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The color of gold alloys for dentistry

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The influence of alloying elements on the properties of gold-based alloys is studied in this paper. The purpose of the study was to establish the regularities of the gold color changing during the process of introduction of one or several alloying elements into the alloy: silver, copper, platinum, palladium, etc. A spectrophotometer with a spherical geometry of measurement was used to obtain the information about the color. The results are shown as the coordinates in the standard $L^*a^*b^*$ color space. The values of the chromaticity coordinates of gold alloys smoothly decrease with the increasing of the content of platinum or palladium. According to this, the yellowness index decreases.

The received data make it possible to analyze the results of experiments. During the development process of gold alloys for dentures, there has been substantiated to limit the content of platinum and palladium to the specific level, on which the alloys color is still perceived as yellow.

Key words: gold alloys, dentistry, alloying elements, color measurement, spectrophotometry, color space, yellowness.

The gold-based alloys are widely used in various fields of production and consumption. Depending on purposes, these alloys have the requirements, which are established by standards and specifications, based on the traditions and demand. In particular, color is one of the important consumer properties for a large part of jeweler alloys and some dental alloys. The consumers of gold dental alloys prefer yellow alloys, because yellow color meets many patients' aesthetic requirements, as an attribute of noble and innocuous metal. Dental prosthetics prefer yellow alloys either, for the production of frames in imitation of ceramics, because this makes it possible to obtain the warm and natural shades of enamel.

Just like other physical and mechanical properties, the color of alloy depends on the selection of composition and percentage of alloying elements. The most studied color is the color of jeweler alloys (Au–Ag–Cu systems, in particular). When alloying of the indicated system alloys, the color changes from yellow-orange to green-yellow shades, and the color strength decreases with the increasing of the content of silver and, especially, copper [1]. The functional standard on the jeweler alloys [2] contains the data, which makes the conclusion that the color of gold alloy depends on the composition of alloying elements, rather than on the content of the basic component (standard).

The colors of jeweler gold-based alloys are given in the Table 1. It can be seen that even with a low level of gold standard, the alloys can have yellow colors or shades, while the high-standard alloys can be white, especially when alloyed with palladium or nickel.

As a rule, the gold alloys, used in dentistry, are the complex multicomponent compositions that may contain silver, platinum and other platinoids, copper and other base metals [3]. The selection of a particular alloy composition depends on its purpose and must provide the fulfillment of certain medical and technical requirements, such as: the corrosion resistance, biological inertness, strength characteristics, technological effectiveness, etc.

It should be noted that certain elements have an appreciable influence on the gold color. For example, silver effectively neutralizes the red shades, caused by alloying with copper; while platinum reduces the yellow color. Alloying with palladium (more than 5% (wt.)) leads to the lightening of gold alloys.

As a rule, the manufacturers of dental alloys indicate the information about the color of the gold alloys in their informational materials as yellow, pale yellow, light yellow, and white. When comparing the products from different companies, it is often found that the alloys with similar compositions could be presented as the alloys with different colors. For example, almost identical alloys Armator DB (UGDO) and V-Classic (Metalor), are presented as white and yellow. This can be explained by the subjectivity of visual perception and shortage of verbal descriptions for all variety of existing colors.

The special devices and methods make it possible to give the definite and objective identification of the color of material. The color measurements (as well as light measurements), which operate with color sensations, apply mostly to physical, psychological and physiological sphere, rather than only to physical sphere [4, 5]. In order to bring the results of instrumental measurements in conformity with real visual sensations, the instrument readings (spectrometric data) are mathematically transformed according to the unified methods approved by the Commission International d`Eclainage

(CIE). Finally, the coordinates in a conditional color space are assigned for any of the colors. For example, the data about the colors of six jeweler alloys of the Au–Ag–Cu system of 585 and 750 standards are specified in the international standard [6] in both verbal designations and the chromaticity coordinates in a three-dimensional color space *x*, *y*, and ρ, according to the CIE Publication No. 15 (Table 2).

When researching of the development [7] of new gold dental alloys[1], there was initially used a method of visual expert evaluations by the intercomparison of the colors of experimental samples and a prototype. There was found that different observers (including technicians and dentists), put the samples in different ways in a row from "more yellow" to "less yellow" according to their individual perception and their conception about colors. Subsequently, the color measurements were held, using a spectrophotometry method to get the objective data, which turned out to be quite productive for the samples with similar compositions and for light yellow alloys.

