucie Chemii Przemysłowej ,(Silicone resins -60 years investigation in Industrial Chemistrym Institute) Polimery 2012,57,399
- [56] Rościszewski P.,Zielecka M.:Silikony,właściwości i zastosowanie,(Silicones, properties and aplication) WNT,Warszawa 2002
- [57] Rościszewski P., Kraszewski W., Kielska B., Klochowicz J., Chyliński W.: Barwna szybkoschnąca farba silikonowa, zwłaszcza dla budownictwa, (Coloured air drying silicone paint especially for building), Pat. polski 115 656 (1978)
- [58] Rościszewski P., Aleksandrowicz S.: Siliconanstrichstoffe fuer Bauindustrie und einige Anwendugsbeispiele (Silicone pains for buildings and some examples of aplication), Plaste u. Kautschuk 1977,24,361
- [59] Rościszewski P.: Erfahrungen und Ergebnisse bei Anwendung von farbigen Fassadenanstrichstoff "Silema B" (Some experiments and aplications results of "Silema B" varnish), Plaste u. Kautschuk 1978, 25, 7
- [60] Rościszewski P., Zielecka M.: Silikonowe farby elewacyjne (Silicone plaster paints for buildings), Mat. Bud. 1981 nr 17/18, 4
- [61] Rościszewski P.: Silikonowe materiały elektroizolacyjne (Silicon electroinsulating materials), Przegl.Elektr. 1969, 45, 491
- [62] Rościszewski P., Kozłowski A., Daniszewska K.: Szybkoutwardzalna dwuskładnikowa kompozycja silikonowa do laminatów

szklanych (The rapid curing silicone composition for glass--silicone laminates), Pat. polski 73 879 (1971)

- [63] Rościszewski P., Zielecka M.: Silikonowe środki hydrofobowe dla budownictwa (Silicone waterproof agents for building), Mat. Bud. 1981, nr 12, 88
- [64] Rościszewski P., Zielecka M.: Podstawy teoretyczne i wytyczne praktyczne hydrofobizacji materiałów budowlanych (Teoretical baseand practical directions for building materials waterproofing), Mat. Bud. 1990, nr 17, 7
- [65] Rościszewski P., Zielecka M.: Rezultaty stosowania silikonowych środków hydrofobowych i farb elewacyjnych w budownictwie (Results of silicone waterproof repelents and plaster paints aplication in building), Mat. Bud. 1982,7,14
- [66] Rościszewski P., Czech Z., Janak W., Raysz E., Kozak B.: Sposób otrzymywania zaprawy krzemoorganicznej zwłaszcza do tynków (Procedur for siliconeorganic mortar especially for plasters), Pat. polski 167484 (1990)
- [67] Rościszewski P., Zielecka M., Bala W.: Silikonowy środek hydrofobizująco-wzmacniający dla budownictwa, zwłaszcza dla konserwacji zabytków (Silicone waterproof and strenghtening agent for buildings and especially for conservation ancient buildings), Pat. polski 142 912 (1985)
- [68] Rościszewski P., Zielecka M., Borkowski J., Daszewski W., Cyruchin K.: Sposób konserwacji zabytkowych obiektów architektoniczno-budowlanych i artystycznych (Procedure for conservation of architecture, building and artistic monuments), Pat. polski 142 912(1984)
- [69] Rościszewski P., Zielecka M.: Anwendung silicium-organischer Produkte zur Konservierung von alten Gebäuden und Denkmalen (Aplication of silicone-organic compounds for old buildings and monuments), Plaste u. Kautschuk 1989, 36, 319
- [70] Rościszewski P., Kielska B.: Fortschritte in der Produktion und beim Einsatz von Silikonanstrichstoffen und Bautenschutzmitteln im Bauwesen in VR Polen (Progres in manufacture and aplication of silicone paints and waterproof agents for buildings in Poland), Plaste u. Kautschuk 1982, 29,488
- [71] Rościszewski P., Zielecka M., Siek Z., Makosik A.: Klej krzemoorganiczny zwłaszcza do materiałów mineralnych i ceramicznych (Organosilicone adhesive especially for mineral and ceramic materials), Pat. polski 155517 (1988)
- [72] Zielecka M., Rościszewski P., Cyruchin K., Staniak H., Byrska A., Gęga H.: Sposób hydrofobowego wykańczania tkanin w środowisku wodnym (Procedure for hydrophobic textile finish in water milieu), Pat. polski 135 741 (1983)
- [73] Zielecka M., Rościszewski P., Cyruchin K.,Byrska A., Gęga H: Sposób antypilingowego wykończanie płaskich wyrobów włókienniczych (Procedure for antypiling finish of plat textile products), Pat. polski 138 676 (1983)
- [74] Zielecka M., Rościszewski P., Moraczewski A., Porcja I., Kupiec S.: Dyspersja krzemoorganiczna do impregnacji zwłaszcza włókien szklanych (Organosilicon dispersion especially for glass fibers impregnation), Pat. polski 155 509 (1988)
- [75] Zielecka M., Rościszewski P., Lendzion A., Cyruchin K., Byrska A., Gęga H., Hodzijewicz F., Dąbrowska L., Jaworska J.: Emulsja impregnacyjna do wykończenia wyrobów włókienniczych, (Impregnative emulsion for textile materials finishing), Pat. polski 163 090 (1990).

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### THREE-PHASE CURRENT \_

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# THE POLE, WHOM WE OWE THREE-PHASE CURRENT

### POLAK, KTÓREMU ŚWIAT ZAWDZIĘCZA PRĄD TRÓJFAZOWY

**Summary**: The article presents the profile and achievements of Michał Doliwo-Dobrowolski, a Polish inventor born in Russia. He played an outstanding role in the history of world electrical engineering at the turn of the 19<sup>th</sup> and 20<sup>th</sup> centuries. He was a pioneer of the three-phase current technique. In 1888, he constructed a sensational, easy to use, cheap to produce and operate, the world's first three-phase squirrel-cage induction motor. The patent, filed on March 8, 1889, started a new era in electricity, which continues to this day, the era of alternating current.

Keywords: three-phase current, electricity, three-phase generator, three-phase motor, phasemeter, frequency meter, three-phase transformer

#### The man, to whom various nationalities are ascribed

Three-phase current is nowadays generally used all over the world. It has numerous advantages; therefore, it is applied universally - especially, for transfer of electric energy to long distances. Only some people, however, know that Michał Doliwo-Dobrowolski was the creator of the idea of three-phase current generators, electrical transformers, transfer lines, and even presently universally employed three-phase engines. When describing his silhouette and achievements, the author of the present development will stubbornly state that he was the Pole. In many elaborations concerning the history of electric engineering, he has been described as the German because in Germany he made his memorable discoveries and presented them on public forum as the representative of German institutions. Moreover, he had initially strong connections with Russia as his mother, Olga Mihajlovna, was the Russian. The future inventor was born in 1862 in Gatczyn near Sankt Petersburg, so the language of his childhood was Russian. However, his father, Józef, was undoubtedly Polish nobleman (Doliwa coat of arms). We may also put here a small question mark at this point because the father of the future genius served in the Russian army, was a colonel and participated in the Crimean War. He did not expose too much his Polish origin as it was an obstacle in his career. However, in the time of challenge, when being the Pole caused repressions to the family, he chose Polishness and emigrated from Russia.

**Streszczenie**: Artykuł przedstawia sylwetkę i dokonania Michała Doliwo-Dobrowolskiego, polskiego wynalazcy urodzonego w Rosji. Odegrał on wybitną rolę w historii światowej elektrotechniki na przełomie XIX i XX wieku. Był pionierem techniki prądu trójfazowego. W 1888 r. konstruował rewelacyjny, prosty w obsłudze, tani w produkcji i eksploatacji, pierwszy na świecie trójfazowy indukcyjny silnik klatkowy. Patent zgłoszony 8 marca 1889 r., zapoczątkował nową erę w elektryce, trwającą do dziś, epokę prądu przemiennego.

**Słowa kluczowe**: prąd trójfazowy, energia elektryczna, prądnica trójfazowa, silnik trójfazowy, fazomierz, częstościomierz, transformator trójfazowy



Fig. 1. Michał Doliwo-Dobrowolski [1] Source: Deutsches Museum Bildarchiv

But it occurred later, when Michał Doliwo-Dobrowolski, as 16 years (!) old boy, commenced (in 1878) the studies at Institute of Technology in Riga.

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His initial education was based upon the Russian language as he graduated a secondary school in Odessa. He undertook the studies in Riga (in Russian language) due to a lack of alternative possibilities; purely Polish higher education institutions did not exist at that time; even in Warsaw, where there was the Russian-language **Imperial** University of Warsaw.

Probably Michał Doliwo-Dobrowolski would have graduated the University of Technology in Riga and – similarly as his father - would have made a carrier in the Russian Empire but an accident happened on 13 March, 1881 in Petersburg: Tsar Alexander II was murdered. Polish student, Ignacy Hryniewiecki was the assassinator; this fact commenced a series of repressions against the Poles. In such context, Dobrowolski (who had nothing to do with the mentioned assassination) was expelled from the University of Technology in Riga in 1881 exclusively due to the fact that he was the Pole!.

#### **Emigration to Germany**

After expelling from the Riga University, Michał Doliwo-Dobrowolski tried to continue his education as a free student in Petersburg, Odessa and Novorossiysk but everywhere his Polish descent was an aggravating factor. When he became deprived of the possibility of obtaining education and decent employment in the country of his birth, Michal Doliwo-Dobrowolski decided to emigrate.

