Dental Materials (2006) **22**, 641–646

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dental materials

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Elastic moduli of cast Ti-Au, Ti-Ag, and Ti-Cu alloys

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Received 8 June 2004; received in revised form 11 May 2005; accepted 11 May 2005

KEYWORDS
Titanium alloy;
Casting;
Young's modulus;
Poisson's ratio;
Density

Summary *Objectives*. This study investigated the effect of alloying titanium with gold, silver, or copper on the elastic properties of the alloys.
Methods. A series of binary titanium alloys was made with four concentrations of gold, silver, or copper (5, 10, 20, and 30 mass%) in an argon-arc melting furnace. The Young's moduli and Poisson's ratios of the alloy castings were determined with an ultrasonic-pulse method. The density of each alloy was previously measured by the Archimedes' principle. Results were analyzed using one-way ANOVA and the Scheffé's test.
Results. The densities of Ti–Au, Ti–Ag, and Ti–Cu alloys monotonically increased as the concentration of alloying elements increased. As the concentration of gold or silver increased to 20%, the Young's modulus significantly decreased, followed by a subsequent increase in value. As the concentration of copper increased, the Young's modulus monotonically increased. The Young's moduli of all the Ti–Cu alloys were significantly higher than that of the titanium.
Significance. The density of all the experimental alloys was virtually independent of the alloy phases, while the Young's moduli and Poisson's ratios of the alloys were dependent. The addition of gold or silver slightly reduced the Young's modulus of the titanium when the alloy phase was single α. The increase in the Young's modulus of the Ti–Cu alloys is probably due to the precipitation of intermetallic compound Ti_2Cu. Copper turned out to be a moderate stiffener that gains a Young's modulus of titanium up to 20% at the copper concentration of 30 mass%.

Introduction

There are many mechanical properties, which characterize a material, such as yield strength, tensile strength, elongation, and hardness. Stiffness, or modulus of elasticity, is one of the most important values once the material is formed to the final shape and used within the elastic range [1]. The low elastic modulus of the material results in large deflection even if it has high yield strength. The modulus is an indispensable design value when computing deflections of prostheses by structural mechanics.

A typical Young's modulus for titanium (100-110 GPa) is about one half that of stainless steel or

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doi:10.1016/j.dental.2005.05.015

Co-Cr alloys [2,3]. A higher modulus of elasticity is required to minimize the elastic deformation when applying titanium to dental prostheses that are subject to comparatively high stress, such as bridges and partial denture frameworks. A higher modulus of elasticity is also favorable to machining titanium [4]. On the other hand, there also exist applications that require a lower modulus of elasticity than titanium. For example, low stiffness offers the advantage of adapting the implant or prosthesis behavior to bones, which have a considerably lower Young's modulus than titanium [3].

The mechanical properties of titanium can be changed through alloying [5,6]. The elastic properties of many titanium alloys have been studied [7–12]. However, few studies have been done on the elastic properties of cast titanium alloys with noble metals. In the present study, a series of experimental binary titanium alloys was made with two noble metals (gold and silver) and copper as alloying elements. While copper is not a noble metal, it was selected because it belongs to the same group of elements of the periodic table as gold and silver (Group 11). The Young's moduli and Poisson's ratios of the as-cast titanium alloys were determined by the ultrasonic-pulse method in order to investigate the effect of alloying on the elastic properties of titanium. The values for two commercial titanium alloys (Ti–6Al–4V and Ti–6Al–7Nb) were also determined.

Materials and methods

Chemical compositions of experimental alloys

Gold, silver, and copper are β-stabilizing elements that reduce the allotropic transformation temperature from β (bcc) to α (hcp) when dissolved in titanium [5,6]. Ti-Au, Ti-Ag, and Ti-Cu alloys are classified as titanium alloys with eutectoid transformation and have eutectoid points at 15.3 mass% Au, 15.6 mass% Ag, and 7.0 mass% Cu, respectively (hereafter, 'mass%' will be referred to as '%' unless otherwise specified) [13–15]. As shown in Table 1, the experimental binary titanium alloys in the present study were selected from both hypoeutectoid and hypereutectoid regions.

