Paper:

Reverse Lift-Off Process and Application for Cu-Zr-Ti Metallic Glass Thick Film Structures

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In technologies involving micro electromechanical systems, lift-off processes combined with sputter deposition are general patterning methods for the formation of amorphous alloy thick film structures. However, the thicknesses of structures fabricated in this manner are not uniform because sputtered particles are blocked by the sidewalls of the lift-off layer. In this paper, a reverse lift-off process is proposed as a new patterning method for fabricating amorphous alloy thick film structures of uniform thickness. In the reverse lift-off process, a template of the desired structure is formed on top of the chosen substrate. The thick film structure is then formed by sputter deposition on the top surface of the template. In contrast to a conventional liftoff process, here the thickness of the structure is uniform because there is nothing to hinder the sputtered particles. To demonstrate this process, we successfully fabricated a Cu-Zr-Ti metallic glass thick film structure with a uniform film thickness and a rectangular cross section across different target structure widths and thicknesses. This demonstrates that the reverse lift-off process is more suitable than conventional liftoff processes for the fabrication of metallic glass thick film structures.

Keywords: micro electro mechanical systems, micro process, lift-off process, copper alloy, amorphous

1. Introduction

In micro electromechanical systems (MEMS), amorphous alloy thin films are often used because of their high tensile strength, low Young's modulus, and desirable magnetic characteristics [1–3]. However, there are two problems in the fabrication of amorphous alloy thick film structures. The first problem is the deposition of the thick film, and the second problem is the thickness uniformity of the thick film structures.

The first problem arises because amorphous alloys tend to crystallize at high temperatures. Sputter deposition is a general purpose deposition method for creating thinner amorphous alloy films. When depositing thicker amorphous alloy films, however, longer sputtering times and/or higher sputtering powers are required. As a result, the amorphous alloy can crystallize because of the heat generated by the sputtering process. To solve this problem, the formation of films thicker than 5 μ m has been investigated using a metallic glass that has a high thermal stability for an amorphous alloy [4–7].

In MEMS technologies, one general patterning method for fabricating thick film structures is to use a lift-off process with a photoresist layer [8]. However, the photoresist layer can outgas when heated for a long time or during high-power sputtering [9]. If the metallic glass incorporates this outgassed oxygen, it can easily crystallize because oxygen promotes nucleation [10]. Fabricating a metallic glass thick film structure requires a method that generates no oxygen, such as a lift-off process using metallic materials that do not outgas [11, 12].

The second problem is the non-uniform thickness of patterned film structures formed using a conventional liftoff process, because the deposition of sputtered particles is hindered by the sidewalls of the lift-off layer. If the aperture of the lift-off layer is wide, the cross-sectional shape of the structure becomes rectangular, as the deposition of sputtered particles is hindered only in areas near the edges of the aperture. If the aperture is narrow, the cross-sectional shape of the structure will deviate from a rectangular shape because the deposition of sputtered particles can be hindered even at the center of the aperture. In contrast, sputtered particles that land on top of the lift-off layer are not blocked by the sidewalls of the lift-off layer, because the lift-off layer rests atop the substrate. Thus, the thickness of the thick film structure is uniform and its cross section is rectangular.

In this paper, a novel method of fabricating amorphous alloy thick film structures is proposed, one that is more suitable for obtaining uniform thicknesses and rectangular cross sections in the produced structures. The method is called the "reverse lift-off process." In this process, a template of the desired structure is formed on top of a substrate. Then, a thick film structure of uniform thickness is formed on the template via sputter deposition. As a practical investigation of the reverse lift-off process, which is explained in detail in Section 3, a Cu-Zr-Ti metallic glass thick film structure was fabricated from the template. The results obtained from using this reverse lift-off process were compared with those from a more conventional liftoff process.



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(b) Lift-off **Fig. 1.** Schematic diagram of a conventional lift-off process.

2. Conventional Lift-Off Process

The lift-off process, shadow mask process, and convex process [13] have all been studied as patterning methods for amorphous thin films. Among them, the lift-off process with a photoresist layer is the most commonly used in MEMS fabrication because the photoresist lift-off layer can form a nanoscale pattern with high dimensional accuracy.

