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Ruthenium: Past and Present

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This article provides information on one of the most interesting elements in the D.I. Mendeleev Periodic Table – ruthenium, discovered 175 years ago by the outstanding Russian chemist Karl Karlovich Klaus. Its most important physical properties, a variety of oxidation states, and a tendency to form countless compounds have been noted, making it unique and indispensable in all areas of science, technology, and in society. We have taken into consideration the structure of ruthenium consumption today as well as a few prospects for its future use.

Keywords: *ruthenium, ruthenium compounds, discovery, chemical properties, application.*

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Рутений: прошлое и настоящее

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В представленной статье приведена информация об одном из интереснейших элементов Периодической системы Д.И. Менделеева – рутении, открытом 175 лет назад выдающимся русским химиком Карлом Карловичем Клаусом. Отмечены его важнейшие физические свойства, многообразие степеней окисления, склонность к образованию бесчисленного количества соединений, что обусловило его уникальность и сделало незаменимым во всех сферах науки, техники и в социальной сфере. Рассмотрена структура потребления рутения в наши дни, а также некоторые перспективы применения в будущем.

Ключевые слова: *рутений, соединения рутения, открытие, химические свойства, применение.*

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One hundred and fifty years ago, the great Russian chemist Dmitry Ivanovich Mendeleev created the Periodic Table of Chemical Elements. A brilliant scientist, Mendeleev came to the conclusion that "... the size of atomic weight determines the nature of the elements." He not only arranged the 63 chemical elements known by 1869 in the order of increasing atomic weight, but also managed to make very bold predictions about the existence of yet to be discovered elements and their properties, leaving empty cells for them. Cell number 44 was occupied by the chemical element ruthenium (translated from Latin *Ruthenia* – Russia)[1], discovered 25 years before Mendeleev's invention by the Russian chemist Karl Karlovich Klaus.

The history behind the discovery of this unique element, a representative of the platinum metal family, is quite well known and has been described in detail in literature [1–3]. Therefore, there is not much point in referring to it in detail in this article. It is sufficient to mention that many famous scientists and scientific historians both in Russia and around the world contacted K.K. Klaus. On the centennial of ruthenium's discovery in 1953, Klaus's Selected Works on the Chemistry of Platinum Metals came out in the Classics of Science series. In 1926, "Materials for the Chemistry of Platinum Metals" were published in Germany. It is impossible not to mention in this article the book by the famous chemistry historian N.N. Ushakova "Karl Karlovich Klaus," published in 1972 [4, 5]. In 1984, the monograph "The Chemistry of Ruthenium" appeared (authors E.A. Seddon, K.R. Seddon) with a volume of over 1300 pages [6].

Within the framework of the present article, it is advisable to consider the main fields of ruthenium's application due to its physical and chemical properties as well as current trends in the development of ruthenium chemistry, the interest in which is not decreasing, but actually steadily increasing.

Ruthenium, like all metals in the platinum group, combines a variety of properties: heat resistance and ductility, corrosion resistance and weldability, reflectance and emission capability, heat and electrical conductivity, and high magnetic characteristics (Table 1) [7].

Due to its unique hardness and strength, ruthenium is an ideal component for imparting the same characteristics in various alloys. As a metal with a hexagonal close-packed lattice, ruthenium is used as an alloying additive. The alloy of ruthenium and platinum is a material used to manufacture fuel cells for artificial space satellites. Ruthenium greatly enhances platinum and increases the melting temperature of heat-resistant platinum alloys (the characteristic of heat resistance is creep resistance). Heat-resistant platinum alloys are unique structural materials capable of withstanding thermal and mechanical stresses in oxidizing aggressive environments for unusually high temperatures of 70–95% of melting temperature for many hundreds and thousands of hours [8]. The concentration of ruthenium in such alloys varies from 0.1 to 10%. Heat-resistant structural materials are used in aerospace engineering, also surpassing molybdenum and tungsten alloys in oxidation resistance. The refractoriness of this platinum group metal is used in the manufacture of thermocouples capable of reaching high temperatures. The smallest proportion (0.1%) of ruthenium added to titanium increases the metal's anticorrosion properties. From 1 to 5% Ru in the main alloy is sufficient to produce high-quality low-current contacts.