Preparation of experimental alloys

Buttons (15 g each) of the titanium alloys corresponding to the desired concentrations were made by melting titanium (>99.8%, grade S-90, Sumitomo Titanium, Amagasaki, Japan) and gold (99.99%, Ishifuku Metal Industry, Tokyo, Japan), silver (99.99%, Ishifuku Metal Industry), or copper (99.99%, The Research Institute for Electric and Magnetic Materials, Sendai, Japan) in an argon-arc melting furnace (TAM-4S, Tachibana-Riko, Sendai, Japan). After the chamber was evacuated to 5 mPa, high-purity (>99.9999%) argon gas (Nipponsanso, Kawasaki, Japan) was introduced until the pressure reached 50 kPa for the melting atmosphere. The titanium 'getter' (a scrap piece of Ti, to remove O_2 from the atmosphere) was melted before melting the buttons. A batch of buttons was melted six times in all; they were turned upside down five times to ensure homogeneity. Titanium buttons were made similarly by melting only the titanium sponge.

Preparation of specimens

To make the specimens of the titanium and the experimental titanium alloys by casting, wax patterns (3.5 mm × 8.5 mm × 30.5 mm) were invested into a magnesia mold (Selevest CB, Selec, Osaka, Japan). According to the manufacturer's instructions, the mold was heated to 850 °C at 6 °C min^{-1} with a 1-h-hold period, followed by cooling to 200 °C in a computerized furnace (KDF-009G, Denken, Kyoto, Japan). Each metal button was arc-melted and cast into the mold by a dental casting machine (Castmatic-S, Iwatani, Osaka, Japan). After casting, the molds were bench-cooled at room temperature. Prior to the tests, approximately 250 μm from all the surfaces of the castings was ground and polished down to #1000 abrasive

Table 1 Chemical compositions and eutectoid points of experimental titanium alloys.

Alloy	Chemical compositions	Eutectoid point
Ti-Au	5, 10, 20, 30% (1.3, 2.6, 5.7, 9.4 mol%) Au	15.3% (4.2 mol%) Au, 832 °C ($\beta Ti \leftrightarrow \alpha Ti + Ti_3Au^a$)
Ti-Ag	5, 10, 20, 30% (2.3, 4.7, 10, 16 mol%) Ag	15.6% (7.6 mol%) Ag, 855 °C ($\beta Ti \leftrightarrow \alpha Ti + Ti_2Ag^b$)
Ti-Cu	5, 10, 20, 30% (3.8, 7.7, 16, 24 mol%) Cu	7.0% (5.4 mol%) Cu, 790 °C ($\beta Ti \leftrightarrow \alpha Ti + Ti_2Cu^c$)

[a] 57.8% (25.0 mol%) Au.
[b] 53.0% (33.3 mol%) Ag.
[c] 39.9% (33.3 mol%) Cu.

paper to remove the hardened surface layer, producing specimens measuring 3 mm×8 mm×30 mm.

Specimens (25 mm in diameter and 3 mm in thickness) sliced from wrought Ti-6Al-4V (annealed, DAT5, Daido Steel, Nagoya, Japan) and wrought Ti-6Al-7Nb (annealed, T-Alloy Tough, GC, Tokyo, Japan) were also prepared as representatives of commercial titanium alloys for reference.

Measurement of elastic constants

An ultrasonic-pulse method is a non-destructive and dynamic way of measuring the modulus of elasticity. Higher precision can generally be obtained by the dynamic rather than static methods [1,7,11]. The Young's modulus (modulus of longitudinal elasticity, E) and Poisson's ratio (ν) of a material are given by the following equations:

$$E = \rho \frac{3V_L^2V_T^2 - 4V_T^4}{V_L^2 - V_T^2}$$

$$\nu = \frac{1}{2}\frac{V_L^2 - 2V_T^2}{V_L^2 - V_T^2}$$

where V_L and V_T are the longitudinal and transverse wave velocity, respectively, and ρ is density of the material [16]. The density of each specimen was measured by Archimedes' principle using an electronic chemical balance (ER-182A, A&D, Tokyo, Japan) at room temperature. Both V_L and V_T were calculated by the thickness of the specimen (3 mm) divided by one half of the round-trip transit time. The thickness of each specimen was measured using a micrometer (MDC25M, Mitutoyo, Tokyo, Japan). An ultrasonic pulser/receiver (5800, Panametrics, Waltham, MA, USA), transducers (M208 and V156, Panametrics), and a computer (AT-680C, Epson, Matsumoto, Japan) equipped with a digitizer (NI5112, National Instruments, Austin, TX, USA) were used to determine the round-trip transit time through each specimen at room temperature. Three specimens were used for each metal.

Statistical analysis

The results for the titanium, experimental titanium alloys, and commercial titanium alloys were compared using one-way ANOVA and the Scheffé's test at a significance level of $\alpha = 0.05$.