The purpose of the study was to determine the conformity to the gold color changing in the process of introduction of one or several alloying elements into the alloy. At the same time, special attention was given to platinoids, which intensively suppress the yellow color. However, their presence in gold alloys is necessary for their usage in the structures in imitation of the metal-ceramics, since platinum and palladium decrease the thermal expansion coefficient and improve the hardness of alloy, while silver raises this coefficient. The selection of optimum combination of the mentioned antogonistic elements appreciably determines the success in the alloys development with the required consumer properties. Also, there were investigated the color characteristics of gold with silver, copper, tin, zinc and indium, in combination of these elements with platinoids.

The spectrophotometer with X-Rite SP-62 grade was used for the measurement of chromatic characteristics[2] of the samples. The device has a spherical geometry of measurement, which allows to exclude the desorption of the color rendition under the influence of metal surface gloss of the samples in the reflected light. The received results, issued in the form of printouts, included:

— the spectral distribution curves;

— the values of chromaticity coordinates in the $L^*a^*b^*$ CIE-76 (CIE LAB) color space and other systems;

— the data on yellowness, which were calculated, according to the ASTM E 313 standard, etc [5].

The coordinates in CIE LAB mean the following: L^* is for clarity; a^* is for a color range in the color circle from green (–120 °) to red (+120 °); b^* is for a color range from dark blue (–120 °) to yellow (+120 °).

The manufacturing of the samples of gold alloys included the following operations: melting of 10–20 g ingots in a resistance furnace; blacksmithing; pressing; and rolling with intermediate heat treatments. The samples surface corresponded to the surface of the polished forming rolls. There were prepared the experimental samples of two-component alloys of gold with platinum, palladium, rhodium, silver, copper, tin, and

Table 1
Colors of the gold-based jeweler alloys

Alloy	Color
375 Standard (37.5% of gold)	
Au–Ag–Cu 375–20	bright yellow
Au–Ag–Cu 375–100	red
Au–Ag–Cu 375–160	red
Au–Ag–Cu 375–250	pink-yellow
Au–Ag–Pd–Cu 375–100–38	yellowish-orange
500 Standard (50.0% of gold)	
Au–Ag–Cu 500–100	red
585 Standard (58.5% of gold)	
Au–Ag 585–415	green
Au–Ag–Cu 585–80	red
Au–Ag–Cu 585–200	reddish-yellow
Au–Ag–Cu 585–300	yellow-green
Au–Ag–Pd 585–255–160	white
Au–Ag–Pd–Zn 585–287–100	white
Au–Ag–Pd–Cd 585–280–100	white
Au–Ag–Ni–Zn–Cu 585–80–8.2–2.5	light yellow
Au–Ni–Zn–Cu 585–12.5–4	white
750 Standard (75.0% of gold)	
Au–Ag 750–250	green
Au–Ag–Cu 750–125	bright yellow
Au–Ag–Cu 750–150	yellow
Au–Ag–Ni–Zn 750–150–7.5	white
Au–Ag–Pd 750–100–150	white
Au–Ag–Pt–Cu 750–80–90	yellowish
Au–Ag–Pd–Ni 750–90–140	white
Au–Ag–Pd–Ni 750–70–140	white
Au–Ag–Pd–Ni–Cd 750–90–85–4	white
Au–Ni–Zn–Cu 750–7.5–2.5	white
958 Standard (95.8% of gold)	
Au–Ag–Cu 958–20	bright yellow
999.9 Standard (~100% of gold)	
Au 999.9	bright yellow

Table 2
Designation of the color of jeweler alloys

Color designation		Chemical composition, % (copper - balance)		Chromaticity coordinates		
Index	Description	Au	Ag	*x*	*y*	ρ
0N	yellow-green	58.5	30.0 – 34.0	0.3383	0.3662	0.90
1N	pale yellow	58.5	24.0 – 26.5	0.3526	0.3700	0.82
2N	light yellow	75.0	15.0 – 16.0	0.3590	0.3766	0.82
3N	yellow	75.0	12.0 – 13.0	0.3601	0.3729	0.79
4N	pink	75.0	8.5 – 9.5	0.3612	0.3659	0.76
5N	red	75.0	4.5 – 5.5	0.3591	0.3604	0.74

[1] The joint developments of the Scientific and Industrial Complex "Supermetal" and the Moscow State University of Medicine and Dentistry

[2] The measures were held by Candidate of Chemical Sciences R. A. Khafizova, and Candidate of Engineering Sciences K. A. Subbotin in "TEKSA" LLC

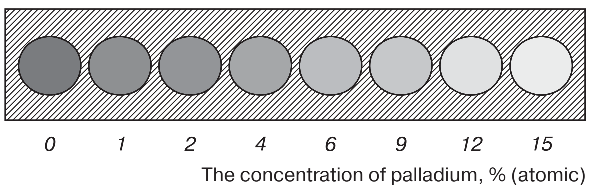

Fig. 1. Appearance of the samples of Au–Pd system alloys for the color measurement