In 1883, he moved for Germany where he immediately undertook further studies at the University of Technology in Darmstadt. At first, he studied at the faculty of mechanical engineering and then, at the newly created faculty of electric engineering. He graduated in 1884 (one year after beginning of his studies). He commenced the employment at the mentioned university as its outstanding graduate. In the period of 1884–1887, he worked under the guidance of professor of electric engineering, **Erasmus Kittler**, and was occupied in, inter alia, electric chemistry and electroplating. It did not, however, inspire him, so he made himself independent and moved to *Allgemeine Elektrizität Gesellschaft* (AEG) and stayed there for a longer period of time and created his key inven-



Fig. 2. Michał Doliwo-Dobrowolski, photo pobably taken in 1883 when he was a student in Darmstadt (Germany) [2] Source: https://www.wikiwand.com



Fig. 3. Michał Doliwo-Dobrowolski (first from the right) as a student and later an assistant to prof. Erazm Kittler (in the center) - head of the Department of Electrical Engineering in Darmstadt [3]

Source: https://de.wikipedia.org/wiki/Erasmus\_Kittler

tories. It should be stressed that Prof. Kittler made a beautiful gesture; instead of disturbing the young man to leave his faculty, he supported his efforts in AEG and gave him a very good certificate. Owing to this fact, the Director General of AEG, Emil Rathenau gave him a freedom in conducting the studies; Dobrowolski focused his attention on the problems of alternate currents. In 1888 he constructed the world's first three-phase current generator.

#### What were the advantages of three-phase current?

At this moment, we should remind the historical background. The first commercially used generators produced a **direct current**. Such current was produced by the world's first electric power plant, constructed in 1882 by **Thomas Alva Edison**. The mentioned factory delivered current to 7200 electric bulbs at Manhattan. The direct current worked and delivered profits but its transmission to bid distances was connected with the energy losses. Edison promised the reward of 50 thousand US dollars to the person, who would be able to decrease the mentioned losses.

**Nikola Tesla**, the Croatian immigrant to the USA found the solution. He became employed at Edison's (whom he admired very much!) company in 1886 and in 1887 he submitted the solution of the mentioned above problem: the losses will be radically lower when the alternating current is produced and transmitted.

Edison was a sworn enemy of alternating current, so he did not accept the solution, did not pay the reward and their paths definitely diverged. The inventory of Tesla occurred to be revelation and when he found a "sponsor, **George Westinghouse**, co-owner of Western Union Company, the alternating current began to be popular. The solutions of Tesla assumed, however, the application of one-phase current what significantly limited their suitability.

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Later on, Tesla employed two-phase current (single-phase electric engines did not want to start up) but they were not successful ideas.

On such background, the concept of **three-phase current** generator by Dobrowolski was technical revelation. Three-phase current may be generated more effectively. The three-phase generator of the same size. The transmitting three-phase line is constructed from lines which weigh and cost by 25% less than the single-phase line, transmitting the same energy. The three-phase motors are cheaper and simpler in construction as compared to the singlephase ones; they have better efficiency and lower vibrations. Three-phase transformers have a compact construction and are more convenient in comparison to the single-phase equipment. In case of three-phase system, we may introduce the current to single-phase receivers (such are found in our flats) and they may occur in 3-times higher quantities.



Fig. 5. Michail Dolivo-Dobrowolsky with colleagues at AEG in Berlin [5] Source: https://de.wikipedia.org/wiki/Michail\_Ossipowitsch\_Doliwo-Dobrowolski

### From inventory to application



Fig. 4. A three-phase engine by Michał Doliwo-Dobrowolski from 1891 [4] Source: http://apw.ee.pw.edu.pl

It is a long list of the advantages of three-phase current but there is no use to name them here; it is sufficient to say that it was the inventory of outstanding meaning in the fates of electric engineering.

#### Cascade of the inventories

A positive receipt of the achievements of Michał Doliwo-Dobrowolski in AEG company caused that "he spread the wings" and the inventories were "pouring from him". In 1889, he constructed three-phase induction engine with squirrel-cage rotor, being a prototype of most of the to-day used asynchronic engines (patent application 8.3. 1889). He obtained also some key patents on three-phase transformers (German patent no 56359 of 29.08. 1889 and the next one of 4.10. 1891). Doliwo-Dobrowolski developed new measuring instruments (phase meter and wattmeter). He found the method for extinction of discharge in high tension switches (the so-called quench chambers). The list of the inventories was long and the year 1889 was undoubtedly most abundant in the whole professional career of our countryman. But we had to wait still for the effects.... In spite of a great substantial value, the achievements of Michał Doliwo-Dobrowolski met the obstacles. The main ones were related with the practical application of the inventories, or their transfer to practice. It happens only in the fairy tales and in imagination of the laics that a genial inventory is quickly and efficiently introduced to practice "for the sake of humanity". In fact, there is one high threshold between the scientific inventory and its universally available application, i.e. economy. To make the inventory popular, it is necessary that someone might gain the profits from such popularization. Unfortunately, in the case of the discoveries of Michał Doliwo-Dobrowolski there were many who would state a loss due to the popularization of his inventories.

The authority of Edison was so high that by the end of the 19th century there were many electric power plants constructed and they produced and sold the direct current according to his technology. Until 1886, in Canada and the USA about 50 such power stations were constructed; until 1888, their number was equal to more than 200. Edison himself constructed 121 power plants in total. There were hundreds of them all over the world, including also a significant number in Germany. No wonder that energy companies did not want to close newly erected power plants and seek for other solutions, even if they were decisively the better ones. In order to get in the market with the new discoveries, AEG and Swiss Oerlikon (which also invested in the studies of Dobrowolski) decided to show his achievements during the World Electric Engineering Exhibition in 1891, held in Frankfurt/Main.

### The crucial show at the Exhibition

During the mentioned above exhibition, AEG company presented a set of different three-phase devices which caused a great interest. Michał Doliwo-Dobrowoslki himself submitted three-phase engine with power of 100 HP which was then the greatest size over the world. The mentioned engine operated the pump which served for generation of 10-m high waterfall, illuminated with the light of 1000 electric bulbs.

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But it was not the most important fact.

The sensation arose from the fact that the inventor fed the mentioned engine via the three-phase energy line of 175 km length, transmitting the current from hydro energy plant (three-phase, of course) in Lauffen. It was easy to build the power plant as well as the objects which were to be fed at the exhibition but the energetic line was a serious problem. To construct it, the whole army of telephony and telegraphy specialists was employed (they constructed earlier the overhead lines of communication conduits); 3283 pillars were erected in total on which – using ceramic insulators – there were suspended the lines, transmitting very high voltage (at the discussed time) of 15.000 V.



Fig. 6. Generator of the power plant in Lauffen am Neckar (contemporary wood engraving) [5]

Source: https://de.wikipedia.org/wiki/Michail\_Ossipowitsch\_Doliwo-Dobrowolski

The obstacles to be overcome were not only of technical nature. The people were afraid of high-voltage line and it was necessary to obtain the consent of the owners of thousands of land plots and, additionally, of the administration of four states, entering the composition of Germany! To calm down the emotions, Michał Doliwo-Dobrowolski assembled the commission (including also journalists who described it later on) before which he demonstrated personally that his energetic line was not dangerous. First, a full voltage on the wires was switched on and then, one of them was broken and the inventor himself came and took it up from the ground, using bare hands. If the residual current circuit breakers (invented by him) in power plant (used until now) had not worked, it would have been his last moment of life... It ended with success. there was no current in the wire, the automatic device reacted correctly, and Michał Doliwo-Dobrowolski survived and convinced the sceptics that his energetic line of alternating current did not pose any danger.

It should be added that more or less at the same time, Edison (the obstinate enemy of alternating current) tried to prove something completely different. To show how much the alternating current was dangerous, he arranged the shows during which he electrocuted dogs, cats, horses and cows – and even orangutan. Moreover, in January 1903, he electrocuted (6000V!) female elephant from Luna Park Zoo in Coney Island, at the presence of "audience", consisting of 1500 persons. He wanted also to perform the public execution of man (William Kemmeler, murderer, being sentenced for death on the electric chair) on electric chair but fortunately, it did not occur.

In spite of the Edison's campaign and unfavourable conditions coming from the existing energy plants, the exhibition in Frankfurt and the related international congress of electric engineers have ended with the success of three-phase current and personally, of Michał Doliwo-Dobrowolski.

# Scientific success becomes changed into business success

The exhibition in Frankfurt decided on the direction of development of electric engineering and electronics. Building of direct current and of single or two-phase power plants was abandoned. Everywhere, three-phase power plants were constructed. Michał Doliwo-Dobrowolski was the outstanding scientist and inventor but he also marked his participation in development of practical electric engineering. In 1895, he constructed the world's first three-phase water power plant on the Rhine river (in Rheinfelden), modifying principally the earlier employed generator which worked in Lauffen. The difficulty consisted in the fact that water turbines were slowly rotating and earlier constructed generators functioned well when had a quickly rotating drive (for example, of steam engine). Dobrowolski solved the mentioned problem and his construction was later copied in many power plants all over the world.

In 1897, he designed also three-phase power plants for Silesian cities Zabrze and Chorzow. It sounds nicely for Polish ears but we let's not forget that in the 19<sup>th</sup> century the mentioned cities belonged to Germany.

At the discussed period, Russia – the country of his birth, remembered the inventor. He was offered the function of the first dean in newly created, Russia's first Department of Electrical Engineering of the Petersburg University of Technology. Dobrowolski did not accept the mentioned function but consulted the matters of the seat of the Department, the program of the studies and purchase of equipment; he also donated his private collection of professional literature to the Department.

There were also noticed various signs of recognition for the achievements of Michał Doliwo-Dobrowolski. It is worthy to mention here Gold Medal of the World Exhibition in Paris and his election to three commissions in Verband Deutscher Elektrotechniker VDE (Association of German Electric Engineers). In 1908, AEG concern nominated him as a director of electrotechnical manufacturing plant in Berlin. In 1911, he was also distinguished by *honoris causa* doctorate of the University of Technology in Darmstadt.

### The success has many fathers, so the doubts were risen up

As it was mentioned above, the beginning of the 20<sup>th</sup> century was connected with the numerous words of recognition for the achievements of Michał Doliwo-Dobrowolski but also, with

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the claims that his contribution to development of electrical engineering was not so much important as was stated by his followers.

It is known that if anyone succeeds, there are always those who want to discredit the mentioned success. The dispute whether the three-phase system may be referred exclusively to the person of Michał Doliwo-Dobrowolski appeared quite often in professional literature and publicist papers at the beginning of the 20<sup>th</sup> century.