Results

Density

The densities of Ti-Au, Ti-Ag, and Ti-Cu alloys monotonically increased as the concentration of alloying elements increased (Fig. 1). The densities of all the Ti-Au, Ti-Ag, and Ti-Cu alloys were significantly higher than that of the titanium ($p < 0.001$). As shown in Fig. 2, changes in the density of the Ti-Au, Ti-Ag, and Ti-Cu alloys were almost linear to the concentration of alloying elements in mol%.

Young's modulus and Poisson's ratio

The Young's moduli of the alloys tested are shown in Fig. 3. Changes in the Young's modulus of the Ti-Au and Ti-Ag alloys were similar. As the concentration of gold or silver increased to 20%, the Young's modulus decreased, followed by a subsequent increase in value. The Young's modulus of Ti-20%Au was significantly lower than that of titanium ($p < 0.001$), while that of Ti-30%Au was significantly higher ($p < 0.001$). The Young's modulus of Ti-20%Ag was significantly lower than that of titanium ($p < 0.001$).

As the concentration of copper increased, the Young's modulus monotonically increased. The Young's moduli of all the Ti-Cu alloys were significantly higher than that of titanium (5% Cu: $p < 0.05$; 10% Cu: $p < 0.01$; 20 and 30% Cu: $p < 0.001$). Ti-30%Cu had the highest Young's modulus (20% higher than the titanium) and Ti-20%Ag the lowest

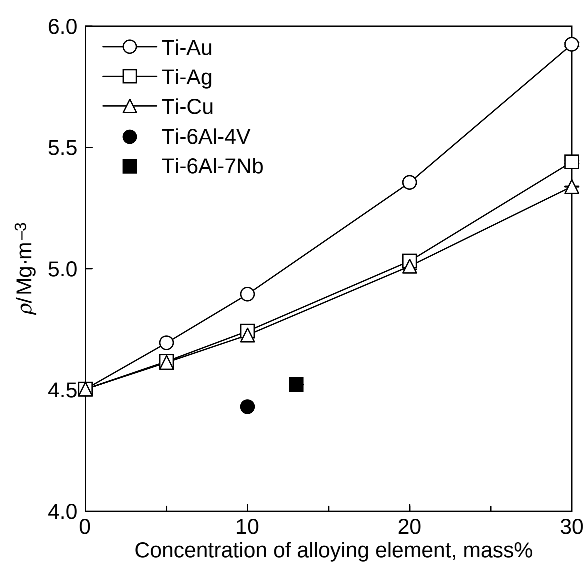

Figure 1 Density (ρ) of the titanium alloys. Error bar: ±SD.

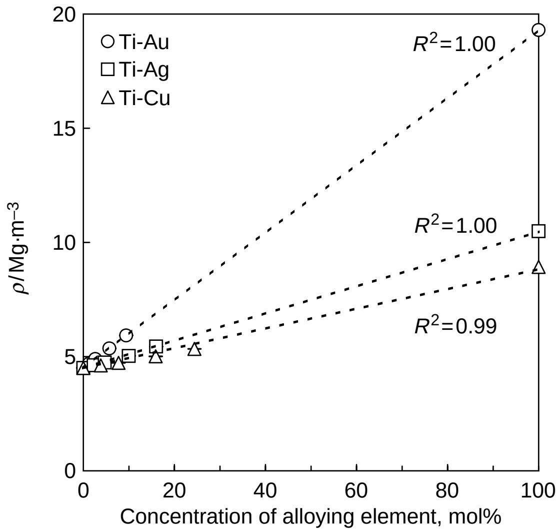

Figure 2 Relationship between the concentration of the alloying element in mol% and the density (ρ) of the experimental alloys. The densities of pure gold, silver, and copper were cited from reference [21]. Error bar: $\pm$SD.

(8% lower than the titanium) among the metals tested. For Ti-6Al-4V and Ti-6Al-7Nb, there was no significant difference from the titanium with regard to Young's modulus ($p>0.05$).

Poisson's ratios of the alloys tested are displayed in Fig. 4. Changes in the ratios of the Ti-Au and Ti-Ag alloys were similar. As the concentration of gold or silver increased to 20%, Poisson's ratio increased, followed by a subsequent decrease in value. Between the titanium and the Ti-Au alloys, the ratios of Ti-10%Au and Ti-20%Au were significantly higher than that of titanium ($p<0.05$ and $p<0.001$, respectively). Between the titanium and the Ti-Ag