The discovery of ruthenium oxide's metallic conductivity in 1962 led to a comprehensive study of the properties of various simple and mixed oxides based on elements in the platinum group [9]. In 1974, N.M. Zhavoronkov, V.B. Lazarev, and I.S. Shaplygin established a correlation between the electronic configuration and the type of conductivity in simple and mixed platinum metal oxides. A semiconductor type of conductivity is shown by oxides with an external configuration d^6 d^8, while a metallic type has a configuration of d^4 and d^5. This discovery made it possible to carry out a directed synthesis of materials with specified conductive properties and ensured unprecedented progress in electronics.

Without ruthenium oxide titanium anodes ORTA and anodes containing, in addition to ruthenium dioxide, additives of iridium oxide – ORTA-I, it would be impossible to imagine the production of chlorine and chlorine-containing compounds – disinfectants, in particular sodium hypochlorite [10] today. They have increased corrosion resistance, electro-catalytic activity, as well as good and reliable polarization characteristics.

Along with the pure metal, various ruthenium compounds are also used. Some are used as powerful oxidizing agents, while others are added to the composition of glass and enamels as dyes, and yet others have fluorescent properties. The inorganic dye called "ruthenium red" deserves special attention (Fig. 1).

It is used in anatomical studies and in histology for more contrasting isolation of the tissues studied and substances when they are studied under a microscope.

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[1] If we turn to the etymology of this name, it is important to remember that Ruthenians is the traditional Latin name for Rus, the Latin distortion of the ethnonym "Rusyns", known to us from the chronicles and "Russian Truth" of Yaroslav the Wise. In German and Danish chronicles written in Latin, the Slavs of the southern Baltic coast were called Ruthenians, i.e. Rus, Rusyns [Merkulov V.I. About one of the names of Russia and Russians in sources: *Rusin*. 2006;4(6):118-122].

Table 1. Physical and chemical properties of Ruthenium [7]

Properties	Group VIII
	Ru
Nucleus charge	44
Amount of stable isotopes	7
Base state electronic configuration	[Kr]4d75s1
Usual oxidation states	+3, +4
Ion radius, nm	0.062
First ionization energy, kJ/mol	710.13
Pauling electronegativity	2.20
Type of crystal lattice	close-packed hexagonal
Density at 20 °C, g/cm	12.45
Melting temperature, °C	2334
Boiling temperature, °C	4080
Normal oxidative potential relative to the potential of a normal hydrogen electrode at 25 °C	+0.45
Electrical resistivity at 300 K, μΩ cm	7.55
Elastic properties at 300 K, GPa	485

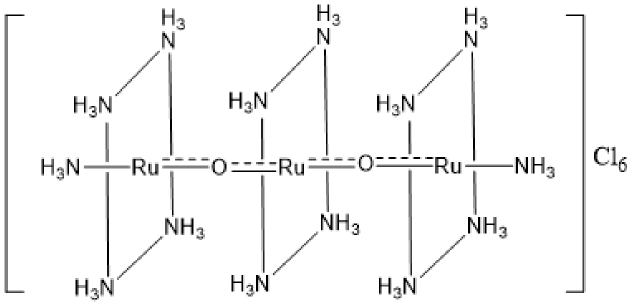

Fig. 1. “Ruthenium red” – the structure of the ammonia complex of ruthenium.

Judging by the latest market data[2], as expected from consideration of the most important properties of ruthenium, the greatest demand for this rare platinum metal is observed in industries such as electronics and electrical engineering, chemical and electrochemical industries. The total demand fluctuates at around 30 tons per year (Fig. 2).

As can be seen from the data presented, the demand for ruthenium fell by 11% due to the use of metal obtained from the processing of secondary raw materials generated in the chemical and electrochemical industries, especially in China. In 2019, according to the diagram, it grew as a result of meeting the demand for it due to available stocks. The main source of ruthenium (Clarke of ruthenium averages 5 × 10^{-7}% wt.) today are sulfide polymetallic ores, in which it is mainly present in a dispersed form. The price of ruthenium reached $250 per troy ounce (as of December 2019).