Figure 1 shows a schematic view of a conventional sputtering and lift-off process. A thick film structure is formed in the aperture of the lift-off layer, but as stated previously the sputtered particles are not deposited uniformly on the substrate because some of them are blocked by the lift-off layer. The thickness of the structure increases in the center of the aperture because sputtered particles can easily reach the substrate surface. In contrast, the thickness of the structure decreases at the edges of the aperture because the sputtered particles cannot reach the surface of the substrate as easily.

If the aspect ratio (height/width) of the aperture is low, the thickness of the structure decreases solely at the edges of the structure, and the cross-sectional shape becomes more rectangular. At high aspect ratios however, the cross-sectional shape of the structure will not be rectangular because the thickness of the entire structure decreases. Therefore, the thickness and cross-sectional shape of the structure will vary in different regions depending on the width of the target structure.

A thin film is formed on the sidewalls of the aperture of the lift-off layer due to the adhesion of sputtered particles. This thin film connects the thick film formed on the liftoff layer and the thick film formed in the aperture of the lift-off layer. It is difficult to remove (or lift off) the liftoff layer with a remover as the lift-off layer is covered by both the thin film and the thick film. However, it is possible to remove the lift-off layer if its height is greater than twice the thickness of the thick film formed in the aperture of the lift-off layer [14, 15]. When the aspect ratio of the aperture of the lift-off layer is large enough, sputtered particles tend to not deposit on the sidewalls near the thick film, and the lift-off layer is not covered perfectly by the thin film and the thick film.



(d) Separation of unnecessary sections

Fig. 2. Schematic diagram of a reverse lift-off process.

3. Reverse Lift-Off Process

In the conventional lift-off process, sputtered particles that encounter the top surface of the lift-off layer are not blocked by the sidewalls of the lift-off layer, because the lift-off layer rests on top of the substrate. The thickness of the film deposited on this top surface is uniform, and its cross-sectional shape is rectangular. This phenomenon is utilized in the "reverse lift-off process." In the reverse liftoff process, a template of the desired structure is formed on top of a pre-fabricated substrate. Then, a thick film structure of uniform thickness is formed on top of the template by sputter deposition.

Figure 2 shows a schematic view of the reverse liftoff process, and **Fig. 2(a)** shows examples of various templates. Structures are created on the top surface of the substrate (either by top-down or bottom-up design) to form a template of the desired thick film pattern. It is necessary that the height of the template be more than twice the thickness of the thick film structure formed in the template openings [14, 15]. This will prevent the thick film structure formed on the top surface from connecting to the material deposited in the template openings. The template can be formed using a method in which a photoresist layer or metallic material is deposited on the substrate (discrete type) or by etching of a Si wafer or metal substrate into the appropriate design (integrated type).

Figure 2(b) shows a diagram the sputtering process. A thick film is deposited onto the template via sputter depo-



(e) Completion

Fig. 3. Implementation of the reverse lift-off process used in this study.

sition, yielding a uniform thickness. **Fig. 2(c)** shows the template separation process. One example of a separation method is the removal of the template using an etchant. It is also possible to separate the structure using a sacrificial layer deposited in advance on the template surface. A film structure formed by this process consists of a thick film section with uniform thickness and thin film sections on the underside edges of the thick film.

The thin film sections on the underside edges of the thick film are unnecessary for a thick film structure with uniform thickness. These sections are thinner and more fragile compared to the thick film sections because the sputtered particles tend to deposit very less on the side-walls of the template, relative to the amount that deposit on the top surface of the template. **Fig. 2(d)** shows the separation process of these sections, where they are broken and separated by an external force. Through utilizing all of these processes, a thick film structure with uniform thickness can be obtained.

Figure 3 shows the implementation of the reverse liftoff process used in this paper. Fig. 3(a) shows a template formed by a difference in heights between the substrate and the spacer units. The substrate is processed in advance to form a template of the desired thick film structure. Fig. 3(b) shows the sputtering of a sacrificial layer and the thick film, where a thick film structure of uniform thickness is formed on top of the template and the spacer prevents the sputtered particles from adhering to the sidewalls of the substrate. Fig. 3(c) shows the spacer removal process, where the sacrificial layer is exposed by removal of the spacer. Fig. 3(d) shows the sacrificial layer removal process, which involves immersion in an acidic or alkaline etchant with ultrasonic vibration. The unnecessary lower sections of the structure are easily broken and separated by etching and ultrasonic vibration due to

their thinner and more fragile makeup. However, residual segments remain on the edges of the backside of the thick film structure, forming small burrs. Using these processes in sequence, a thick film structure of uniform thickness with a rectangular cross section was produced as shown in **Fig. 3(e)**.