There were indicated the studies of G. Ferraris who created the theory of rotating magnetic field, being the basis for functioning of generators of motors of Dobrowolski. There was also mentioned C. Bradley, who owned the patent for a similar solution as that one by Michał Doliwo-Dobrowolski, but he has never implemented it in practice. The Americans were most obstinate and indicated the studies of mentioned above Nikola Tesla, who was oriented to the same direction but he did not create a technical solution, having a practical meaning. The dispute on the mentioned subject was settled definitely by the special VDE commission which published (in 1957) the univocal opinion, indicating Michał Doliwo-Dobrowolski as the creator of three-phase electric engineering systems.

Unfortunately, the author of the described above achievements did not see this moment. The period of the World War I deteriorated much his health. After termination of the war but before undertaking again his work at the University of Technology in Darmstadt, he became infected with the "Spanish" flu and died on 15, October 1919. But he lives in the grateful memory of all electricians!.

### References

- [1] Deutsches Museum Bildarchiv;
- [2] https://www.wikiwand.com;
- [3] https://de.wikipedia.org/wiki/Erasmus\_Kittler;
- [4] http://apw.ee.pw.edu.pl;
- [5] https://de.wikipedia.org/wiki/Michail\_Ossipowitsch\_Doliwo--Dobrowolski;

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### GROUND-SOURCE AND AIR-SOURCE HEAT PUMPS AS THE HEAT SOURCE FOR A SINGLE-FAMILY FREE-STANDING HOUSE

GRUNTOWA I POWIETRZNA POMPA CIEPŁA JAKO ŹRÓDŁO CIEPŁA DLA BUDYNKU JEDNORODZINNEGO WOLNOSTOJĄCEGO

**Summary**: In the paper, information concerning the ground and air pumps to supply a free-standing, single family house with thermal energy has been presented. The principle of functioning of the heat pumps, based upon the Carnot cycle and Linde cycle has been discussed. There was also discussed COP, i.e. Coefficient of Performance, indicating to what degree the heating equipment utilizes the heat from natural environment in relation to electric energy, supplying the heat pump. Working factors, as employed in the heat pumps, have been presented, as well.

Keywords: ground-source pump, air-source pump, Carnot cycle, Linde cycle, heat source, working liquid, bivalent system, ventilator

Streszczenie: W artykule przedstawiono informacje dotyczące gruntowej i powietrznej pompy ciepła zastosowanych do zasilania energią cieplną budynku jednorodzinnego wolnostojącego. Przedstawiona została zasada działania pomp ciepła w oparciu o obieg Carnota i Lindego, omówiono również współczynnik COP wskazujący, w jakim stopniu urządzenia grzewcze wykorzystują ciepło pozyskane ze środowiska naturalnego w stosunku do energii elektrycznej zasilającej pompę ciepła. Przedstawione zostały również czynniki robocze stosowane w pompach ciepła.

**Słowa kluczowe**: pompa gruntowa, powietrzna, obieg Carnota, obieg Lindego, źródło ciepła, płyn roboczy, układ biwalentny, wentylator

### Introduction

The heat pump is a device which collects a heat from a lower low-temperature source and transmits it upwards to the upper source with a higher temperature. The direction of the heat flow is always reverse to a natural one and requires the appropriate energy supply. We can distinguish compressor heat pumps and the absorption and adsorption heat pumps [1]. The most popular solution includes the heat pumps, equipped with the electric energy-driven compressors.

In the case of the absorptive and adsorptive heat pumps, the thermal energy necessary for working of the mentioned pumps may be produced in the process of natural gas combustion which is supplied in a liquid os gaseous form.

The lower heat source should be characterized by a sufficiently high and stable temperature; it is important as during the heat intake, its temperature should not be too much lowered; therefore, the source heat energy should be successively supplemented [2, 4]. The air is a popular lower heat source. The heat pump utilizing air as the lower heat source is the cheapest as investment but such solution becomes less effective when the air temperature drops below 5°C [3]. Under the climatic conditions, occurring in Poland, it is necessary to adapt the construction of the building, insulation, ventilation system and the method of heating the building to variable effectiveness of the work of heat pumps.

The heat pumps, collecting thermal energy from the air, as being the only one source of the buildings' heating, may be adapted to heating of the buildings, corresponding to the standards of low- or zero-energetic buildings [7, 8, 9].

Surface and ground waters are the effective source of heat for the pumps; such solution should be, however, precisely analyzed as the situations may occur when after a dry weather period, the level of water will be low and/or water will freeze in winter time [10, 11, 12].



Fig. 1. Evaluation of primary sources

From Fig. 1 it is followed that the effectiveness of the heat pump has the highest value when water is the lower heat source, however, the availability of ground water is limited.

In the case when the ground is the heat source, we may assume that the effectiveness of the heat pump and the availability of the heat source are sustainable.

In the case when the air is the heat source, its availability is unlimited but the air temperature variations may result in lowering of the effectiveness of the pump work; it is especially unfavourable at a low air temperature and simultaneous high demand on heat for warming [12, 13, 14].

In the case when water or ground are the lower heat source, the circuit of lower source should be filled by factory-made glycol solution or with brine, possessing the certificate which confirms the suitability to work in such application [15].



Fig. 2. Left-hand Carnot cycle

where:

 $T_0$  – temperature of boiling

 $T_{k}$  – temperature of condensation

 $Q_{d}$  – the supplied heat in evaporator

L<sub>s</sub> – work of compressor

 $Q_a = Q_d + L_s$  – dissipated heat in condenser

The circuit which defines the state and thermodynamic transformation of refrigerant, circulating in the heat pump is called the Linde cycle [9].

### The principle of functioning of heat pumps

The heat pump transmits the thermal energy from the lower source with a low temperature (e.g. water, ground or air) to the source, or upper source with a higher temperature (e.g. heating installation).

The set of transformations, after which the status of working liquid– after the passage of the successive conversions – comes back to the initial point, is an ideal circuit, the so-called Carnot cycle in which all transformations are reversible. In the case of heat pump, the Carnot cycle is a course of changes, illustrated in coordinates T-s, running in the counter-clockwise direction. The mentioned cycle is a left-hand Carnot cycle [10].





where:

- 1-2 isentropic compression of vapour
- 2-3 cooling of overheated vapour
- 3-4 condensed vapour
- 4-5 isentropic throttling

5-1 – boiling at the constant pressure of evaporation Po and constant temperature  $T_{\rm o}$ 



Fig. 4. Scheme of heat pump work

The blue line illustrates the saturated internal curve inside which the area of wet vapour is found.

In the circulation of compressor heat pump, there are 4 basis elements, through which the refrigerant flows successively: evaporator, compressor, condenser and expansion valve (throttling element) [10].

In the heat pumps for which water or ground is the lower heat source, the plate heat exchangers are employed; their construction ensures high efficiency of heat exchange, especially in the case of installation distributing the refrigeration factor in the exchanger. The uniform distribution of refrigerant on the whole surface of heat exchange ensures optimum utilization of the evaporator's surface [10, 11, 12].



Fig. 5. Heat exchanger without distributing installation [6]



Fig. 6. Heat exchanger with the distributing installation [6]

The above thermographic illustration show how the refrigeration liquid is distributed in evaporator. The refrigerant without support of distribution may fill the space of evaporator unevenly; then the surface of heat exchange is ineffectively utilized. In the case of distribution of the mentioned factor, the uniform penetration of the evaporator is possible. The heat pump for which the air is the lower heat source is furnished with lamellar heat exchangers. They are characterized by a high surface on the side of the air (primary side) because the thermal capacity of the air is smaller than the heat capacity of the mixtures of glycols or brines, used in the case when ground or water is the lower source of heat [11, 8, 5, 6].

The compressors increase the pressure and temperature of the factor, flowing out from the evaporator. The electric motordriven compressors are most frequently employed; they are most convenient under the domestic conditions. In the modern heat pumps, there are used rotary and spiral compressors; vane compressors and piston compressors are older devices. The work of vane compressor is calmer as compared to piston compressor because its working part is not moving by reciprocating movement but by circulating and steady, uniform movement; it is accompanied by smaller vibrations. It is smaller in size and has smaller number of elements which are responsible for gas compression. Such features make it reliable and very stable [7, 8]. Construction of compressors has been constantly improved, aiming at betterment of their durability and energetic effectiveness.

During the condensation process, plate heat exchangers are employed at first; they are characterized by a high value of the stream of heat exchange; the other constructions of heat exchangers are employed as late as in the range of heat power above 100 kW.

The task of the expansion valve of heat pump is to lower the pressure of liquid refrigerant which after dissipation of heat to heating system in condenser remains still under a high pressure, until the pressure at which its evaporation takes place; it enables the intake of heat by refrigerant in lower heat source. The expansion valve regulates the flow of refrigerant so as the quantity of in-flowing refrigerant may completely evaporate in the evaporator and the overheated vapour passes to compressor [16, 17, 18]. To maintain the stable values of the temperature and pressure of the refrigerant before entrance to the compressor, it is purposeful to install the expansion valve, driven by thermostatic or electronic sensor [1, 2, 3, 4].

It is advantageous to employ the so-called economizer in the heat pump. The mentioned device consists of heat exchanger and additional expansion valve. The stream of working factor in a liquid form after leaving the condenser is divided into two parts: a lower part of the stream (10-20%) is directed to additional expansion valve and the exchanger of the economizer where it is heated up by the remaining part of the stream with a higher temperature which flows through the exchanger; it is injected to the compressor as the overheated vapour. The greater part of the factor's stream, after cooling down in the exchanger of the economizer, is directed to the main expansion valve and then, to evaporator [8].

The increase in temperature of the vapour in the exchanger of the economizer is higher than lowering of temperature of the factor in a liquid form. The application of the economizer protects the compressor from a flow of liquid factor and decreases the demand on driving power (intake of electric energy) of the

compressor, improves the effectiveness of the process and durability of the compressor [4, 5].



Fig. 7. Scheme of the heat pump with economizer [8]

#### Working liquid in the heat pumps

Factors R410A and R407C together with polyester oil for greasing of the compressor are most frequently used as refrigerators in the heat pumps. R-410A is the azeotropic mixture of difluoromethane (R-32) and pentafluoromethane (R-125). R-407C is the zeotropic mixture of difluoromethane (R-32), pentafluoromethane (R-125) and 1,1,1,2-tetrafluoroetane (R-134a). Both mentioned above factors are colourless and have an ethereal fragrance.