It is advisable from our point of view to pay attention to a very promising future source of ruthenium – spent nuclear fuel (SNF): among the fission products of uranium, we see a significant amount of light metals of the platinum group – Ru, Rh, and Pd mainly in the form of a mixture of stable isotopes. It is known [11, 12] that, depending on the type of reactor, the combustion mode of the fuel, and

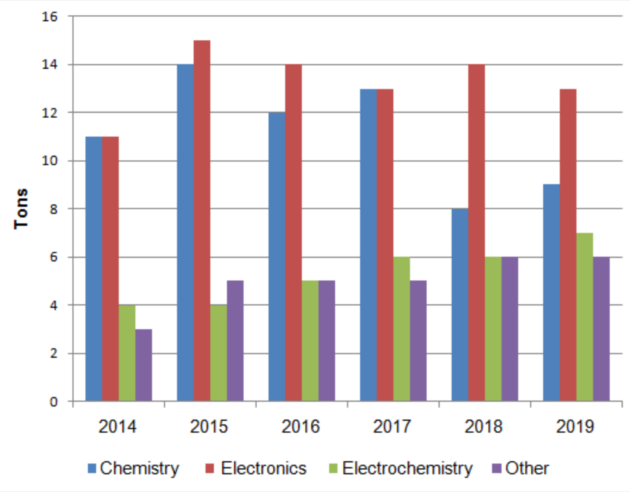

Fig. 2. Ruthenium Market Overview 2014–2018 (2019 – forecast).

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[2] http://www.platinum.matthey.com (accessed November 18, 2019).

its storage time, the concentration is from 0.7 to 29.4 g per liter of liquid waste, and the fraction of inactive ruthenium in nitric acid solutions of SNF is many times greater than the fraction of its radioactive isotopes. Therefore, current trends in improving environmental safety in the management of highly active SNF waste are the separation of platinum fragmentation metals for their subsequent transmutation and practical use.

Ruthenium is a unique element in the periodic system that has an amazing ability to manifest up to 10 different degrees of oxidation in its compounds, from –2 ($K_2[Ru(CO)_4]$) to +8 (RuO_4) (Table 2).

One of the most interesting examples is the formation of complexes with molecular nitrogen. $[RuN_2(NH_3)_5]X_2$ is the first of the known complexes obtained by Allen and Senof in 1965 by the interaction of $RuCl_3$ with hydrazine hydrate in an aqueous solution. In this case salt-like compounds of the composition $[RuN_2(NH_3)_5]X_2$ ($X = Cl^-$, Br^-, I^-, BF_4^-) [6] were isolated from the solution.

An important distinguishing feature of the chemistry of ruthenium is its ability to form nitroso complexes [6]. Nitro complexes are the only class of ruthenium compounds that are resistant to redox transformations in aqueous solutions. They have the group $(RuNO)^{3+}$, which is extremely stable even in "harsh" conditions. The discovery of the ability of nitroso complexes to enable the reversibly photoinduced transition to a long-lived metastable state provides rare opportunities for the synthesis of hybrid materials combining photoactivity with other physical properties, such as conductivity, magnetism, special optical properties, etc. [14]. Objects in which magnetic properties can reversibly change under the influence of light are of great scientific and practical interest, in particular, as materials to record information. The combination of these properties in one crystal lattice and their synergism can lead to new physical phenomena and new applications in molecular electronics.

Catalysis is one of the most common uses for platinum metals, including ruthenium. In recent decades, due to significant advances in the chemistry of coordination and organometallic compounds, for example, processes of homogeneous and heterogeneous catalysis involving catalytically active complexes "fixed" to the surface of a solid have been widely developed. Homogeneous and catalytic are the industrial processes of hydroformylation of olefins, carbonylation of alkenes, synthesis of aldehydes, etc. [6, 15]. In the electrolytic oxidation reaction of alkyl aromatic compounds, the pyridine complex Ru (IV) is used, while for the synthesis of aromatic acetaldehydes $RuCl_3$[3] is used. It was established that acidified methanol solutions of the complexes $[Ru_3(\mu\text{-}O)(CH_3COO)_6(PPh_3)_3]or[Ru_2(\mu\text{-}O)(CH_3COO)_4(PPh_3)_2]$ catalyze the hydrogenation reaction of alkenes.