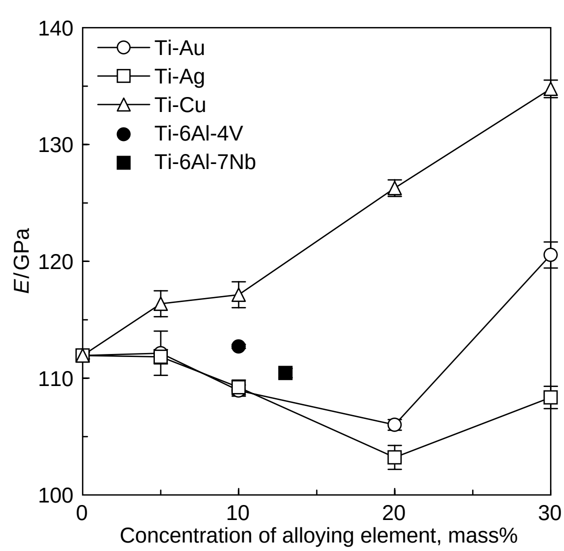

Figure 3 Young's moduli (E) of the titanium alloys. Error bar: $\pm$SD.

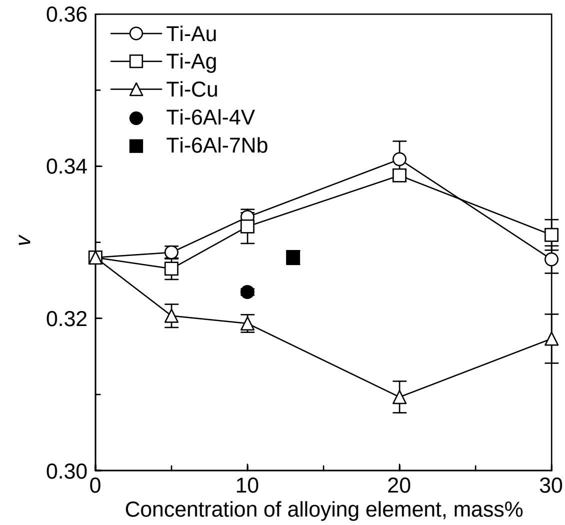

Figure 4 Poisson's ratios (ν) of the titanium alloys. Error bar: $\pm$SD.

alloys, the ratios of Ti-20%Ag were significantly higher than that of titanium ($p<0.001$).

As the concentration of copper increased to 20%, the Poisson's ratio of the Ti-Cu alloys decreased, followed by a subsequent increase in value. Between the titanium and Ti-Cu alloys, the ratios of all the alloys were significantly lower than that of titanium (5% Cu: $p<0.05$; 10% Cu: $p<0.01$; 20% Cu: $p<0.001$; 30% Cu: $p<0.01$). Ti-20%Au had the highest and Ti-20%Cu had the lowest Poisson's ratio of the metals tested.

Discussion

The alloy phases of as-cast Ti-Au, Ti-Ag, and Ti-Cu alloys previously studied using X-ray diffractometry (XRD) are summarized in Table 2 [17-19]. In the Ti-Au and Ti-Ag alloys, intermetallic compounds Ti_3Au or Ti_2Ag were found with 30% Au or 30% Ag, respectively. At the lower concentration, only the α phase was found in these two alloys, and intermetallic compounds either did not exist at all or the minimal amount of intermetallic compounds

Table 2 Alloy phases determined by X-ray diffractometry [17-19].

Alloy	Concentration of alloying element			
	5%	10%	20%	30%
Ti-Au	α	α	α	$\alpha+Ti_3Au$
Ti-Ag	α	α	α	$\alpha+Ti_2Ag$
Ti-Cu	α or $\alpha+Ti_2Cu$	$\alpha+Ti_2Cu$	$\alpha+Ti_2Cu$	$\alpha+Ti_2Cu$

that possibly existed made it difficult to detect anything using XRD. In the Ti-Cu alloys, an intermetallic compound (Ti_2Cu) was found with a copper concentration of 5 [18] or 10% [17] and more. The densities of the Ti-Au, Ti-Ag, and Ti-Cu alloys were virtually independent of the alloy phase within the single α phase structure or the eutectoid structure regions and were determined by the ratio of the number of constituent atoms (rule of mixtures) [20]. On the other hand, the Young's moduli and Poisson's ratios of the alloys were dependent on the alloy phase.

The densities of the titanium ($4.50\,\mathrm{Mg \cdot m^{-3}}$), Ti-6Al-4V ($4.43\,\mathrm{Mg \cdot m^{-3}}$), and Ti-6Al-7Nb ($4.52\,\mathrm{Mg \cdot m^{-3}}$) were in good agreement with the published values [2,21]. The densities of all the experimental alloys increased as the concentration of the alloying elements increased. The lower density of titanium than that of noble alloys is one point of difficulty when centrifugally casting this material [22]. In this respect, the increase in the density of titanium by alloying is preferable. However, the densities of the experimental alloys were still considerably lower than those of conventional dental casting alloys, such as gold and Co-Cr alloys. This is advantageous to make bulky maxillary prostheses lighter [23].