Factor R410A is employed in the heat pumps with inverter compressor (with a variable efficiency); it facilitates work of the compressor in a high spectrum of its performance and the working temperature of R410A is higher than that one of R407C [8, 9].

#### Effectiveness of COP coefficient

The effectiveness of the heat pump is measured by the coefficient of performance (COP) [2, 3]. COP is a ratio of value of thermal energy, generated in the heat pump which is used for utility purposes and value of the energy necessary for the supply of motor of the heat pump compressor.

Formula: 
$$COP = P_a / N_a$$

where:

 $P_{a}$  – heating power of the heat pump {kW],

Nel – power necessary for the drive of the compressor's motor, consumed from the electric network [kW].

The coefficient of refrigeration performance (*eng.* EER, energy efficiency rating) is a ratio of refrigeration power, obtained in the refrigerating equipment and power, consumed by the mentioned device (power of compressor); it is employed for evaluation of the efficiency of the heat pump and of refrigeration, 2].

The relationship between COP and EER of the compressorbased heat pump is simple: COP = 1+ EER. If we speak about ideal heat pump where the losses of energy do not occur, then we receive the formula of the efficiency coefficient as follows:

$$COP_{max} = T_s/T_s - T_p$$

where:

T<sub>s</sub> – is a temperature of liquid in condenser [K],

 $T_p$  – is a temperature of liquid in evaporator [K].

From the above formula it is followed that the coefficient of performance is the highest, the smallest is a difference of temperature between the evaporator and condenser of the pump.

In the case of heat pumps, the lower is the temperature of the lower source, from which the evaporator consumes the heat, the lower the performance of the system is.

Value of COP coefficient is significantly affected by energy losses which may result from friction between the elements of the compressor, friction of working liquid flowing in the heat pump and deficiencies of thermodynamic processes, occurring in the circulation of the pump; if we consider the mentioned above losses, we may assume that the real value of Cop is by a half lower than the theoretical value [4, 5].

The efficiency of the heat pump is one of the most important criteria of the choice of specified equipment, determining the costs of its operation. The comparison of COP values is reliable when the mentioned coefficient was determined (calculated) under the same conditions for each of the equipment elements [6, 8]

#### Air-source heat pump

The installation of the air-source heat pump is relatively simple and it belongs to the advantages of the discussed equipment. The cheaper devices with a simple construction are most popular; they are not adapted to work at a low air temperature and in practice, they may be operated at the air temperature in the limits of -70 to 5oC.

There are also manufactured heat pumps which are adapted to the work at the air temperature lower than -10oC, but they are more expensive devices.

The air-source pumps which do not work effectively at subzero air temperatures should not be the only one equipment for heating of the house; on the other hand, they may be used in bivalent system for the support of the work of boiler during the heating season (in spring-autumn period); they may be also employed in preparation of domestic hot water at the period when the demand on energy for heating is small [8, 9, 10]. In such case, we may utilize cheaper devices, with lower power compressors and smaller heat exchangers [6].

The heat pumps with the traditional compressors, operating on the principle "switch on/switch out" consume more electric energy with the jumping (discrete) consumption of energy during start of compressor what results in decrease of the pump's efficiency. In most of the working conditions, the discussed above pumps produce the excess of heat what often forces the application of additional buffer tanks which take over the heat excess and then, give it back gradually to heating installation. It results from the necessity of maintaining the minimum time of the compressor's work and limiting to minimum the frequency of its start-ups [2, 2].

More advantageous solution includes the application of inverter compressors which are not switched out but change smoothly the number of their revolutions and adapt the compressor's performance to its actual demand on heat energy. It is also possible to use two compressors in one heat pump (or two-rotor compressors) what enables better adaptation of heating power to varying demand on the heat.

The application of economizer in the heat pump system facilitates also the flexible adaptation of the heat pump heating power to the demand on the heat in a building [4, 5].

When the pump operates in monovalent system, the heat power of the pump should be chosen in accordance with the planned thermal charge which should be calculated in conformity with the requirements of Standard PN-EN 1283. In the case of bivalent system, that is, work of the air pump, supported by electric heaters, the sum of the power of two operating devices should always correspond to the planned thermal charge. The power of the pump is dependent on the bivalent point, i.e. air temperature at which the second heating device, supporting the pump, is started up [12, 13, 14].

 $P_{pc}/P_{ma} = t_w - t_o/t_w - t_z$ 

where:

 $\mathsf{P}_{\mathsf{pc}}-\mathsf{is}$  the heat power of the heat pump,

- $P_{max}$  is the power of thermal charge,
- $\rm T_{\rm w}$  is the temperature of the heated accommodation,
- $T_{b}$  is the temperature of bivalent point
- $T_z$  is the external temperature

The heat pumps (air, water) are produced in the following variants:

- for assembling at home such solution is not recommended due to the fact that, the air must be supplied and extracted additionally,
- for assembling outside the house external devices of Split type; it is easier to install them and recommend at the situation when the site near the house is available, Generally, there is no problem with such possibility. Many models of Split type have the outer unit, which is suitable for fixing it to the house façade; it is also possible to place it at a certain distance from windows. To fix the pump, the even hardened surface at a small distance from the house is necessary [14, 15].

#### Ground-source heat pump

The heat contained in the ground is received using heat exchangers, most frequently with pipe construction. We may distinguish vertical and horizontal types.

Vertical ground-source heat exchanges are classified into:

single – U-pipe,

- double U-pipe
- coaxial

Horizontal ground-source heat exchangers are linear and spiral.

In Polish climate conditions, the mean air temperature during a year near the ground is equal to 7-9oC whereas the mean temperature of the earth surface has a value approximate to the annual mean air temperature what results from the effect of the following factors: heating of the ground by solar radiation, radiation of the heat from the ground and effect of geothermal heat [2, 3, 4].

The temperature in the ground is changed dependently on the season of the year e.g. the range of the temperature changes in the ground  $(t_{gw})$  at the depth  $h_{gr}$  = 1.5 m is found in the limits of 6 - 13°C [11, 12].



*Fig. 8. Run of changes in temperature of the ground during a year, depending on the season of the year and the depth of the ground* 

In order to calculate the size of vertical ground heat exchanger, we have only to know the refrigeration power of the heat pump. If we do not have such information, it is sufficient to know that it corresponds to the heat power, decreased by the power of compressor. On the other hand, if we do not have information about the power of compressor, but we possess the knowledge on the COP coefficient, the refrigeration power will be calculated according to the following formula:

$$Q_{chl} = COP - 1/COP * Q_{arz}$$

Before assembling the vertical heat exchanger, we should examine the ground on which the mentioned heat exchanger will be placed because we must determine the thickness and the coefficient of thermal conductivity  $\lambda$  of the particular ground layers, and calculate the mean value of the coefficient of thermal conductivity  $\lambda$  for the area where the pipe of the exchanger is to be placed [1, 2]. If we know the mean coefficient of the ground conductivity, we may determine the unit heating capacity g<sub>v</sub>



Fig. 9. Scheme of installation with vertical ground heat exchanger [8]

[W/m], and calculate the depth of placing the exchanger from the following formula:

$$L_o = Q_{obt} / g_v [m]$$

In the case of exchanger, consisting of the pipes arranged horizontally, the depth of 0.2 - 0.5m below the limit of freezing is considered as optimum. If water course is found at the small depth, the favourable solution includes placing the pipes just there because the heat pump will reach the higher COP coef-

ficient in such situation. The pipes of horizontal heat exchanger should be arranged in the earlier prepared excavation; they should be laid in a form of coil in the whole surface of the exchanger, with the preservation of a specified space between the neighbouring segments [6, 7, 8]. The spaces should not be lower than 0.4 m or higher than 1.2 m. They should be adapted to the type of the ground as it determines its regeneration ability. The heat capacity of the exchanger results not only from the length of the pipe but also from the size of the ground area on which the pipes are arranged [1].



Fig. 10. Installation with the horizontal ground heat exchanger [4]

# Characteristics of building, supplied with thermal energy, generated by the heat pumps

The air-source and ground-source heat pumps have been installed in a single-family building with the following characteristics: free-standing, single-family building, without cellar, with the habitable attic, with the total surface amounting to 189 m<sup>2</sup> and cubature of 625 m<sup>3</sup> and with the gable roof. The thickness of the outer walls was 40 cm together with the insulation. The discussed building is situated in the first climatic zone for which the minimum external temperature is equal to -16°C. The number of inhabitants of the mentioned building does not exceed 5 persons.



Fig. 11. A single-family, free-standing building with a gable roof [17]

# Air-source heat pump as the heat source in the single-family building

There is presented hereby the air-water heat pump with the external and internal unit of VAILLANT type VWL plus 102/3 with capacity of 10kW and COP coefficient = 2.5. It cooperates – in bivalent system – with two-function gas boiler with a closed combustion chamber of VUW 202/3-5 type, with capacity of 20kW [7, 8]. The air-source pump and the boiler are found in a storage room. The tank made of stainless steel with capacity of 175 dm<sup>3</sup> is used for storage of the domestic hot water [6, 7].

In the storage room, there was placed also a tank by Vaillant company type allSTOR VPS/3, of 300 dm<sup>3</sup> capacity for heating of water [4,5], which functions as a buffer, collecting thermal energy and allowing to limit frequency of the heat pump switching on.

The boiler starts up, supporting the work of the heat pump when the outer temperature is lowered down to -°C. When the outer temperature falls down below -12°C, the pump shuts down and the boiler remains the only one source of heat.



Fig. 12. Air-source heat pump by VAILLANT [12]

# Ground-source heat pump as the heat source in the single-family buildingl

The heat pump based upon brine and water, as manufactured by VAILLANT of type geo THERM VWS 171/3, with capacity of 17.4 kW and with tank heater for domestic hot water of type VIH R 150 by Vaillant company is operated in monovalent system [11,12]. The pump is found in the storage room. The lower source consists of 4 ground probes, being 97 m long each of them; they are made of HDPE pipes with diameter of 32 x 2.9 mm. The diameter of feed pipes is 63 x 5.8 mm (the diameters of the pipes were chosen according to the heating capacity when propylene glycol was the liquid in the circuit of the lower source (33%)). The ground installation of vertical heat exchanger was performed in conformity with the guidelines of designing and acceptance of the installation together with the heat pumps [3, 4].



