УДК 543.422.8
DOI https://doi.org/10.32782/pcsd-2025-4-8

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Бібліографічний опис статті: Ткачук, Г., Ткачук, А., Стремецький, О., Марчук, О. (2025). Рентгенофлуоресцентне дослідження сплавів, що містять золото та срібло. *Проблеми хімії та сталого розвитку*, 4, 59–68, doi: https://doi.org/10.32782/pcsd-2025-4-8

РЕНТГЕНОФЛУОРЕСЦЕНТНЕ ДОСЛІДЖЕННЯ СПЛАВІВ, ЩО МІСТЯТЬ ЗОЛОТО ТА СРІБЛО

У роботі представлені результати дослідження хімічного складу сплавів, що містять золото та срібло, а також інші метали, із застосуванням сучасного неруйнівного методу рентгенофлуоресцентного аналізу (РФА). Актуальність дослідження зумовлена необхідністю контролю якості дорогоцінних металів на ринку ювелірних виробів, де часто зустрічаються відхилення від заявлених стандартів. Метою роботи було виявлення та аналіз фактичного складу ювелірних виробів, медалей та монет різного історичного періоду та походження.

Для дослідження було використано спектрометр ElvaX з програмним забезпеченням, за допомогою якого проведено кількісний та якісний аналіз сплавів. У ході дослідження ювелірних виробів із золота встановлено, що проба більшості проаналізованих зразків не відповідає клеймуванню. Зокрема, у сучасних виробах 585 проби вміст золота коливався в межах 58.14–58.55 %, що для деяких виробів є нижчим за мінімально допустимий стандарт. Виявлено, що для здешевлення сплавів срібло в лігатурі часто замінюють цинком, який є значно дешевшим. Також в одному зі зразків виявлені небажані домішки нікелю, який є потенційним алергеном. Аналіз срібних виробів також продемонстрував значні відхилення: вміст срібла у деяких зразках був нижчим за заявлену 925 пробу, тоді як в інших значно перевищував її.

Дослідження золотих та срібних медалей та монет підтвердило історичні особливості їх виробництва. Наприклад, золота монета часів Миколи II майже ідеально відповідає заявленій 900 пробі, а срібні полтинники викарбувані в 20-х р.р. минулого століття мають завищений вміст срібла. Аналіз сучасних монет Євросоюзу, *України та Польщі підтвердив використання спеціалізованих сплавів, таких як нейзильбер та Nordic Gold, що відповідає відомим технологіям карбування. Отримані результати підтверджують, що метод РФА є надійним інструментом для швидкого та ефективного контролю складу металевих виробів, дозволяючи виявляти невідповідності та фальсифікації, що особливо важливо як для експертних установ, так і для споживачів. Загалом, проведений аналіз демонструє високу ефективність РФА для експертного дослідження.*

Ключові слова: *рентгенофлуоресцентний аналіз, сплави золота, сплави срібла, металічні сплави, ювелірні вироби, монети.*

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To cite this article: Tkachuk, H., Tkachuk, A., Stremetskyi, O., Marchuk, O. (2025). Renthenofluorestsentne doslidzhennia splaviv, shcho mistiat zoloto ta sriblo [X-ray fluorescence study of alloys containing gold and silver]. *Problems of Chemistry and Sustainable Development*, 4, 59–68, doi: https://doi.org/10.32782/pcsd-2025-4-8

X-RAY FLUORESCENCE STUDY OF ALLOYS CONTAINING GOLD AND SILVER

This paper presents the results of a comprehensive study of the chemical composition of alloys containing gold and silver, as well as other metals, using the modern non-destructive X-ray fluorescence (XRF) analysis method. The relevance of the research is determined by the need for quality control of precious metals in the jewellery market, where deviations from declared standards are often encountered. The aim of the work was to identify and analyze the actual composition of jewellery, medals, and coins from various historical periods and origins.

For the study, an ElvaX spectrometer was used to perform quantitative and qualitative analysis of the alloys. During the analysis of gold jewellery, it was found that the fineness of most of the analyzed samples did not correspond to the hallmark. Specifically, in modern 585-fineness items, the gold content ranged from 58.14–58.55 %, which for some items is below the minimum permissible standard. It was discovered that to cheapen the alloys, silver in the ligature is often replaced with zinc, which is significantly cheaper. Additionally, an undesirable impurity of nickel, a potential allergen, was found in one of the samples. The analysis of silver items also showed significant deviations: the silver content in some samples was lower than the declared 925 fineness, while in others it significantly exceeded it.