4. Cu-Based Metallic Glass

In this paper, a thick film structure of Cu-Zr-Ti metallic glass was fabricated using these techniques. Cu-Zr-Ti was selected because Cu-based metallic glass is a promising material for future use in MEMS due to its high tensile strength and fatigue strength [16, 17]. Here, a Cu-Zr-Ti metallic glass thick film was deposited using radio-frequency magnetron sputtering (Sanyu Electron Co., SVC-700RF). The Cu-Zr-Ti metallic glass target used for the sputter deposition was made from an oxygenfree copper ingot, zirconium ingot and a titanium ingot using an arc melting method.

The physical properties of the Cu-Zr-Ti metallic glass were measured using a film sample that was 5 μ m thick. The Young's modulus of the sample was 62.3 GPa, and the tensile strength was 0.9 GPa, as measured by tensile tests at room temperature using a thermomechanical analyzer (TMA; Rigaku TA-60). The composition of the sample was Cu64Zr28Ti8 (at%) according to energy dispersive X-ray spectroscopy (EDX; Shimazu μ EDX-1200). The glass transition temperature of the sample was 734 K and the crystallization temperature was 768 K, as measured by differential scanning calorimetry (DSC; UL-VAC DSC9400).

5. Fabrication of Cu-Zr-Ti Metallic Glass Thick Film Structure and Evaluation of its Shape

A Cu-Zr-Ti metallic glass thick film structure was fabricated using a reverse lift-off process. Fig. 4 shows the sputtering jig used to fabricate the structure. As shown in Figs. 4(a)-(c), the sputtering jig was assembled by inserting spacer units into a stainless steel substrate. Fig. 4(d) shows an enlarged photo of one section of the template shown in Fig. 4(c). Fig. 5 shows the dimensions of the template in the areas labeled A and B in Fig. 4(c). The Cu-Zr-Ti metallic glass thick film structure was formed on this template.

Figure 6 shows the areas on the sputtering jig marked A and B in **Fig. 5**, after a 0.1 μ m-thick Cr adhesion layer, a 6 μ m-thick Cu sacrificial layer, and a 10 μ m-thick Cu-Zr-Ti metallic glass layer were deposited.

Figure 7 shows the thickness of the Cu-Zr-Ti metallic glass thick film structure, as measured by scanning white light interferometry (Canon Inc., Zygo New View 5032) with the template shown in **Fig. 6** (widths of 0.1 mm in the C-C' area, 0.2 mm in the D-D' area, 0.3 mm in the E-E' area, 0.5 mm in the F-F' area, 0.7 mm in the G-G' area, 0.8 mm in the H-H' area, and 1.0 mm in the I-I' area). As











(b) Schematic diagram of Area B (Fig. 4(c)) Fig. 5. Template dimensions.

shown in **Fig. 7**, a Cu-Zr-Ti metallic glass thick film structure of uniform thickness with a rectangular cross section was obtained regardless of the width of the target structure.



(a) Area A



(b) Area B **Fig. 6.** Substrate after sputtering the Cu-Zr-Ti metallic glass.

Figure 8 shows a Cu-Zr-Ti metallic glass thick film structure separated from its substrate by immersion in nitric acid for 12 hours. During the separation process, the structure was exposed to ultrasonic vibrations at 100 kHz to promote separation from the substrate. Fig. 8(a) shows an obverse view of the structure, and Fig. 8(b) shows a reverse view of the structure. The area marked L in Fig. 8(b) is the other side of the area marked J in Fig. 8(a). Similarly, area M is the reverse view of area K. Figs. 8(c) to 8(f) show areas J-M in Figs. 8(a) and 8(b). As shown in Fig. 8, the fabrication of a Cu-Zr-Ti metallic glass thick film structure was successful.

Figure 9 shows a burr that remained on one edge of the reverse side of the structure. Fig. 9(a) shows an enlarged view of a burr in area N of Fig. 8(d). Fig. 9(b) shows the shape of the burr, as measured by a stylus-type surface profile instrument (ULVAC Dektak150) in the O-O' region of Fig. 9(a). The width of the burr was 17.7 μ m, and its height was 5.1 μ m.