It has been found [16] that the reaction for acetylene hydrochlor chlorination under conditions of homogeneous and heterogeneous catalysis selectivity (100%) and with a sufficient rate (at least 1 mol/lh h) catalyze all products formed in $RuOHCl_3$ – CH_3COOH – $(CH_3CO)_2O$, including by-products of the synthesis of the gross composition of $Ru_xO_yCl_z$, containing 40–50% ruthenium, which is of great practical importance. $[Ru_2(CH_3COO)_4Cl]$ has the highest catalytic activity in the case of direct application to activated carbon.

Table 2. Oxidation states and stereochemistry of ruthenium compounds [13]

Oxidation state	CN	Stereochemistry	Compound example
–2 (d^{10})	4	Tetrahedron	$[Ru(CO)_4]^{2-}$
0 (d^8)	5	Trigonal bipyramid	$[Ru(CO)_5]$
+2 (d^6)	4	Tetrahedron	$[RuH\{N(SiMe_3)_2\}(PPh_3)_2]$
	5	Square pyramid	$[RuCl_2(PPh_3)_3]$
	6	Octahedron	$[Ru(CN)_6]^{4-}$
	10	«Sandwich»	$[Ru(\eta^5\text{-}C_5H_5)_2]$
+3 (d^5)	6	Octahedron	$[RuCl_6]^{3-}$
+4 (d^4)	6	Octahedron	$[RuCl_6]^{2-}$
+5 (d^3)	6	Octahedron	$[RuF_6]^-$
+6 (d^2)	4	Tetrahedron	$[RuO_4]^{2-}$
	5	Trigonal bipyramid	$[RuO_5]^{4-}$
+7 (d^1)	4	Tetrahedron	$[RuO_4]^-$
+8 (d^0)	4	Tetrahedron	RuO_4

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[3] Ruthenium trichloride is well known as a catalyst for the synthesis of ammonia. The commercial drug $RuCl_3 \cdot H_2O$ is quite popular not only in catalysis, but also in preparative practice.

It was found [16] that the selectivity of ruthenium catalysts does not decrease when the CO content in acetylene exceeds 3% vol., providing the ability to work with pyrolysis acetylene.

It is impossible not to pay attention to the possibility of using ruthenium, one of the least expensive platinum group metals, in automobile exhaust gas afterburning catalysts. Ru-containing bimetallic catalysts obtained by autoclave thermolysis are effective in the oxidation of hydrocarbons [17, 18]. Table 3 shows the conversion of *n*-hexane during complete oxidation at 250–450 °C in a flow reactor using various mono- and bimetallic catalysts [17, 18]. Obviously, at 250°C on Pt, Pd, Ru, Pt–Ru, Pd–Ru and other catalysts, the conversion of *n*-hexane is ~ 50–70%, and at a temperature of ≥300 °C it exceeds 90%. In Ru catalysts, a 50% conversion of *n*-hexane is achieved at 230–250 °C, and 90% conversion is achieved at 280–335 °C, which, according to temperature characteristics, corresponds to a catalyst containing 0.2% of the mass.

It was shown [17, 18] that in the studied reaction, the Pd catalysts turned out to be significantly less active than platinum, ruthenium, and platinum-ruthenium: the temperatures of the complete conversion of *n*-hexane to them are almost 100 °C higher for each given value and are 350–370 °C and 385–390 °C, respectively, for 50% and 90% conversion. The addition of an equal amount of ruthenium to the Pd catalysts leads to a noticeable decrease in temperature (by about 50–100 °C) for both semi- and complete conversion of *n*-hexane.

Although the totality of the data presented demonstrates a significant practical effect from the use of ruthenium-based catalysts and its compounds, it certainly cannot compete with the enormous role that the achievements of the Nobel laureates in chemistry in 2001 and 2005 played and will play.

Primarily, this is a set of work carried out under the leadership of Ryōji Noyori to create chiral redox catalysts for the pharmaceutical industry. The chiral catalysis of hydrogenation reactions made possible the synthesis of enantiomers of very simple chiral molecules, which can be used as building blocks – synthons for the preparation of complex structures of natural origin [19]. Stereoselective hydrogenation methods have made available many terpenes, vitamins, amino acids, antibiotics, alkaloids, prostaglandins, and other biologically important compounds, and a number of them have begun to be produced by the industry. The scientist proposed a ruthenium (II) catalyst, BINAP (its structure is shown in Fig. 3), which hydrogenated many types of molecules with different functional groups. In particular, the Noyori complex is used as a catalyst in the production of (R)-1,2-propanediol for the industrial synthesis of the antibiotic levoflaxazine.