The study of gold and silver medals and coins confirmed the historical features of their production. For example, a gold coin from the time of Nicholas II (sample 12) almost perfectly matches the declared 900 fineness, while silver «poltinniks» (samples 14, 15) have an inflated silver content. The analysis of modern coins from the European Union, Ukraine, and Poland confirmed the use of specialized alloys such as Neusilber and Nordic Gold, which is consistent with *known minting technologies. The results confirm that the XRF method is a reliable tool for quick and effective control of the composition of metal products, allowing for the detection of inconsistencies and falsifications, which is particularly important for both expert institutions and consumers. Overall, the conducted analysis demonstrates the high effectiveness of XRF for expert examination.*

Keywords: *X-ray fluorescence analysis, gold alloys, silver alloys, jewellery, coins.*

Gold and silver items, traditionally used as jewellery, remain popular due to their aesthetic qualities and corrosion resistance. However, pure gold is a soft metal, meaning jewellery made from it quickly deforms. To enhance wear resistance, reduce cost, and expand the colour palette, other metals are added to gold, forming alloys.

The mass content of the precious metal in an alloy is called the fineness and is marked with a digital hallmark. In Ukraine, as in many other countries, the metric fineness system is used, which indicates the number of grams of pure metal per one kilogram of the alloy (Instruction, 1999).

The colour of a gold alloy depends on the composition of the ligature (alloying elements). For example, yellow gold is achieved with a balanced content of zinc, copper, and silver. The intensity of red gold is determined by the proportion of copper, while white gold requires the addition of platinum, palladium, silver, or, in some cases, nickel. Notably, the use of nickel in jewellery manufacturing in Europe has been banned since 2011 due to its allergenic properties (Tkachuk, 2023).

Unfortunately, counterfeits are often found in the jewellery market, which makes controlling the composition of these items essential. One of the most effective methods for this purpose is X-ray Fluorescence Analysis (XRF), which allows for the rapid and non-destructive determination of the qualitative and quantitative composition of the material. XRF is actively used in forensic science and the jewellery industry, particularly for checking fineness conformity (Tkachuk & Navrotska, 2022).

The aim of this work is to investigate the chemical composition of gold and silver alloys, as well as other metal items, using the XRF method, and to compare the obtained results with the declared characteristics of the items.

The study employed the ElvaX X-ray fluorescence spectrometer (Beckhoff, 2007). The analysis was conducted on the following sample groups:

– gold jewellery: three modern, yellow gold items with 585 fineness of Ukrainian manufacture (Samples 1, 2, 3); red gold earrings with 585 fineness, dating from the 1990s (Sample 4);

Table 1

Qualitative and quantitative composition of gold jewelry

Sample Number	Element	Acquisition Time, s	Line Series	Intensity	Content, %
1	Au	121	L	–	58.191 ± 0.146
	Cu		K	–	30.722 ± 0.121
	Zn		K	–	8.845 ± 0.113
	Ag		K	–	2.242 ± 0.033
2	Au	120	L	–	58.144 ± 0.131
	Cu		K	–	30.503 ± 0.112
	Zn		K	–	7.497 ± 0.100
	Ag		K	–	3.274 ± 0.034
	Ni		K	–	0.583 ± 0,031
3	Au	121	L	–	58.546 ± 0.128
	Cu		K	–	26.849 ± 0.108
	Ag		K	–	7.807 ± 0.049
	Zn		K	–	6.797 ± 0.095
4	Au	121	L	936,912	39.800 ± 0.079
	Ag		K	320,000	39.133 ± 0.093
	Cu		K	616,841	21.067 ± 0.056
5	Au	141	L	550,127	74.546 ± 0.092
	Ag		K	182,283	15.511 ± 0.070
	Cu		K	78,821	9.943 ± 0.074

yellow gold ring with 750 fineness, produced in the USSR during the 1960s (Sample 5);

– silver jewellery: a ring with 925 fineness from Greece (Sample 6); a bracelet from Sri Lanka (Sample 7); a necklace from Ukraine, 19th–20th centuries (Sample 8); a brooch with 925 fineness, dating from the beginning of the second half of the 20th century (Sample 9);