Fig. 13. Ground-source heat pump manufactured by VAILLANT [12]

### Comparison of air-source and ground-source pumps which feed the installation for central heating and domestic hot water in single-family, free-standing building

From the viewpoint of the operating costs, application of the installation with the ground pump is more profitable solution. It is determined by the lower source from which the heat is taken, i.e. ground. The temperature of the ground is more stable than the temperature of the air. Therefore, the ground pump works at a small range of the changes in the temperature of liquid in evaporator and its sub-elements may be cheaper as compared to the parts of air pumps [2].

The advantages of the ground pump include a high COP coefficient amounting to 4.9 and the possibility of work in monovalent system. In the case of correctly designed and performed ground heat exchanger, the ground pump may be independent heating device, covering the demand on the heat energy in the building.

The COP coefficient of the air-source heat pump, as being equal to 2.5, is by a half lower than in the case of the groundsource heat pump. Besides it, the thermal capacity of the air pump is decreased together with the decline of the air temperature, that is, in the period when the demand on the thermal energy for heating of the house is increasing [1].

The defect of the air-source heat pump consists in the emission of the noise, which is caused by the work of outer unit (ventilator of evaporator). The level of acoustic pressure in the case of location of the external unit at the distance of 18 m from the building is equal to 34.9 dB, i.e. it is found at the limit of the permitted standard of the noise, penetrating the residential building.

The advantages of the application of the air-source heat pump include the costs of performing the whole heating system, including the air pump, operated in the bivalent system, with the double-function gas boiler. In this case, the costs are significantly lower as compared to the costs related to the ground-source heat pump.

### Bibliography

- Koczyk H.: Ogrzewnictwo praktyczne projektowanie, montaż, eksploatacja, Systherm Serwis Danuta Gazińska S.J, Poznań, 2005
- [2] Mizielińska K., Olszak J.: Gazowe i olejowe źródła ciepła małej mocy, Oficyna Wydawnicza Politechniki Warszawskiej, Warszawa, 2011
- [3] Sosnowski S., Tabernacki J., Chudzicki J.: Instalacje wodociągowe i kanalizacyjne, Instalator Polski, Warszawa, 2000
- [4] Polska Organizacja Technologii Pomp Ciepła: Wytyczne projektowania, wykonania i odbioru instalacji z pompami ciepła, Część I: Dolne źródła do pomp ciepła, Port PC, Kraków, 2013
- [5] Sękowski K., Juchnicki J.: Poradnik projektanta i wykonawcy, System Kan-therm, Warszawa, 2005
- [6] Viessman, Podręcznik architekta, projektanta i instalatora Pompy ciepła, str.28
- [7] Kosieradzki J., Pompy ciepła-kierunki rozwoju, Rynek Instalacyjny nr 7-8/2011, str.39
- [8] Viessmann, Podręcznik architekta, projektanta i instalatora-Pompy ciepła
- [9] Danfoss, Poradnik pomp ciepła Danfoss
- [10] Rozporządzenie Ministra Infrastruktury z dnia 12 kwietnia 2002r. w sprawie warunków technicznych, jakim powinny odpowiadać budynki i ich usytuowanie, stan prawny na 1 stycznia 2014r.
- Polska Norma PN- EN 12831: Instalacje ogrzewcze w budynkach. Metoda obliczania projektowego obciążenia cieplnego
- [12] Polska Norma PN- 92 B- 01706: Instalacje wodociągowe
- [13] Polska Norma PN- 92 B- 01707: Instalacje kanalizacyjne
- [14] Mistrz-instalacji.pl
- [15] Ogrzewanie.info.pl
- [16] Dartech.elk.pl
- [17] Instal.centrum.com.pl
- [18] Instal.reporter.pl

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# TECHNICAL INFORMATION PORTAL The largest database of on-line publications www.sigma-not.pl

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# TECHNICAL COOLING SYSTEMS IN OFFICE PREMISES

### CHŁODZENIE TECHNICZNE POMIESZCZEŃ BIUROWYCH

**Summary:** In the paper, the characteristic of the object was presented. The problems concerning comfort which should be met by the office rooms were discussed. The questions relating to the heat profits and the methods of obtaining the mentioned profits were presented. Also, the need of ventilation of the office premises and the problems resulting from cooling of the mentioned rooms were discussed. The characteristics of the air system and their types were outlines. In the paper, two air supply systems: CAV and VAV were compared.

*Keywords*: ventilation, air-conditioning, dew point, venting system, air, dry cooling, wet cooling, heat gains

Streszczenie: W pracy została przedstawiona charakterystyka obiektu. Poruszone zostały kwestie dotyczące komfortu, jaki powinny spełniać pomieszczenia biurowe oraz zagadnienia dotyczące zysków ciepła i w jaki sposób można było te zyski ciepła pozyskać. Omówiono potrzebę wentylacji pomieszczeń biurowych, a także problemy wynikające z chłodzenia tych pomieszczeń. Zostatała przedstawiona charakterystyka systemów powietrznych, a także ich rodzaje. W pracy zostały porównanie między sobą dwa systemy powietrzne CAV i VAV.

**Słowa kluczowe**: wentylacja, klimatyzacja, punkt rosy, system wentylacji, powietrze, chłodzenie suche, chłodzenie mokre, zyski ciepłay

### Introduction

Ventilation is an organized or non-organized air exchange from a given room with the aim to its refreshing. We can distinguish natural (non-organized) and mechanical (organized) ventilation [1].

Natural ventilation is the air exchange into a fresh one in a given room, as a result of the difference in pressure, caused by difference in temperature between the internal and external air. The mentioned exchange occurs in the case of leakage and through the openings in the building, with the support of auxiliary, electric current-driven devices [2, 3]. In effect, there is no possibility of regulating the quantity of the air exchange and its parameters (temperature, humidity). Hence, there is the name: non-organized ventilation.

Mechanical ventilation is also the air exchange nut using the mechanical, electrically driven equipment [4]. It enables regulation of the amount of the supplied and exhausted air. The whole air exchange is possible due to designing of individual supply-exhaust ventilation channels; therefore, mechanical ventilation is also called the organized system [5].

On the other hand, air-conditioning is a process which gives the specified parameters to the air in a closed space, being necessary for maintaining the appropriate thermal comfort due to hygienic requirements and those resulting from the technological needs. Full conditioning takes place when the following thermodynamic processes are implemented: heating, cooling, wetting and drying [6, 7].

The dew point is the temperature which - under the given pressure and gas composition - may commence the process of condensation. In the air-conditioning, the dew point is the important factor. In the premises which are air-conditioned, there is always the need of maintaining the appropriate values of the air [even in winter time) [8]. At the external temperature of 26°C and humidity of 50%, the temperature of dew point will be equal to 15°C. We should always remember that the temperature of cooling water has not to drop below 16°C. Exceeding the mentioned temperature will commence the process of condensation - the humidity will appear on the internal partitions in the room. The change in the value of humidity in the given premise is affected also by opening of windows [9, 10]. With the air conditioning on, the windows should be not opened because it causes exceeding of the dew point in the room [9]. If the air-conditioning system will be operating above the dew point, we will have to deal with the so-called dry cooling. A high cooling performance may be obtained by a wet cooling. The temperature of cooling water will drop then below the dew point and the humidity will appear on the heat exchanger (in such situation, it is necessary to install a tray for condensate under the heat exchanger [10].

#### Air-conditioning in office premises

The main task of the air-conditioning and ventilation of the office rooms is as follows:

- maintenance of thermal comfort in a given room,
- neutralization of the resulting heat gains,
- · supply of the appropriate amount of the fresh air,
- maintaining of the set air temperature,
- maintaining of the set air humidity,
- maintaining of the set air velocity [5, 6].

When calculating the heat gains, we may determine demand on a fresh air stream which should be supplied to a given room in order to neutralize the gains of heat [8]. It concerns the amount of heat, expressed in watts [W] which should be removed from the mentioned room in order to maintain the appropriate thermal comfort.

# Heat profit resulting from the sun and transparent partition (windows)

The summer gain of heat, coming from the sun is understood as the sum of solar radiation which penetrates the room via windows and the sum of the heat stream which results from a difference between the external air temperature and that one of the inside the room. The mentioned parameters are dependent on the season of the year, hour of the day, situation of a given window in relation to the world parts as well as on latitude and atmosphere transparency [5].

$$\boldsymbol{Q}_{ok} = \boldsymbol{F} \cdot \left[ \varphi_1 \cdot \varphi_2 \cdot \varphi_3 \ast \cdot (k_c \cdot R_s \cdot l_{cmax}) + k \cdot (t_z - t_p) \right] [W]$$

where:

F - is a surface of window [m<sup>2</sup>];

 $\phi_{\!\!1}$  – is a participation of glass surface in the surface of window;

 $\phi_2$  – is a correction due to the height above the sea level;

 $\phi_{\scriptscriptstyle 3}$  – is a coefficient, considering the type of glass and of the equipment;

- $k_c$  is the coefficient of accumulation;
- $R_s$  is the coefficient of sunny area and total area;

 $I_{max}$  – is the maximum value of the radius intensity;

- k is the coefficient of window penetration [W/m<sup>2</sup>K];
- $t_z$  is the calculation temperature of external air;

 $t_n$  – is the calculation temperature at a given room.

### Profit obtained from the non-transparent partition (wall)

The summer period considers also the heat gains coming from the walls. As a result of solar radiation, the temperature of the wall surfaces is changed, so the appropriate amount of heat penetrates the room [4].

$$\boldsymbol{Q}_{\text{ść}} = \boldsymbol{F} \cdot \boldsymbol{K} \cdot \boldsymbol{\Delta}_{\boldsymbol{tc}} \left[ \boldsymbol{W} \right]$$

where:

F – is the area of the surface of non-transparent partition [m<sup>2</sup>];

K – is the coefficient of partition heat penetration, adopted as 0.2 [W/m²K];

 $\Delta_{tr}$  – is the equivalent difference of temperature [K].