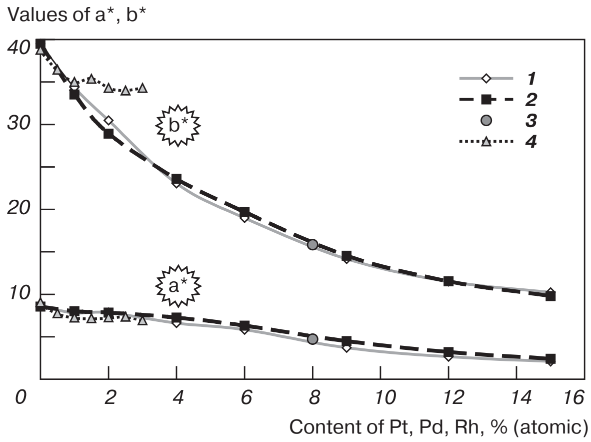

Fig. 2. The influence of platinoids on the chromaticity coordinates of gold alloys: *1* — Au–Pt; *2* — Au–Pd; *3* — Au–Pt–Pd; *4* — Au–Rh

zinc, as well as three- and four-component alloys containing platinoids and one of the foregoing elements, and samples of some dental alloys.

The samples of gold and two-component Au–Pd alloys are shown in Fig. 1. In the alloying process, their color changes from yellow to white (practically colorless). The first two or three left samples (from the gold sample) can be called yellow, the next two samples can be called light yellow, the last but one and the last one can be called white. However, it is hard to distinguish visually the differences between the following samples. On the other hand, the spectrophotometric color measurements produce considerably different and logical results (Fig. 2).

The results, which are given in the Fig. 2, show that the values of chromaticity coordinates of two-component gold alloys decrease gradually with increasing of the platinum or palladium content, and with more abruptly decreasing of the share of prevailing yellow color (*b**) in comparison with red color (*a**), especially at the initial stages of alloying. The curves, built for Au–Pt and Au–Pd alloys, are practically similar. The chromaticity coordinates of the three-component gold alloy, alloyed both with platinum and palladium (4% (atomic)), are situated on the same curves. Alloying of gold with rhodium is also accompanied by the decreasing of values of *a** and *b**, but only to 1% (atomic); the color does not change after that. This fact can be explained by the overstepping of boundary of solid solutions at more high concentrations of rhodium, which excess is spent for the formation of the second phase. This can be confirmed by a hardness diagram (Fig. 3) for the Au–Rh alloys in which the solid-solution strengthening is observed to 1% (atomic) of rhodium. The hardness does not change at its higher content. The gold forms the solid solutions in the whole researched range of platinum and palladium concentrations. Therefore, the changing of chromaticity characteristics is smooth and has no knees, as well as the changing of hardness (Fig. 3). Inherently, there is a smooth decreasing in the color saturation [8], and its approaching to white color (achromatic).

The similar regularities of the influence of platinoid concentration on the gold alloys color are observed in the changing diagrams of the yellowness index (YI) of the samples (Fig. 4). The estimation of yellowness is important for the solution of the applied problem of the dental alloys development. According to the Fig. 4, the yellowness index YI = 17.22 is revealed instrumentally even with a sample of almost white (visual perception) alloy of gold with 15% (atomic) of palladium. Formally, a sample can only be considered to be white at zero values of *a**, *b**, and YI. Therefore, proceeding from the practical experience, the criterion of "yellowness loss" can only be conditionally established for a gold alloy. It is considered, that the alloys with YI < 20 are perceived visually as white with a weak yellow shade, while the alloys with YI < 25 are perceived as pale yellow. In particular, "Super-LB" gold alloy (CASDENT-B), developed by Scientific and Industrial Complex "Supermetal" and Moscow State University of Medicine and Dentistry, has a light yellow color, which yellowness value is YI = 27.27.