– gold and silver coins and medals: gold medals from 1958 and 1987 (Samples 10, 11); a gold coin from the time of Nicholas II (late 19th – early 20th century) (Sample 12); a silver commemorative coin, «Princess Olga,» 1999 (Sample 13); silver «poltinnik» coins from the USSR, 1924 and 1925 (Samples 14, 15);

– items made from other alloys: an ornament made of a copper and silver alloy (Sample 16); a brass ornament (Sample 17); USSR 1 ruble coins (Samples 18, 19); European Union coins with denominations of 2€, 1€, 50¢, 1¢ (Samples 20, 21, 22, 23); a Polish coin with 1 grosz denomination (Sample 24); a Ukrainian coin with 2 UAH denomination (Sample 25).

Spectra and quantitative characteristics were obtained using the ElvaX software, which also determined the fineness of the items. The analysis time for each sample is specified in the corresponding tables.

The results of the gold jewellery analysis are presented in Table 1 and Figure 1. The analysis duration, typically 120–141 seconds, was chosen to ensure high statistical reliability with an error margin of ±0.079–0.146 %. The analysis utilized the L-series lines (around 9.7 keV) for gold, and

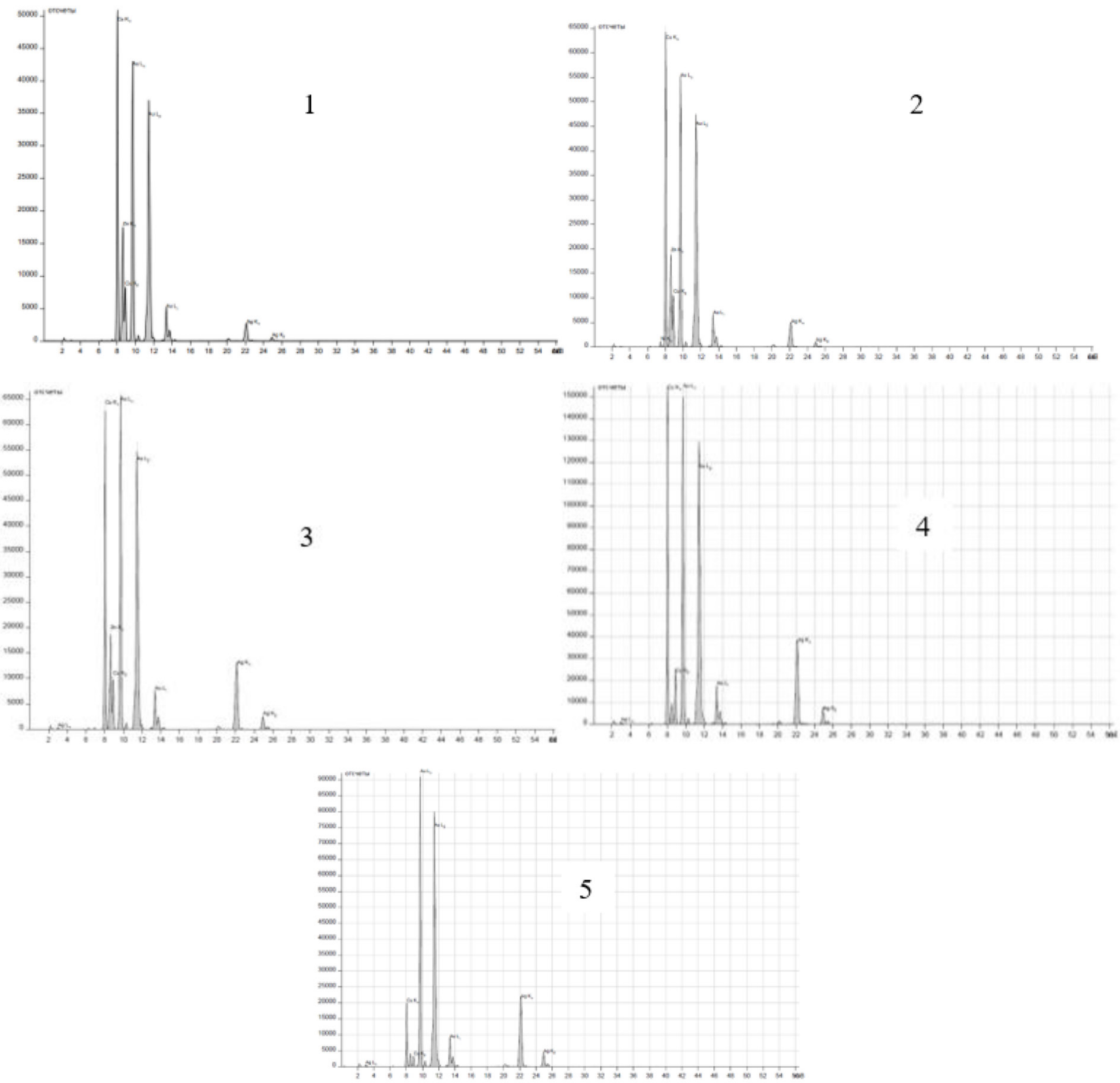

Fig. 1. XRF spectrum of gold jewelry

Table 2

Qualitative and quantitative composition of silver jewelry

Sample Number	Element	Acquisition Time, s	Line Series	Intensity	Content, %
6	Ag	31	K	1,579,772	92.246 ± 0.066
	Cu		K	62,168	7.754 ± 0.075
7	Ag	41	K	57,131	97.970 ± 0.294
	Cu		K	2,734	1.886 ± 0.082
	Au		L	266	0.144 ± 0.028
8	Ag	31	K	1,541,077	96.535 ± 0.053
	Cu		K	25,822	3.465 ± 0.057
9	Ag	31	K	554,818	95.278 ± 0.082
	Cu		K	12,427	4.722 ± 0.094

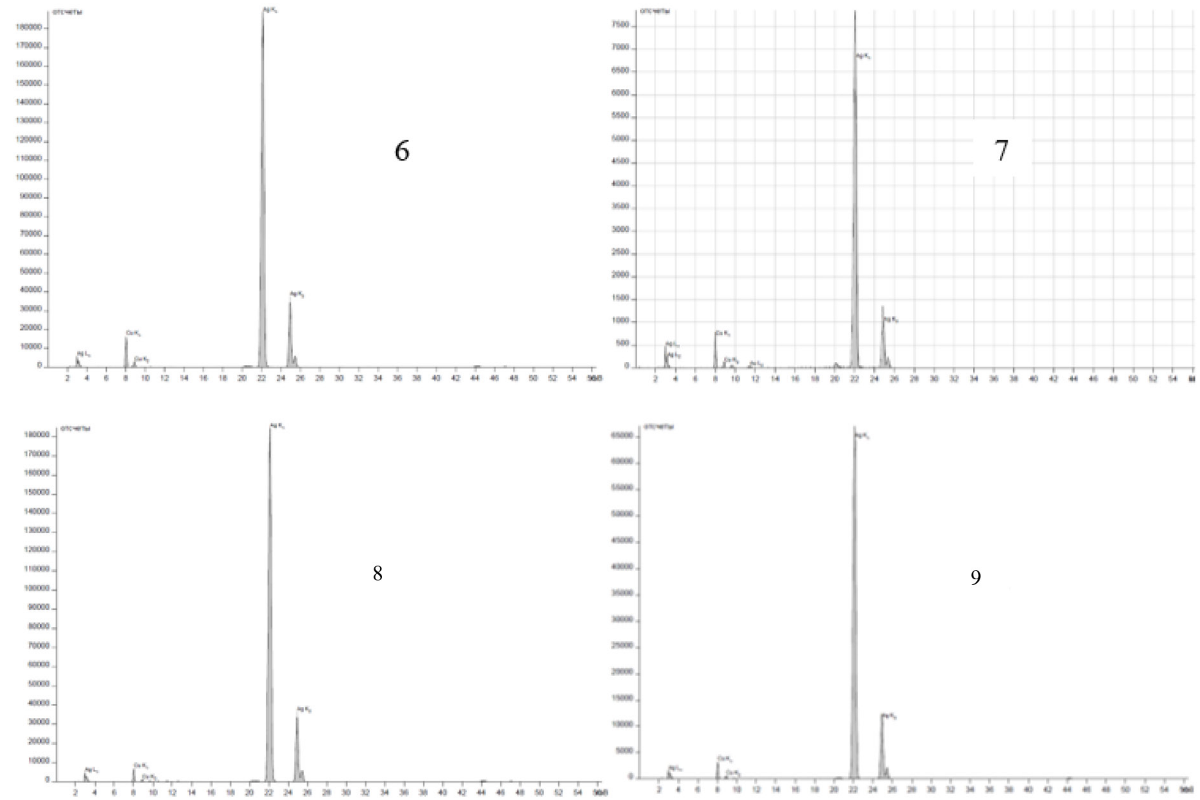

Fig. 2. XRF spectrum of silver jewelry

Table 3

Qualitative and quantitative composition of gold medals and gold and silver coins