The cross-sectional area of the burr was approximately 6.1% of the smallest cross-sectional shape in the structure (0.1 mm wide by 10 μ m high). The burr size was negligible compared with the overall Cu-Zr-Ti metallic glass thick film structure. Based on these observations, a Cu-Zr-Ti metallic glass thick film structure of uniform thickness with a rectangular cross section was successfully fabricated.



Fig. 7. Thickness of the Cu-Zr-Ti metallic glass.

(b) Reverse view

(d) Area L



(a) Obverse view



(c) Area J



(e) Area K (f) Area M **Fig. 8.** Cu-Zr-Ti metallic glass thick film structure.



Fig. 9. Burr thickness on the Cu-Zr-Ti metallic glass thick film structure.



Fig. 10. Cu-Zr-Ti metallic glass thick film structures with different film thicknesses.

6. Fabrication of Thicker Films

It is necessary to confirm the thickness uniformity and cross-sectional shape of any amorphous alloy thick film structure thicker than 10 μ m, because the rectangular geometry of the structure cross-section can be affected by larger burrs caused by the increased thickness of the structure. Therefore, we investigated the formation of thicker films using the reverse lift-off process.

Figure 10 shows Cu-Zr-Ti metallic glass thick film structures with thicknesses of 10 μ m or 30 μ m, and widths of 0.2 mm or 1.0 mm. These structures were fabricated and measured as described in Section 5. The thicknesses of the structures thicker than 10 μ m were as uniform as those of the 10 μ m-thick structure.

Figure 11 shows burrs on structures that were $10 \,\mu m$ or



Fig. 11. Comparison of burrs on Cu-Zr-Ti metallic glass thick film structures with different film thicknesses.

30 μ m thick. These burrs were measured using a stylustype surface profile measuring instrument, as shown in Fig. 9. In the 10 μ m-thick structure, the cross-sectional area of a burr is 50.2 μ m², while for the 30 μ m-thick structure the cross-sectional area of a burr is 130.4 μ m². Therefore, when the thickness of the structure was tripled (from 10 μ m to 30 μ m), the cross-sectional area of burr increased approximately 2.6 times. The volume of the burr relative to the volume of the structure decreased with increasing thickness of the structure. Therefore, the shape of the structure should not deteriorate with increasing thickness of the structure. As stated above, it is possible to fabricate Cu-Zr-Ti metallic glass thick film structures of uniform thickness with rectangular cross-sections, even if the structures themselves feature variable widths and thicknesses.

7. Comparison with Conventional Lift-Off Process

We have previously studied the shape of a Cu-Zr-Ti metallic glass thick film structure made using a conventional lift-off process [12]. Fig. 12 shows a schematic of the conventional lift-off process using a 100 μ m-thick KMPR 1035 photoresist layer (Nippon Kayaku Co., Ltd.).

The Cu-Zr-Ti metallic glass thick film structure was formed via sputter deposition and a conventional lift-off process in the aperture of a KMPR 1035 lift-off layer. The structure consisted of a 50 nm-thick Ti adhesion layer, an approximately 10 μ m-thick Cu-Zr-Ti metallic glass layer, and a 100 nm-thick Au antioxidant layer. When the photoresist layer was removed, the Cu-Zr-Ti metallic glass thick film structure was complete. The Au antioxidant layer prevented corrosion of the Cu-Zr-Ti metallic glass in the etchants used to remove the KMPR 1035 photoresist layer [18, 19].

Figure 13 shows the cross-sectional shapes of Cu-Zr-Ti metallic glass thick film structures formed using either the reverse lift-off process or a conventional lift-off process. The shapes were measured by scanning white light interferometry. The structures were approximately 10 μ m



Fig. 12. Conventional lift-off process with a KMPR1035 photoresist layer.



Fig. 13. Comparison of cross-sectional shapes of Cu-Zr-Ti metallic glass thick film structures.



Fig. 14. Thicknesses of Cu-Zr-Ti metallic glass thick film structures formed using two different processes.

thick and 0.1 mm, 0.2 mm, 0.5 mm, or 1.0 mm wide.