### Heat gain resulting from electric light

During winter time when the solar radiation is not high, the heat gains coming from the switched electric light are also taken into consideration, [2].

$$\boldsymbol{Q}_{\boldsymbol{o}} = \boldsymbol{N} \cdot \boldsymbol{\varphi} \cdot \boldsymbol{\alpha} \cdot \boldsymbol{k} [W]$$

where

N - is the total capacity of the installed light [W/m<sup>2</sup>];

 $\phi$  – is the coefficient of coincidence;

 $\alpha$  – is the coefficient, considering the exhaust of heat via ventilated fittings;

k - is the coefficient of accumulation.

### Heat gain coming from equipment

Heat is also emitted by the equipment, installed in the room (Table 1). The mentioned values are estimated and their sum is always dependent on the time in which a given device is working [3, 4].

Tab. 1. Heat gain, coming from electric devices [8]

Type of equipment	Nominal capacity	Time of equip- ment operation	Sensible heat gains	
	[W]	[min/h]	[W]	
PC Computer	100 ÷ 150	60	100 ÷ 150	
Terminal	60 ÷ 90	60	60 ÷ 90	
Dot matrix printer	20 ÷ 30	15	5÷7	
Laser printers	800	15	200	
Plotter	20÷ 60	15	5÷15	
Scanner	180	30	90	
Copying machine	1600 ÷ 1700	45÷55	1200 ÷ 1550	
Electric typewriter	50	60	50	

### Maintaining the determined humidity, temperature and air velocity

The temperature of the appropriate value should ensure a thermal balance to a human body in the surrounding environment. The optimum value is dependent on the following factors: insulation of the cloths, physical activity of man and the season of the year [5, 6, 7].

The optimum values for summer winter periods are given in Table 2 and 3.

#### Tab. 2. Optimum values for the summer time [4]

SUMMER								
Parameter	Unit	Small physical activity	Medium physical activity	High physical activity				
Optimum values (air-conditioning)								
Temperature in a given room	[°C]	23 ÷ 26	20 ÷ 23	18÷21				
Range of relative humidity	[%]	40 ÷60						
Maximum air velocity	[m/s]	0.3	0.4	0.6				
Admitted values (ventilation)								
Temperature at heat gains up to 50 W/m <sup>2</sup> of the floor	[°C]	t <sub>z</sub> + 3						
Temperature at heat gains above 50 W/m² of the floor	[°C]	t <sub>z</sub> + 5						
Relative maximum humidity	[%]	70						

#### Tab. 3. Optimum values for winter [4]

WINTER							
Parameter	Unit	Small physical activity	Medium physical activity	High physical activity			
Temperature of a given room	[° C]	20 ÷ 22	18÷20	15 ÷ 18			
Optimum range of relative humidity	[%]	40 ÷ 60					
Minimum relative humidity	[%]	30					
Maximum air velocity	[m/s]	0.2	0.2	0.3			

#### The need of ventilating the office premises

The office premises are the place where we should have the ensured appropriate air temperature of work. To this end, air-conditioning is used. The air-conditioning system is aimed at maintaining the appropriate temperature in a given room throughout the whole year, irrespectively of the external conditions. The mentioned temperature should be always automatically regulated owing to the heater or cooler [5, 6]. It prevents from the exceeding the relative humidity  $\phi$  = 70% which is a limit of thermal comfort in the room. If we want to ensure the more precise regulation of the air humidity in the air, we speak then about the full air-conditioning system which is able to maintain automatically all appropriate conditions of microclimate [7].

When determining the minimum air stream which should be supplied to the premise depending on the number of the present persons, the standard PN-83/B-03430/Az:3 200 [1, 2] is taken into consideration. In order to keep the set the temperature and humidity parameters, when the windows are not opened, the stream of the air from the air-conditioning system should be equal to 30 m/h/. On the other hand, when the windows had been opened, the mentioned stream would amount to 20 m<sup>3</sup>/h. In the calculations of the stream of the required air, we have to consider the factors (heat gains), the higher values of which would cause

the rise of the demand on the fresh air, as being supplied to the room [3, 4, 5].

#### The problem of cooling the office premises

When choosing the system of ventilation of the office premises, we should pay attention to the following factors:

- natural ventilation will never ensure the air stream to the room for the whole year, also due to the hygienic aspects;
- in multi-storey buildings, there are big differences in the pressures, occurring at their higher floors; it is caused by the effect of wind on external partitions what may cause the impossibility of ventilating the premises at opened windows (in this case, the system of double facade may be a good solution) [4, 5];
- when the windows are opened, the street noise may be a problem;
- atmospheric contamination of the air which comes from the neighbouring streets; it is caused by sealing of the buildings and non-opening of the windows;
- heat gains coming from solar radiation, heat gains coming from the people, light and electric devices are the cause of the rise of temperature inside the premises;
- cooling of the office rooms ensures the comfort for the customers and renders the attractiveness to the building.

### The systems of cooling the office premises

The choice of the system of cooling of the office rooms should be always considered individually. It is determined by the following factors: number of storeys, size of the surface, transparent size of the surface (window), construction of the building (constructional partitions) and the number of the present persons [2, 3].

### The air systems

### CAV (Constant Air Volume) System

It is a system which supplies the air to the specified zones in a room with the constant yield but with the varying temperature of the air blow. The air is factor which shapes the microclimate in the premises. When the temperature outside is increased, the temperature of the supplied air is lowered. The regulation of the air temperature is ensured by automatic system of heater and cooler in the air handling unit [1, 2, 3].

### VAV (Variable Air Volume) System

VAV system is the solution where the air stream is variable and temperature is constant. The demand of each room zones on heat is equalized by the change in the intensity of the delivered air stream. When the temperature in the room is increased, the stream of the air is increased; when it drops, the stream is decreased. The changes in the air in the particular premises are carried out using VAV regulators, based upon the signals obtained from the rooms (temperature). Then, the change of the angle of throttle takes place [1, 2, 3].

### Single-pipe systems (CAV and VAV)

Single –pipe (one-conduit) system supplies air with the appropriate temperature and with a given flow to the premises. In the particular branches, the regulators, which will maintain the flow at the set level, may be installed. The sensor informs currently about the recommended value of the air stream. The mentioned sensor is installed in the given room [2, 4].



Fig. 1. Single-pipe system with a separate regulator of the blown and exhausted air [2]



Fig. 2. Single-pipe system with zone heaters [2]

### Double-pipe (VAV) system

Two-pipe VAV system is characterized by two air streams, running in two separate channels. Warm air flows via one channel and the cold air flows in the other one. Each of the channels is directed to each of the rooms where they are mixed in the mixing chamber. The mixing chamber is responsible for mixing of the both streams and it supplies the air with a required temperature to the air diffuser. The shortcoming of the system consists in too much developed system of ducts [2, 4].



Fig. 3. Double-pipe system [2]

### The system with direct evaporation (direct cooling)

# VRF (Variable Refrigeration Flow) /VRV (Variable Refrigerant Volume) (this last name is reserved by the producer)

The variable flow of refrigerant consists of external unit and several internal parts. The external unit is found in the room. It is composed of evaporator (exchanger of the direct evaporation of refrigerant), throttling element, ventilator and a control element [2, 4]. The external unit is the refrigeration aggregate which is equipped with condenser, compressor with varying capacity and

ventilator. If the thermal load is increased and there is a need of temperature regulating, the automatic regulation causes start of the successive compressors; owing to this fact, the change in the flow of refrigerant goes smoothly [4, 5].

#### Split system

The Split system consists of the following elements: external module (condenser), internal module (evaporator with ventilator), compressor and throttling element. The both modules are linked by two conduits in which the refrigerant flows in as form of water and gas. On the other hand, the air form the room is cooled down in evaporator, emitting the heat to the refrigerant. The gas refrigerant is compressed in the compressor (its temperature is increased) and it becomes a liquid with a high pressure. In the condenser, the heat from the refrigerant is exhausted to external air. The present gas is condensed under the high pressure and pressure are lowered. The cooled down refrigerant goes to evaporator and there it is again heated up by the internal heat and changes into gas [6, 8].



Fig. 4. Scheme of functioning of air-conditioning system of Split type [6]



Fig. 5. External unit of air-conditioning system of Split type [6]

### Multisplit

Multisplit system differs from the Split system in respect of the possibility to install up to 5 internal units. The total refrigeration capacity of such system does not exceed 16 kW. The external unit is always connected with the external unit by the individual system of pipelines. The expansion valve is situated near to the external unit (as in the case of Split system). Each of the internal unit is operated irrespectively of the other, using pilot and from the panel, installed in a given room [7, 8].



Fig. 6. Multisplit system [7]

#### System of refrigerating water

In the system of cooling water (the so-called ice water), cooling down of the air is effected in the evaporator for cooling of the liquid instead of the air cooler. It is the so-called plate or jacket-pipe evaporator, being also named water chiller. Water at a low temperature (supply; 5 - 100C, return: 10 - 160C) is distributed via the system of pipes from the source to the receivers. The receivers are fan-coil units (heat exchangers of water-air type) [8, 11].

During the transitory and winter period when the external temperature drops, it is possible to utilize the natural cold source i.e. external air. It may be implemented using the additional, built-in exchanger in the chiller, installed near air condenser. Without starting the chiller, the external low temperature will ensure cooling of the transfer medium. Such possibility is called a free cooling [11, 12].



Fig. 7. System of cooling with transfer medium with the closed three-way valve of free cooling [11]

### Fan-coil units and induction units

This name refers to the devices aiming at maintenance of a constant temperature in a given room. Water or its mixture with the refrigerant is the refrigerant in the fan-coil units. The discussed devices are installed under the window and in the suspended ceiling. The air delivered to the fan-coil units is centrally treated in ice water aggregate. We can distinguish induction units and fan-coil units [9, 10].

The induction units utilize the power of induction of the external air; they suck the room air in and direct it to the heat exchanger and then, to the blow element [8].

Fan-coil units possess a fan in which the filter may be installed before the exchanger. The fan-coil unit may be connected to the network of ventilation ducts [7, 8].