Sample Number	Element	Acquisition Time, s	Line Series	Intensity	Content, %
10	Au	120	L	7,635,779	79.952 ± 0.038
	Cu		K	2,188,384	18.413 ± 0.028
	Ag		K	272,384	1.635 ± 0.008
11	Au	121	L	10,187,186	98.876 ± 0.02
	Cu		K	127,699	1.124 ± 0.011
12	Au	121	L	9,275,587	91.647 ± 0.020
	Cu		K	961,591	8.122 ± 0.019
	Ag		K	36,048	0.231 ± 0.005
13	Ag	30	K	1,373,192	91.59 ± 0.12
	Cu		K	26,862	8.41 ± 0.18
14	Ag	31	K	1,604,525	97.397 ± 0.048
	Cu		K	20,005	2.603 ± 0.049
15	Ag	31	K	1,432,794	95.95 ± 0.10
	Cu		K	12,869	4.05 ± 0.14

the K-series lines for the lighter alloying elements: copper, zinc, and silver.

The ElvaX spectrometer software determined the fineness of samples 1 and 2 to be only 500, while Sample 3 was identified as 585. This indicates that the gold content in the first two items (58.19 % and 58.14 %, respectively) was lower than the minimum permissible standard for 585 fineness (58.33 %). This discrepancy may result from rounding errors or a violation of manufacturing technology (Artyukh, 2018). The gold content in sample 3 (58.55 %) did, however, match the declared fineness.

Analysis of the alloying elements in the modern yellow gold items (samples 1–3) revealed that after gold, copper was the most prevalent element (21–31 %). Crucially, zinc (7–8.5 %), which is significantly cheaper than silver, constituted a larger proportion of the alloy than silver (2.2–8 %). This confirms that jewellers may be replacing silver with zinc to reduce the manufacturing cost of the items. Additionally, a minor content of nickel (~0.5 %) was detected in sample 2, which is an undesirable impurity (Lemberge, n.d.).

Sample 4 contains Au 39.80 %, Ag 39.13 and Cu 21.07 % which does not correspond to any standard fineness. However, neither zinc nor nickel was detected in this item. Sample 5, manufactured in the USSR and marked as 750 fineness, showed