The structures formed using the reverse lift-off process had the same design as the samples discussed in Section 4. The deposition conditions were the same in the sputtering portion of each lift-off process. Using scanning white light interferometry, we were unable to measure the side surface shape of the sample formed using each lift-off process. This was because scanning white light interferometry is used to measure the shape of an object based on reflected light, but very little light reflected from the nearly vertical side surfaces of the structure [20]. In comparison with upper the surface shapes of the samples, the cross-sectional shapes of the samples formed using a conventional lift-off process had more curved surfaces than those of samples formed using the reverse lift-off process.

Figure 14 shows the thicknesses of Cu-Zr-Ti thick film structures formed with either a conventional lift-off process or the reverse lift-off process. In the conventional lift-off process, when the structure is narrower, its thickness decreases. This is because fewer sputtered particles reach the substrate as many are blocked by the KMPR 1035 lift-off layer when the aperture is narrow. In the reverse lift-off process, even if the Cu-Zr-Ti metallic glass thick film structure is narrow, the thickness of the structure will be uniform and its cross section will be rectangular.

Figure 15 shows X-ray diffraction (XRD) patterns for Cu-Zr-Ti thick film structures made using a conventional lift-off process or the reverse lift-off process. An imaging plate X-ray diffraction device (Rigaku Rint-rapid) was used for the measurements. The sample fabricated using the conventional lift-off process was measured before sputtering of the Au antioxidant layer. Sample fabrication using the reverse lift-off process was performed as described in Section 5. The Cu-Zr-Ti thick film structure made using the reverse lift-off process was amorphous; in contrast, the structure formed using the conventional liftoff process was crystalline.

Metallic glasses are easily crystallized because nucleation is promoted when the metallic glass absorbs oxygen,





Fig. 15. Comparison of XRD patterns of Cu-Zr-Ti metallic glass thick film structures.

and the raw material of the KMPR 1035 photoresist layer is an epoxy resin. We assume that the KMPR 1035 photoresist layer generated oxygen gas when the heat from the sputter deposition increased its temperature, causing the Cu-Zr-Ti metallic glass to crystallize. The reverse liftoff process using a metallic template is better suited for the fabrication of metallic glass thick film structures because the process does not rely on materials that can generate oxygen.

Unlike the conventional lift-off process, the reverse liftoff process can be used to fabricate Cu-Zr-Ti metallic glass thick film structures with uniform thickness and a rectangular cross section. The reverse lift-off process is therefore more suitable for the fabrication of metallic glass thick film structures than the conventional lift-off process.

8. Conclusion

A novel method of fabricating amorphous alloy thick film structures of uniform thickness with rectangular cross sections, called the "reverse lift-off process," was proposed. In the reverse lift-off process, a template of the desired structure is formed on top of the substrate. A thick film structure of uniform thickness is then formed on this template by sputter deposition. As a practical demonstration of the reverse lift-off process, different templates were formed with different distances between the substrate and the spacer unit. The substrate and spacer unit were made from stainless steel to avoid outgassing. A Cu-Zr-Ti metallic glass thick film structure was fabricated from the template using the reverse lift-off process, and the reverse lift-off process.

Several different Cu-Zr-Ti metallic glasses of uniform thickness with rectangular cross-sections were successfully fabricated, with variable structure widths and thicknesses. The reverse lift-off process is more suitable for the fabrication of metallic glass thick film structures than the conventional lift-off process because it avoids crystallization of the film and results in structures with better thickness uniformity and more rectangular cross-sections.

9. Future Work

Using the reverse lift-off process, it is possible to fabricate thick film structures of uniform thickness that could not otherwise be fabricated using conventional MEMS processes. However, it is necessary to further examine the following problems before the reverse lift-off process can be universally applied as a MEMS fabrication method. The first problem is to reduce the width of the thick film structures from the millimeter-scale to the micro- or nanoscale. The second problem is the integration of the thick film structures into electric circuits. Using the reverse liftoff process, the fabrication of integrated MEMS devices with both electrical components and mechanical components made from thick film structures of uniform thickness becomes more feasible. The third problem is to attach electrical components to the thick film structure before separating the thick film structure from the substrate. MEMS with thick film structures can offer increased productivity if they are fixed to a substrate because a substrate with multiple integrated MEMS can be processed all at once in a single process.

A novel reverse lift-off process applicable to various MEMS devices will be proposed based on future investigations of the above three problems.

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