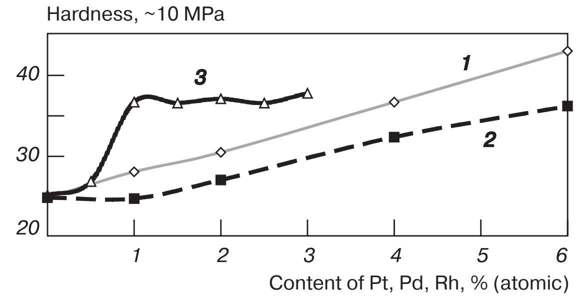

Fig. 3. The influence of alloying on the hardness of gold alloys: *1* — Au–Pt, *2* — Au–Pd, *3* — Au–Rh

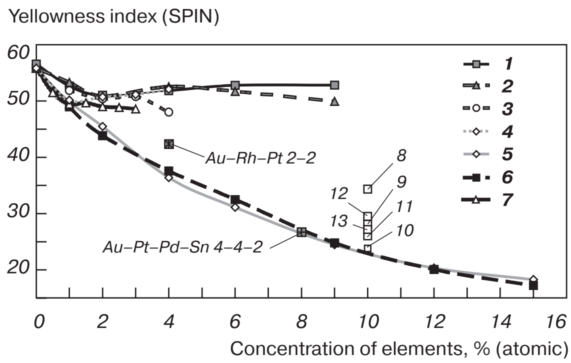

Fig. 4. The influence of alloying on the yellowness of gold alloys: *1* — Ag; *2* — Cu; *3* — Sn; *4* — Zn; *5* — Pt; *6* — Pd; *7* — Rh; *8* — Au–Pt–Pd 4–4; *9* — Au–Pt–Pd–Rh 4–4–2; *10* — Au–Pt–Pd–Ag 4–4–2; *11* — Au–Pt–Pd–Cu 4–4–2; *12* — Au–Pt–Pd–In 4–4–2; *13* — Au–Pt–Pd–Zn 4–4–2

In comparison with the platinoids, the particular alloying elements, such as silver, copper, and zinc, have a lower effect on the yellowness of two-component gold alloys (Fig.4). In more complex multicomponent alloys, with joint alloying of gold with platinum, palladium, and other elements, rhodium and tin appreciably improve the yellowness of the alloy, while silver worsens it.

The obtained data make it possible to analyze objectively the experimental data and reasonably limiting of the platinum and palladium content in the gold alloys to a level at which the alloy color is perceived as yellow.

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New gold alloys for dentistry

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The purpose of this investigation was to create a new yellow gold alloy for the casting of metal-ceramic dentures' frames. The yellow color corresponds to the consumer and aesthetic needs of some patients, because of the sign of noble and innocuous metal. At the same time, it is necessary to ensure the correspondence of the alloy properties with the properties of the ceramics, applied on the metal frame. For this purpose, the thermal expansion coefficient of the alloy (TEC) should be in the range of $(13.5–14.5) \cdot 10^{-6}$ K^{-1}, with the heating from 20 °C to 600 °C.

Platinum and palladium are the main alloying elements of the majority of gold alloys for metal-ceramics. These metals increase the characteristics of strength. Copper, tin, and other precious and base metals are also included in these alloys.

Alloying of gold with platinum and palladium in the two-component alloys, leads to the decrease in the TEC. However, TEC increases with introduction of copper, silver, and tin. The multidirectional influence of the alloying elements is the achievement factor in the compliance of TEC with the given values of the alloy. However, in multicomponent systems, the mutual influence of individual components on the alloy properties is unpredictable. This also applies to the color characteristics of the alloys, which vary in the direction of yellowness reduction with the increasing of platinum and palladium concentration, while other elements may have the opposite effect on the results. The yellowness index (YI), calculated according to the results of spectrophotometric studies, was chosen as an objective indicator of color. In this study, the requirement for YI was given as not less than 25. The color of such alloys can be called light yellow.

All investigated alloys contain 85% (wt.) of gold. Therefore, the higher corrosion resistance and biological inertness of finished dental products were ensured.

Two the “most yellow” alloys were selected among the alloys that met the requirements of both yellowness and TEC. The adhesive properties of these alloys met the requirements of the State Standard R 51736-2001 to the alloys for metal ceramics: bond strength with ceramics, applied on the surface of a metal sample, was above a minimum value, equal to 25 MPa (Table 2).

Key words: gold, platinum, palladium, alloying elements, alloy, yellowness, thermal expansion, metal-ceramics.

Since the late 1990s, the Scientific and Industrial Complex “Supermetal” (SIC “Supermetal”), in a creative collaboration with the Moscow State University of Medicine and Dentistry (MSUMD), has been developing and producing the dental alloys on the basis of noble metals. The ideology of the creation of new alloys, their compositions and properties, as well as the analysis of the products of other manufacturers, and the market situation, are presented in [1, 2].

The gold alloy for the metal-ceramics grade PLAGODENT (Super-KM) is the most popular alloy, produced by SIC “Supermetal”. In orthopedic dentistry, the term “alloy for metal-ceramics” means an alloy, suitable for application as a metal frame of metal-ceramic denture [3]. The metal frame

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