The Young's modulus of both Ti-Au and Ti-Ag alloys decreased as the concentration of the alloying element increased to 20%. It is known that the elastic moduli of metals change in proportion to the number of atoms of solute when the concentration of an alloying element is low [11,24,25]. The Young's moduli of gold and silver are reported to be 78 and 71 GPa [21], respectively, 30% or more lower than that of titanium (112 GPa). Although the alloy phases of Ti-Au and Ti-Ag alloys up to 20% Au or 20% Ag were all α, it is possible that their crystal structure changed to imperfect α by adding gold or silver, resulting in a lower Young's modulus.

The Young's moduli of Ti-30%Au and Ti-30%Ag were higher than those of the Ti-20%Au and Ti-20%Ag, respectively. The alloy phases changed from a single α phase to an α+intermetallic compound Ti_3Au or Ti_2Ag at 30% Au or 30% Ag. It is known that the elastic properties of two-phase materials follows the rule of mixtures and vary in a roughly linear manner between those of the two component phases [8,11,25,26]. Intermetallic compounds generally have higher elastic constants than the constituting elements [11,24,27]. Ti-Al and Ti-Sn alloys are reported to have a local maximum value of Young's modulus at the composition of the intermetallic compounds [11,28,29]. As an exception, a Ti-55%Ni alloy known as a shape-memory alloy is an intermetallic compound (TiNi) with a lower Young's modulus than both titanium and nickel [2,21]. Although the Young's moduli of the intermetallic compounds were not measured in the present study, it is quite possible that they are higher than those of the constituting elements and that the increases were caused by the precipitation of intermetallic compounds.

Unlike the Young's moduli of Ti-Au and Ti-Ag alloys, the Young's moduli of the Ti-Cu alloys monotonically increased with the concentration of copper. The most likely explanation is that the reported Young's modulus of copper is 128 GPa [21], above that of the titanium. What is more, the intermetallic compound Ti_2Cu, which probably has a higher Young's modulus than either titanium or copper, precipitated at a lower concentration of the alloying element, and/or the amount of compound was greater than that in the Ti-Au or Ti-Ag alloys.

Ti-6Al-4V is the best-known high-strength titanium alloy. Minimum requirements for the yield strength of annealed Ti-6Al-4V (780 MPa) [30] and Ti-6Al-7Nb (800 MPa) [31] are more than five times greater than that of grade 1 ELI (extra low interstitial) titanium (140 MPa) [32]. However, their Young's moduli were similar to that of titanium. This suggests that, although the yield strength of titanium is significantly increased by alloying, it does not necessarily follow that its Young's modulus will also increase [25]. Both Ti-6Al-4V and Ti-6Al-7Nb alloys contain a mixture of α and β phases at room temperature and are classified as $\alpha+\beta$ alloys [2]. Aluminum (α stabilizer) in these alloys increases the modulus, whereas, vanadium and niobium (both β stabilizers) decrease it [2,11]. The effects of these alloying elements on the Young's moduli of Ti-6Al-4V and Ti-6Al-7Nb were virtually cancelled.

The Poisson's ratios of all the alloys tested were approximately 0.3, which is typical for metals, including dental alloys [1,25,33]. It is known that the Poisson's ratio of many intermetallic compounds is lower than that of ordinary alloys [29]. Assuming that the intermetallic compound Ti_2Cu has a lower Poisson's ratio than titanium, the decrease in the Poisson's ratio of the Ti-Cu alloy as the concentration of copper increased can be explained by the increase in the amount of Ti_2Cu. The decreased Poisson's ratios of Ti-30%Au or Ti-30%Ag alloys from those at 20% Au or 20% Ag support the existence of the intermetallic compounds in these alloys.

It was found that alloying titanium with gold or silver slightly lowers the Young's modulus when the alloy phase is single α. Copper can be used as a moderate stiffener for titanium, but it does not increase the modulus to as much as those of Co-Cr alloys. It is evident that other properties should also be considered before applying the alloys in the present study to dentistry.

Acknowledgements

The authors are extremely grateful to Sumitomo Titanium Corporation, for providing the high-purity titanium sponge. The authors heartily thank the GC Corporation as well for providing the titanium alloys. This study was partially supported by a Grant-in-Aid for Scientific Research (B) (2) No. 12470411 from the Japan Society for the Promotion of Science.

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