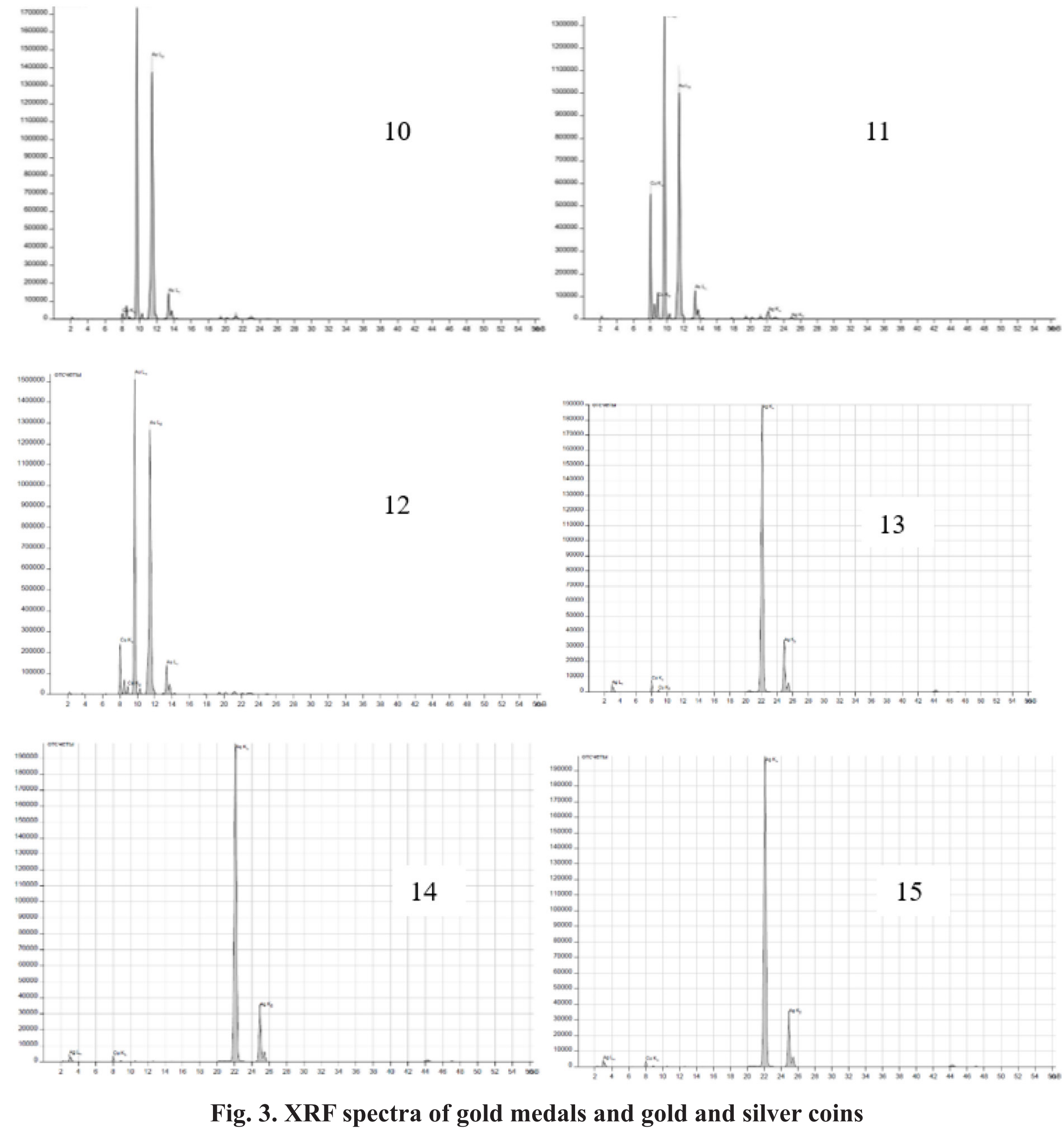

Fig. 3. XRF spectra of gold medals and gold and silver coins

a content of according to the XRF results, placing it Au 74.55 % closer to 585 fineness than to 750. The ratio of alloying elements in this sample is also distinct, with a higher silver content (15.51 %) than copper content (9.94 %) (Li, 2015).

The results from the analysis of silver jewellery are presented in Table 2 and Figure 2. The analysis time, ranging from 31 to 41 seconds, was considered sufficient to achieve an acceptable error margin of ±0.053–0.294 %. The K-series lines with an energy of approximately 22 keV were used for this analysis.

According to the XRF results, the silver content in Sample 6 was 92.25 %, which is slightly below the standard for 925 fineness. Consequently, the software identified it as 875 fineness. Sample 7 contained silver 97.97 % and copper 1.89 % with a minor gold impurity, a composition that does not correspond to any standard fineness. The necklace (Sample 8) showed a composition of Ag 96.54 % and Cu 3.47 %, corresponding to 960 fineness. The brooch (Sample 9), though marked as 925 fineness, showed an analysis result of Ag 95.28 % and Cu 4.72 %, significantly exceeding the 925-fineness standard. These results clearly demonstrate the considerable variations in the composition of silver items (Chiojdeanu & Cristea-Stan, 2011).

The results from the analysis of the gold medals and gold and silver coins are presented in Table 3 and Figure 3.

The gold medal from 1958 (Sample 10) contained gold (~80 %), suggesting the use of an alloy different from what may have been declared. The gold medal from 1987 (Sample 11) had a composition 98.88 % of gold. Since is a surface analysis method was applied, this very high gold content is likely explained by the presence of gold plating on the item. The gold coin from the time of Nicholas II (Sample 12) confirmed the declared 900 fineness with a composition of Au 91.65 %, Cu 8.12 %, Ag 0.23 %.