Moreover, the second Nobel Prize is for his contribution to the development of the metathesis

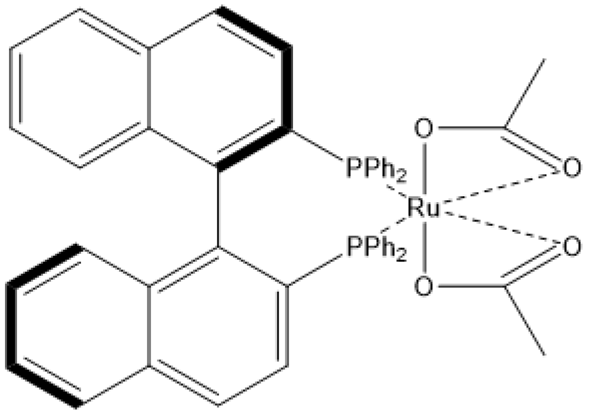

Fig. 3. Structure of the complex (R)-Ru$(OAc)_2$ (BINAP).

Table 3. Conversion of *n*-hexane during complete oxidation at 250–450 °C in a flow reactor using mono- and bimetallic Ru-containing catalysts [17]

Composition of the catalytically active phase, %	*N*-hexane conversion at temperatures, %				
	250 °C	300 °C	350 °C	400 °C	450 °C
0.2 Pt	47.2	88.2	99.5	99.9	99.9
0.1Pd	19.9	23.9	38.7	94.9	99.9
0.2Pd	8.9	27.5	51.1	99.2	99.9
0.1Ru	49.5	71.4	98.4	99.0	99.9
0.2Ru	59.1	97.8	98.6	99.0	99.9
0.1Pt–0.1Ru	47.5	94.7	97.8	98.9	99.9
0.2Pt–0.2Ru	48.0	86.6	98.6	99.4	99.9
0.1Pd–0.1Ru	4.5	92.2	98.1	99.1	99.9
0.2Pd–0.2Ru	15.4	24.0	98.9	99.2	99.9

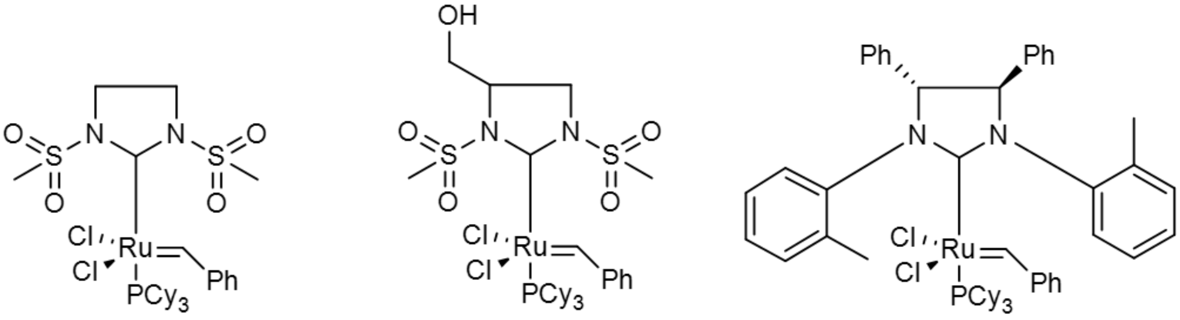

Fig. 4. Grubbs Catalysts.

method in organic chemistry. Metathesis reactions are catalyzed by ruthenium catalysts that are known as Grubbs catalysts (Fig. 4) [20]. Grubbs catalysts exhibit high reactivity in various types of metathesis and high tolerance to various functional groups. Ruthenium catalysts show low sensitivity to air, moisture, and minor impurities in solvents. They can be stored for several weeks in the open air without decomposition. Reactions with Grubbs type catalysts are often slightly slower than with ones with molybdenum, but their availability and ease of manufacture make them the most convenient for use.

In conclusion, special attention should be paid to a socially significant sphere of application for ruthenium – medicine. This should include various drugs based on ruthenium, primarily drugs for the treatment of cancer.