Fig. 8. The circulation of the air in the fan-coil unit [8]

#### Table 2. Comparison of the air systems [8] (bold type represents the advantages of the system)

Air systems				
CAV	VAV			
There is a possibility of regulating the air temperature	Lack of temperature regulation. There is a possibility of regulating the tem- perature by installation of individual air heaters, fed with warm water or electric energy. In such case, it is unfavourable to situate the source of warm air in the inter-ceiling space as it is connected with the feeling of cold in the vicinity of windows; it also increases the costs of installing the individual heaters.			
Favourable solutions for the objects where the constant heat gains and the constant number of persons in a room occur	Maintenance of the set parameters in many zones			
Saving of refrigeration capacity (in the case of one room)	Lack of saving of refrigeration capacity (in the case of one room)			
Impossibility to regulate the flow	The possibility to regulate the flow, depending on the need			
impossibility to regulate the now	Smooth regulation of the flow			
The regulators' work cannot be connected with the possibility of cooperation with the whole managed building (BMS – Building Management System, BMS)	Regulators' work is connected with the possibility of cooperation with the whole managed building (BMS – Building Management System, BMS)			
More expensive operation of the system	Cheaper operation of the system			
Constant power consumption by a fan due to the demand on the air at a given moment	Variable power intake by a fan			
Supply of the air to all premises, even those non-utilized	Lack of the supply of the air to all premises, even those non-utilized			
Failure to keep the set parameters in many zones	Keeping the set parameters in many zones			

### Summing up

The effective system of refrigerating the office premises is a complex matter. We cannot univocally determine which system is the best. The choice of the system is determined by several factors. The first one includes architectonic conditions of the building, i.e. the place where the offices are situated and whether it is a new building or the older one. If the building has the suspended ceilings, we may choose the air system and air-water system where the air channels may be installed in the mentioned ceilings. The air-water systems have the advantage in relation to the size of the channels of the installation. We mean here the cross-sections smaller than 1:300. The second factor which decides on the choice of the appropriate refrigeration system includes the type of the premises which will be cooled down. We must answer the question: how many persons will be staying in a given room, what temperature is to be maintained in a given room, what will be the heat gains and how many rooms will be cooled down. Maintenance of the appropriate temperature and humidity in a room must always be regulated by the sensors, installed in the rooms. The automatic system must be adapted to the existing internal and external conditions. It is important to calculate the heat gains in the design stage what is connected with the required demand of blown heat stream. We must also remember about the financial and economic questions. We must decide whether the heat gains are recognised as enough high as to install the air-conditioning system or it is sufficient to ensure the mechanical ventilation without all functions of air-conditioning. We should also remember about the natural ventilation, i.e. opening of the windows (airing of the premises), the system of double facades and utilization of glazed atriums. However, in the case of natural "cooling down", there is no possibility of regulating the values of temperature, humidity and the amount of the supplied air.

Reassuming, there is no ideal, model solution which refrigeration system is the best. We should always remember that each office building should be considered individually and in all aspects. When analyzing a given building and considering its all architectonic, technical or economic potentials, we cannot forgot about the needs of its users

#### Bibliography

- Aleksander Pełech Wentylacja i Klimatyzacja podstawy (Oficyna Wydawnicza Politechniki Wrocławskiej 2009)
- [2] Kazimierz M. Gutowski Chłodnictwo i klimatyzacja (Wydawnictwo Naukowo-Techniczne 2007)
- [3] Marian B.Nantka Wentylacja z elementami klimatyzacji (Wydawnictwo Politechniki Śląskiej, 2011)
- [4] Henryk G. Sabiniak Chłodnictwo w klimatyzacji (Politechnika Łódzka 2011)
- [5] Edward Szczechowiak Klimatyzacja z chłodnictwem, 2007
- [6] www.wentylacja.com.pl
- [7] www.instalator.pl
- [8] www.ozon.pl
- [9] www.rekuperacja.com.pl
- [10] www.instalacje.com.pl
- [11] www.pro-vent.com.pl
- [12] www.murator.pl

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### THE 27<sup>TH</sup> INTERNATIONAL SCIENCE CONFERENCE

### PROBLEMS OF ANIMAL PRODUCTION INTENSIFICATION WITH RE-GARD TO ENVIRONMENT PROTECTION, EU STANDARDS AND ALTERNATIVE ENERGY PRODUCTION, INCLUDING BIOGAS

PROBLEMY INTENSYFIKACJI PRODUKCJI ZWIERZĘCEJ Z UWZGLĘDNIENIEM OCHRONY ŚRODOWISKA, STANDARDÓW UE I PRODUKCJI ENERGII ALTERNATYWNEJ, W TYM BIOGAZU

In September, 15, 2021, the Institute of Technology and Life Sciences – the National Research Institute in Falenty had the honour to host the 27th International Scientific Conference under the patronage of the Minister of Agriculture and Rural Development, Polish Society of Biomass POLBIOM and the National Representation of PhD candidates. The subject of the conference was: "Problems of animal production intensification with regard to environment protection, EU standards and production of alternative energy, including biogas". The conference was transmitted on-line by company Your Transmissions and is available at platform YouTube (https://www.youtube.com/watch?v=yb9Tu\_ Sq4Cg).

During the Conference, there were discussed many problems concerning the sustainable development in animal and vegetal production, environment protection with the elements of infrastructure at the rural areas, limitation of GHG and ammonia emission and renewable energy. The mentioned problems were divided into the following sessions:

plenary session

- renewable energy and the modern management of farm animals, being friendly to environment and climate, and
- poster session.



Fig. 1. From the left: PhD Adam Brysiewicz (ITP-PIB), PhD Andrzej Seliga (ITP-PIB), prof. Henryk Sobczuk (ITP-PIB), prof. Wacław Romaniuk (ITP-PIB), PhD Kamila Mazur (ITP-PIB), PhD Adam Koniuszy (ZUT Szczecin), PhD Andrzej Karbowy (ZUT Szczecin)



Fig. 2. prof. Wacław Romaniuk (ITP-PIB), dr PhD Kamila Mazur (ITP-PIB)



Fig. 3: From the left: MSc. Kinga Borek (ITP-PIB), PhD Dawid Kozacki (ITP-PIB), MSc. Magdalena Bagińska (ITP-PIB)

During the duration of the Conference, the training bloc relating to research programmes had place. In the mentioned bloc, two presentations were submitted: 1- Mr Tomasz Mróz from the National Contact Point of The National Centre for Research and Development (NCBR) – "Horizon Europe – Cluster 6. Food, bioeconomy, natural resources, agriculture and environment", 2 – Aleksander Bomberski, the national Broker of Innovation – " Activity>Cooperation>as an instrument for multi-subject implementation of innovations in Polish agri-food sector".

The 27<sup>th</sup> International Scientific Conference was attended by 100 participants from Poland and abroad (Russian Federation, Belarus and Latvia) representing the following institutions:

- University of Technology in Koszalin,
- High School of Business,
- West-Pomeranian University of Technology in Szczecin,

### CONFERENCE \_\_\_\_\_



Fig. 4. dr inż. Adam Brysiewicz (ITP-PIB)



Fig. 6. From the left: prof. Henryk Sobczuk (ITP-PIB), prof. Wacław Romaniuk (ITP-PIB), PhD. Adam Koniuszy (ZUT Szczecin), PhD Andrzej Karbowy (ZUT Szczecin), PhD. Kamila Mazur (ITP-PIB),

- Warsaw University of Life Sciences in Warsaw (SGGW),
- Agricultural University in Cracow,
- University of Life Sciences in Poznań,
- Institute of Animal Production, National Research Institute in Cracow,
- West Pomeranian Agricultural Chamber,
- University of Life Sciences in Lublin,
- University of Technology in Białystok,
- University of Zielona Góra,
- Experimental Station of Varieties Evaluation in Krzyżew,
- Mazovian Advisory Centre of Agriculture
- · Ministry of Agriculture and Rural Development,
- BELAGROMECH, Mińsk, Belarus,
- the V.R. Viliams GNU VNII Kormov, Moscow, Russia (Institute of Animal Feeds),
- RUNIP "IMSKH NAN Nelarus", Mińsk, Belarus (Institute of Energetics),
- Latvian University of Agriculture, Jelgava, Latvia,
- GNU NII SH of Agricultural Scientific-Research Institute, Kirov, Russia,
- State Institute of Engineering and Economy in Knyaginino, Russia,
- Agricultural Farm Tadeusz Radzięciak,
- AGRAVES Ltd.,
- TRUSTT,
- De Heus,
- TESTMER Warszawa Ltd.,
- WOLF System Ltd.,
- Lely East Ltd.

During the Conference, 37 lectures were delivered and 19 posters submitted, including 32 presentations held in foreign languages (in Russian and in English).



Fig. 5. From the left: MSc. Kinga Borek (ITP-PIB), PhD Karolina Kolasińska (ITP-PIB), PhD Dorota Kluszczyńska (ITP-PIB)



Fig. 7. From the left: PhD Andrzej Seliga (ITP-PIB), prof. Henryk Sobczuk (ITP-PIB), prof. Wacław Romaniuk (ITP-PIB)

Thy organization of the 27<sup>th</sup> International Scientific Conference was possible owing to the following co-organizers:

- Faculty of Engineering of Renewable Energy Sources of the West Pomeranian University of Technology in Szczecin,
- High School of Agribusiness in Łomża,
- Warsaw University of Life Sciences,
- Agricultural University in Cracow,

The Sponsors of the Conference were:

- WOLF System Ltd.,
- TESTMETER Warszawa S.A.,

Media patronage:

- Journal "Polish Technical Review",
- Journal "AURA Environment Protection",
- Top Agrar Poland,
- Internet Daily B2B "Now Environment".

As a result of the received papers of the participants of the 27<sup>th</sup> International Conference, two monographs will be published as follows:

- 1. Monograph: "Improvement of agricultural production technology, including renewable energy, with consideration of the requirements of sustainable development",
- Monograph: "Problems of animal production intensification, with regard to environment protection and alternative energy production, including gas".

Institute of Technology and Life Sciences – the National Research Institute in Falenty would like to express gratitude and thanks to Co-organizers, Sponsors, Patrons and Participants for the support and active participation in the Conference!