The silver commemorative coin "Princess Olga" from 1999 (Sample 13), declared as 925 fineness, showed an Ag content of 91.59 %, which is closer

Table 4

Qualitative and quantitative composition of non-jewelry art products and coins

Sample Number	Element	Acquisition Time, s	Line Series	Intensity	Content, %
16	Cu	30	K	609,602	74.73 ± 0.16
	Ag		K	824,458	25.27 ± 0.07
17	Cu	31	K	962,090	65.26 ± 0.10
	Zn		K	737,133	34.74 ± 0.09
18	Cu	31	K	925,133	56.94 ± 0.11
	Zn		K	459,765	29.10 ± 0.10
	Ni		K	210,702	10.99 ± 0.06
	Mn		K	31,611	2.97 ± 0.06
19	Cu	31	K	879,341	54.19 ± 0.11
	Zn		K	507,179	54.19 ± 0.11
	Ni		K	202,337	10.44 ± 0.06
	Mn		K	43,546	10.44 ± 0.06
20	Cu	121	K	5,039,532	76.681 ± 0.035
	Zn		K	1,444,329	18.441 ± 0.037
	Ni		K	381,559	4.877 ± 0.018
21	Cu	120	K	6,139,683	77.432 ± 0.036
	Ni		K	1,645,825	22.568 ± 0.032
22	Cu	21	K	2,967,861	94.843 ± 0.069
	Zn		K	223,458	5.157 ± 0.046
23	Cu	21	K	3,016,710	96.510 ± 0.068
	Fe		K	136,896	3.490 ± 0.037
24	Cu	120	K	5,237,297	77.754 ± 0.031
	Zn		K	1,940,193	20.957 ± 0.036
	Fe		K	105,628	1.289 ± 0.015
25	Ni	21	K	3,051,365	99.508 ± 0.0
	Fe		K	22,173	0.492 ± 0.016