It should be noted that the ability to exhibit biologically active properties was stated by the great Klaus in “Neue Beiträge zur Chemie der Platinmetalle. Über das Ruthenium verglichen mit dem ihm ahnlichen Osmium” [21][4]. In it, he cited a strange — from his point of view — example of the inexplicable sensitivity of individuals to drugs of other platinum metals. In the article, he mentions the name of an assistant to the famous professor at the University of Dorpat, Ozanne, who, even after 31 years of working with platinum residues, could not enter the chemical laboratory in which platinum products were stored.

Carrying out detailed studies of the properties of ruthenium and its closest analogue, osmium, Klaus drew attention to the fact that volatile compounds of ruthenium irritate the respiratory system. In fact, the manifestation of similar and other properties of ruthenium compounds is understandable, since ruthenium in the Mendeleev Periodic Table is in the same group with iron, which are considered vital elements [22].

As a result of the study of the physicochemical and pharmacological properties of a large number of compounds synthesized to date, it was possible to identify several classes of ruthenium compounds with external sphere ligands that have antitumor activity that significantly exceeds platinum-containing preparations. In addition, ruthenium compounds are not toxic to humans, making the production of an antitumor substance based on it safe.

In 2000, clinical trials of the Ru (III) octahedral complex with imidazole (Im) and dimethyl sulfoxide (dmso) *trans*-(ImH) [Ru^{III}Cl4 (dmso)(Im)] (NAMI-A) began as the antimetastatic agent, the first of a number of ruthenium complexes (Fig. 5).

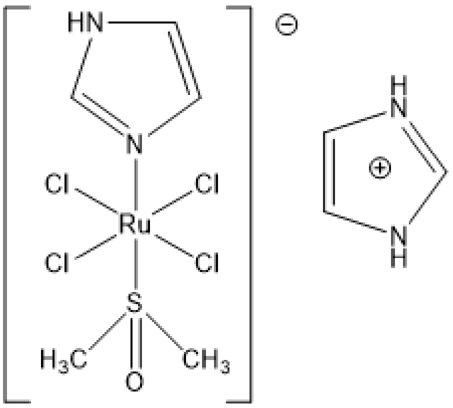

Fig. 5. Antitumor agent (NAMI-A).

We note that the ruthenium complex NAMI-A is slightly toxic to primary tumor cells and is promising in preventing the spread of cancerous tumors (metastases) [22].

Binding to biological targets is an important part of the modes of action of ruthenium complexes and organometallic compounds. As shown in the clinically

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[4] “New advances in the chemistry of platinum metals. Comparison of the properties of ruthenium and osmium” (translated from German).

studied Ru (III) complexes, they bind rapidly to serum proteins, and bind to albumin and transferrin, which is most likely especially important for selective transport into a cell. It is assumed [23, 24] that it is responsible for the selective activity of some Ru complexes and is responsible for lower side effects compared with established chemotherapeutic agents based on platinum. The latter bind relatively non-selectively to DNA, which inhibits replication and transcription and, finally, leads to the induction of controlled cell death, i.e. apoptosis. However, DNA is not necessarily the only and/or primary target for organometallic ruthenium compounds. It has recently been shown [24] that they can also act as kinase inhibitors or show preferential binding to proteins compared to DNA.

The activity and specificity of antitumor metal-containing compounds can be easily varied using the ligand environment. The introduction of a target-oriented organic fragment into the complex structure will significantly increase the antitumor properties of the compounds. In the work [24], lonidamine and bexarotene were selected as target-oriented ligands: lonidamine specifically inhibits aerobic glycolysis in cancer cells, while enhancing glycolysis in normal cells; bexarotene is known as an antagonist of the retinoid X receptor. Ru (II) compounds, leaders with high specificity for malignant neoplasms, were identified *in vivo* experiments.

This is just a small fraction of ruthenium compounds for medical applications. Both ruthenium clusters [24], and the nitroso complexes [25] mentioned above, and diruthenium complexes with amino acids [26] are being studied.

The properties of ruthenium are so unique; the chemistry of ruthenium is so multifaceted that it is impossible to cover everything in a short article, so we did not even attempt to do it. In summary, we only emphasize that the possibilities of ruthenium are endless and inexhaustible; it is unlikely to completely know about ruthenium in the future like other metals in the platinum group. Ruthenium is still waiting for its researchers, thanks to which, undoubtedly, this "Russian" elemen.

The authors declare no conflicts of interest.

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