### FINAL GALA OF THE COMPETITION "YOUNG INNOVATOR", "NUMERUS PRIMUS INTER PARES" AND OLYMPICS OF TECHNICAL KNOWLEDGE

GALA FINAŁOWA KONKURSU "MŁODY INNOWATOR", "NUMERUS PRIMUS INTER PARES" I OLIMPIADY WIEDZY TECHNICZNEJ

The Laureates of the initiatives of association movement, which addressed to the young generation, had to wait a long time for the solemn summing up of the mentioned event and receiving the rewards. It concerned the XLVII Olympics of Technical Knowledge and the XIV edition of the Competition *Young Innovator*. The pandemic Covid-19 was the reason for such delay; it changed many plans in our life, not only in the association but also in public life.

The both mentioned events were carried out and terminated in the school year 2020/2021. The Laureates and finalists received the appropriate certificates but "finis coronat opus"! All persons, involved in the above activities, i.e. participants, guardians or supervisors, organizers and all supporters and sponsors of the mentioned initiatives, with the patronizing Ministry of Education and Science (MEiN), deserved for such meeting! We should also add the summing up of the XXXOV edition of the Competition Numerus Primus inter Pares, concerning the best specialist technical journal and that one, popularizing technical knowledge and culture. Its organizer is the Association of Culture and History of Engineering (TKiHT). The summing up of the mentioned Competition was - until now held together with the end of the Olympics of Technical Knowledge (OWT). Without the knowledge carriers such as technical journals, it is difficult to reach the objectives set for the OWT - i.e. encouraging the school youth to become interested in engineering, rising up the level of technical knowledge and culture and also, to help on ensuring the inflow of the appropriate candidates for technical studies.

It is not surprising, therefore, that the Gala in Warsaw Engineer House of NOT, summing up the above mentioned initiatives was attended by the meaningful guests, including, inter alia: Artur Górecki - Director of the Department of Teaching and Manuals at MEiN and Justyna John - the main specialist in the mentioned above Department; Piotr Zakrzewski - Deputy President of the Patent Office of the Republic of Poland - the partner of the competition together with the accompanying persons from the Department of Innovations and Communications of the Patent Office of the Republic of Poland, Prof. dr hab. Andrzej Dobrowolski, PhD., -Vice-Rector of Military University of Technology (WAT); and also, the representatives of the sponsors of the Competition: Agnieszka Rudzka from Polish Oil and Gas Company (PGNiG SA), Zofia Tyszkiewicz from ADAMED Foundation, Jan Rosiński - the chief process engineer at Toruń Factory of Medical Materials (in Polish TZMO SA), it could not miss the representatives of organizers and, namely:



Ewa Mańkiewicz-Cudny – the President of FSNT-NOT; Prof. dr hab. Stanisław Wincenciak, PhD. – the Chairman of the Main Committee of OWT and Prof. dr hab. Wojciech Radomski, PhD., - Vice-Chairman of the Main Commission of OWT, dr hab. Michał Szota, Eng., – the President of the Association of Polish Inventors and Rationalisers (SPWiR) – the co-organizer of the competition **Young Innovator** and Engineer Kamil Wójcik, M.Sc. – The President of the "NOT – Information Science" company, the co-organiser of the Olympics of Technical Knowledge (OWR).

The Gala was commenced with a short speech of the President of FSNT-Not, Ewa Mańkiewicz-Cudny. Then, Director Artur Górecki read the letter of the Minister of Education and Science, Przemysław Czarnek, addressed to all participants of the solemn meeting (the letter enclosed). Piotr Zakrzewski, the Vice-President of the Patent Office of Poland took the floor.

### THE LAUREATES











After the official part, the rewards and distinctions in the competition "Young Innovator" were presented to the laureates. The Jury chose laureates in three categories: primary school, secondary school of comprehensive education (lyceum) and technical college (technikum).

*In the category: primary school*, the first place was gained by Damian Dudek and Adam Ździebko from class 6 of the Tadeusz Kościuszko Primary School in Połaniec for the project: "System of multiple use of water", developed under the guidance of Jerzy Nowak, MSc. A Special Award in this category was granted by the President of FSNT-NIT and it was Jakub Rozbicki from class IV of the Stanislaw Staszic Assembly of Post-gymnasium Schools No. 1 in Siedlce, who received this award for the project: "SOS System for older people" prepared under the guidance of Engineer Włodzimierz Michalak, MSc. In the category: secondary school of comprehensive education, the first place was gained by Patryk Górski and Gabriela Rutkiewicz from the LXXX Leopold Staff Lyceum in Warsaw for the project: "Creation of the project and a prototype of life-saving launcher "LIFE GUN", prepared under the guidance of engineer Kazimierz Okraszewski, MSc. It should be mentioned that in the mentioned category, FSNT-NOT granted also a distinction which was obtained by Paweł Michoński, Paweł Szczepański and Oskar Aleksandrowicz from the United Europe I Social Lyceum of the Social Education Society (STO) in Słupsk for the project: "Automatic extinction system in personal vehicles", developed under the guidance of Grażyna Linder, MSc.

*In the category: technical (vocational) college*, the first place was obtained by the team: Mikołaj Kolo, Mikolaj Kłakulak and Filip Szafraniak from Technical School No. 1 of the Complex of Technical

### THE LAUREATES



Schools in Ostrów Wielkopolski for the project: "Drone from AED", prepared under the guidance of Dr Paweł Sobczak.

Subsequently, the awards were presented to the authors of the projects, distinguished by the Association of Polish Inventors and Rationalisers. The complete list of the laureates is available at: *www. not.org.pl.* 

In the second part of the described ceremony, there were presented the awards of the XXXIV competition *Numerus Primus inter Pares* for the best specialist technical periodical and the journal, popularising engineering knowledge and culture. In the group of magazines, popularising the engineering knowledge and culture, the Jury granted the title of Numerus Primus inter Pares to number 3/2020 of *Przegląd Budowlany*, published by Polish Association of Building Engineers and Technicians (PZITB). In the category of specialist periodicals, the tile of Numerus primus inter Pares was granted to number 9/2020 of *Wiadomości Elektrotechniczne*, the monthly, published by SIGMA-NOT. The Jury granted also two distinctions. It was obtained by *Wiadomości Naftowe i Górnicze*, published by the Scientific-Technical Association of Engineers and



Technicians of Petroleum and Gas Industry (SITPNiG) and also, for the innovative form of on-line publication of *Polish Technical Review*, published by SIGMA–NOT.

The third part of the gala was dedicated to awarding the Laureates of *XLVII Olympics of Technical Knowledge* which was held for the first time at the electronic platform. It was attended (those who were logged-in) 1532 pupils form 207 secondary school (technicalvocational and lyceums of comprehensive education). As a result of the successive qualifications, a group consisting of 17 Laureates of XLVII was generated (Tab. 1).

It is very nice accent of the continuity of OWT that the Laureates receive the successive numbers of diplomas in the documentation, conducted since the first edition in 1975.

We express our words of congratulations to all Laureates and we thank very much to the patrons and sponsors!

Janusz M. Kowalski FSNT-NOT

### THE LAUREATES

### LAUREATES of the XLVII Olympics of Technical Knowledge in the school year 2020/2021

Mechanical-building group							
No.	First name and family name	School	Tutor	Points	Place		
1	Korneliusz OBARSKI	The Queen Jadwiga Private Secondary School in Lublin	Piotr KONONOWICZ	92	I		
2	Michał TROJANOWSKI	V Secondary School (Lyceum) in Bielsko-Biała	Andrzej KOŹMIC	90			
3	Bartosz DĄBROWSKI	The Adam Mickiewicz Secondary School in Białystok	Sławomir ŻUBER	80			
4	Igor KĘDZIERAWSKI	The Stefan Czarniecki I Secondary School in Chełm	Hanna JAROSZ	79	IV		
5	Adam STANOWSKI	The Kujawy Land I Secondary School in Włocławek	Mariusz SOBCZAK	78	V		
6	Piotr KLUBA	The A. Frycz Modrzewski VI Secondary School with Bilingual Departments in Rybnik	Grzegorz ŁOPATKA	77	VI		
7	Filip LANGIEWICZ	The Stefan Czarniecki I Secondary School in Chełm	Hanna JAROSZ	73	VII		
8	Aleksander SARZYNIAK	The Complex of Secondary Schools no 1 in Kwidzyn	Joanna POŹNIAK	71	VIII		
9	Kacper OMIELIAŃCZYK	The Duchess Anna Jabłonowska, born Sapieha II Secondary School in Białystok	Piotr CHOMIENIA	70	IX		
10	Wojciech WEREMCZUK	The Jan Kochanowski VI Secondary School with Bilingual Departments in Radom	Sławomir LICHOTA	68	X		
		Electric-electronic group					
No.	First name and family name	School	Tutor	Points	Place		
1	Szymon STRZELCZYK	The Complex of Post-Primary Schools in Kleszczów The Jan Paul II Technical College of Modern Technologies in Kleszczów	Paweł KELM	93	I		
2	Krzysztof SŁONKA	The Complex of Electronic Schools in Lublin	Teresa NOWOSAD	88			
3	Karol WILK	The Complex of Technical and Secondary Schools in Kędzierzyn-Koźle	Eliza BARON	79			
4	Wiktor NOWACKI	The Mikołaj Kopernik Complex of Schools of Telecommunication and Technology in Poznań	Stanisław SITEK	75	IV		
5	Marek PAWŁOWSKI	The Defenders of Polish Post Complex of Schools of Telecommunication and Technol- ogy in Cracow	Mirosław PASZKOWSKI	66	V		
6	Mateusz WAWRZYNIAK	The King Jan III Sobieski Complex no. 6 of Schools in Jastrzębie-Zdrój	Jacek KLUBA	65	VI		
7	Piotr SOWIŃSKI	The M. Kasprzak Complex of Schools No. 36 in Warsaw	Małgorzata NADOLNA	61	VII		

The remaining participants of the competition of the 3rd degree (central) held on 22.05. 2021 at the Military University of Technology in Warsaw, obtained the number of points, required by the rules of XLVII OWT and obtained a title of **finalist**.



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