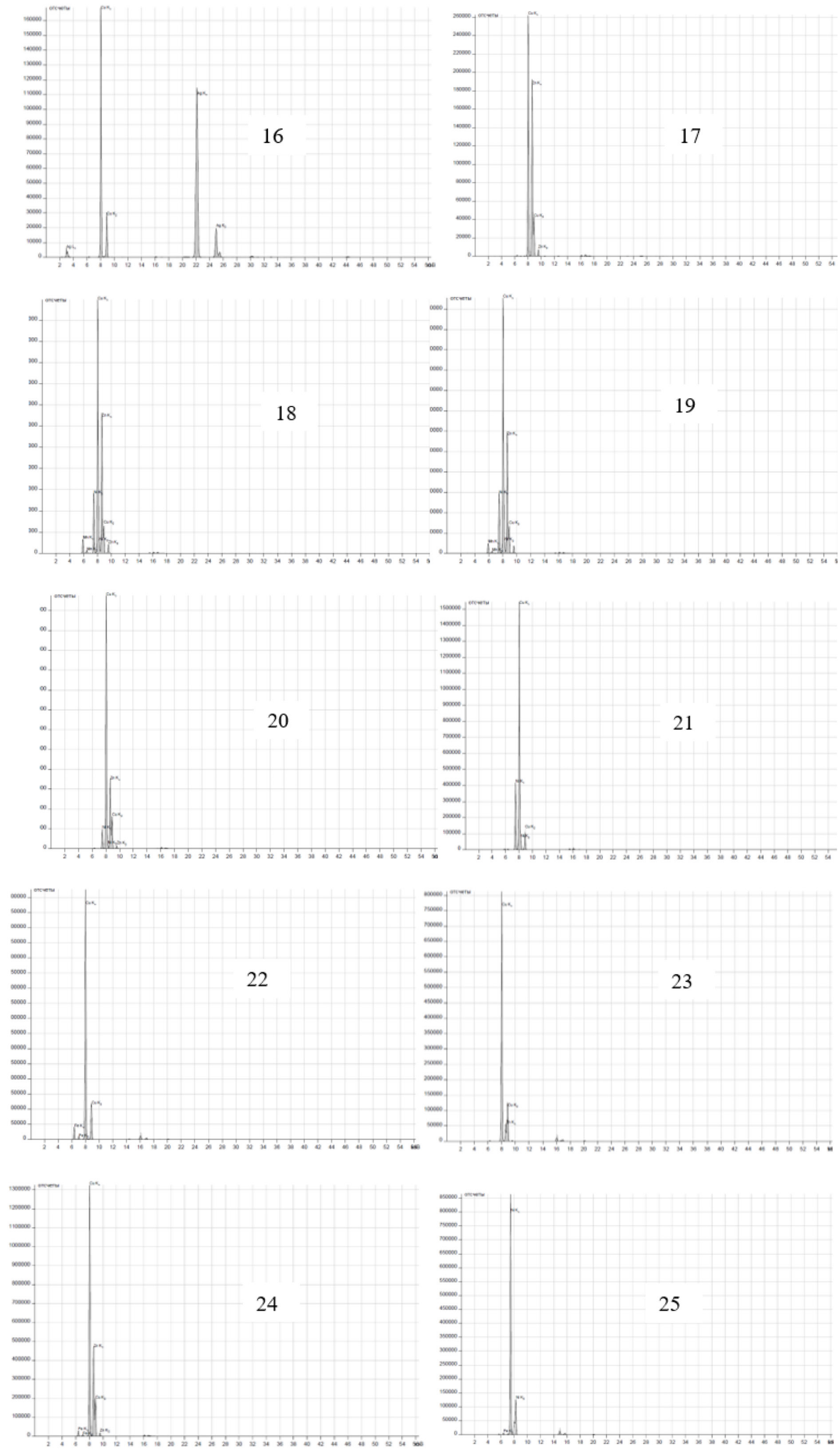

Fig. 4. XRF spectra of non-jewelry art products and coins

to the 875-fineness determined by the instrument (Serrano, 2023). The silver "poltinnik" coins from 1924 (sample 14) and 1925 (Sample 15), despite being marked as 900 fineness, demonstrated an elevated silver content (97.40 % and 95.95 %, respectively).

In addition to the gold and silver items, we investigated ornaments that do not belong to the jewellery category but can be classified as art metal products or costume jewellery, as well as various coins. The results of these studies are shown in Table 4 and Figure 4.

Sample 16, containing 74.73 % Cu and 25.27 % Ag, is a copper-silver alloy that does not adhere to traditional jewellery standards. It is most likely classified as costume jewellery. The composition of the brass ornament (Sample 17) confirmed that it is made of a copper-zinc alloy (Mazuki, 2023).

Analysis of the USSR coins from 1964 and 1970 (Samples 18, 19) confirmed they were made from cupronickel (a copper-zinc-nickel alloy, often referred to as German silver or Neusilber), known for its silvery colour and high durability. The composition of the European Union coins (2€, 1€, 50¢) and the Ukrainian 2 UAH coin aligns with known minting technologies: bi-metallic alloys for the euro coins (Cu-Zn-Ni and Cu-Ni), the Nordic Gold alloy (Cu-Zn) for the 50¢ coin, steel cores with a coating of copper (1¢) or nickel (2 UAH).

The study of the chemical composition of jewellery, medals, and coins using the XRF method demonstrated its high effectiveness as a non-destructive tool for quality control of precious metals. The significant discrepancies found between the hallmarking and the actual composition, especially in modern items, point to systemic issues in the jewellery market, specifically suggesting possible falsification and non-compliance with established standards.

In particular, it was confirmed that to reduce the cost of gold alloys, silver in the ligature is often replaced with cheaper zinc. Furthermore, an undesirable impurity of nickel, a known allergen, was detected in one of the samples. These results underscore that the XRF methodology is critically important for expert institutions seeking to protect consumers from unscrupulous manufacturers. In contrast, the analysis of historical items, such as the gold coin, confirmed that minting technologies in the past were generally more accurate, and the alloy composition adhered to the declared fineness. The investigation of other metal items, including European Union and Ukrainian coins, also confirmed that their composition aligns with known technologies, such as the use of cupronickel (Neusilber) and Nordic Gold alloys, further demonstrating the reliability of XRF as a method for metal identification across various applications.

Based on the conducted analysis, several key conclusions were drawn:

1. The study confirmed that a significant portion of modern jewellery items deviates from the declared fineness, indicating a mismatch between the marking and their actual chemical composition.
2. It was established that silver in modern gold items is often replaced by cheaper zinc, and the presence of undesirable impurities, such as nickel, was detected in some samples.
3. Analysis of historical gold and silver coins showed that their composition generally corresponds to the declared characteristics, reflecting historical production features.
4. The investigation of coins and other metal products confirmed adherence to established technological standards for minting.
5. Ultimately, the obtained data proves that XRF analysis is an indispensable, rapid, and non-destructive tool for quality control, essential for detecting falsifications in the precious metals market.

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Стаття надійшла: 02.11.2025
Прийнято: 21.11.2025
Опубліковано: 30.12.2025