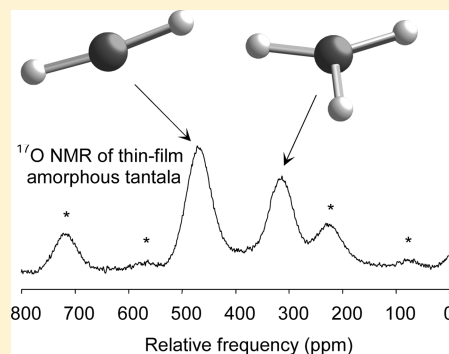


Structure of Amorphous Tantalum Oxide and Titania-Doped Tantalum Oxide: ^{17}O NMR Results for Sol–Gel and Ion-Beam-Sputtered Materials

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ABSTRACT: Amorphous thin films of high-refractive index metal oxides such as Ta_2O_5 are critical in multilayer optical components such as the main mirrors of the Laser Interferometer Gravitational-Wave Observatory (LIGO), but their atomic-scale structures are not well-known. Amorphous, pure, and titania-doped tantalum oxides were prepared by sol–gel synthesis and by ion-beam sputtering. The samples were successfully enriched with the nuclear magnetic resonance (NMR)-active ^{17}O isotope by heating them in $^{17}\text{O}_2$ gas, and they were studied using ^{17}O solid-state NMR. ^{17}O MAS NMR spectra of pure tantalum show two resonances that correspond to two- and three-coordinated oxygens, allowing the average oxygen coordination numbers to be estimated from the relative peak areas. The average coordination number for oxide ions seems to increase as tantalum changes from amorphous to crystalline. Titania-doped tantalums, $(\text{Ta}_{1-x}\text{Ti}_x)_2\text{O}_{5-x}$ with $x = 0.10, 0.25,$ and 0.50 , were also studied. At the lower two dopant levels, Ta and Ti cations are well-mixed in the sense that TiO_2 -like domains are not observed, but Ti is preferentially associated with three-coordinated oxygens. This increases the mean oxygen coordination slightly, which may be related to previously reported improvements in thermal noise characteristics of materials for multilayer interferometer mirror coatings. ^{17}O MAS NMR spectra of thin-film tantalum deposited by ion-beam sputtering and ^{17}O -enriched after synthesis are very similar in resonance positions and relative peak ratio (and, thus, average oxygen coordination number) to data for the sol–gel materials, which suggests that the amorphous structure may be similar, regardless of the preparation methods when materials are annealed at $450\text{--}500\text{ }^\circ\text{C}$.



KEYWORDS: solid state NMR, Ta_2O_5 , tantalum, LIGO, sol–gel, optical coating

INTRODUCTION

The short-range structures of “normal” oxide glasses (especially silicates, borosilicates, and phosphates), typically formed by cooling from a melt, are relatively well-understood after decades of spectroscopic, X-ray, and neutron scattering studies. High-resolution solid-state nuclear magnetic resonance (NMR) has played an important role in this elucidation, as such materials often contain cations of nuclides that readily yield information-rich spectra (e.g., ^{11}B , ^{27}Al , ^{29}Si , and ^{31}P). More recently, entire families of amorphous oxides of heavier metals have been developed that are usually formed as thin films by various vapor deposition and sputtering methods and have widespread and valuable technological applications. The structures of such materials are largely unknown for a variety of technical reasons, but they are potentially especially interesting, because they fall outside of the classical conceptual realm of “network” glasses, usually entirely lacking “network forming” cations. Applications of NMR to such materials have been particularly limited, as their constituent metal cations usually lack “accessible” NMR nuclides. However, amorphous heavy-metal oxides do share a potential NMR nuclide with light-element glasses, which has been proven to be very informative in recent studies, namely, ^{17}O . However, the necessary enrichment in this isotope can be challenging, particularly in materials such as these, which readily crystallize upon heating.

In particular, metal oxide thin films are widely used in optical applications such as high-reflective or antireflective coatings. These

often consist of alternating layers of amorphous oxide materials with a high refractive index and with a low refractive index. The latter is often silica, the structure of which (at least as a bulk glass) has been very widely studied. However, the structures of the heavier transition-metal oxides often used for the high-index layers are much less well-known, in part because they often can only be synthesized as thin films.

Amorphous tantalum oxide ($\text{a-Ta}_2\text{O}_5$) is often used as a high-refractive-index coating material for high-performance multilayer mirrors. A notable example is the type of mirror at the heart of the Laser Interferometer Gravitational-Wave Observatory (LIGO),^{1,2} which is made of alternating $\lambda/4$ -thick layers of $\text{a-Ta}_2\text{O}_5$ and amorphous SiO_2 . $\text{a-Ta}_2\text{O}_5$ also has been studied as a material with a high dielectric constant in memory devices and other electronic applications. In particular, titania-doped Ta_2O_5 shows a much increased dielectric constant.³

Despite its excellent optical performance, the $\text{Ta}_2\text{O}_5/\text{SiO}_2$ coating has been identified as an important source of thermal noise in LIGO mirrors.⁴ Careful study with different numbers and thicknesses of alternating layers of amorphous Ta_2O_5 and SiO_2 showed that the noise originated mainly from the tantalum.⁵ Therefore, there has been much effort to understand this

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phenomenon and thus to minimize the thermal noise: for example, the addition of titanium oxide to amorphous tantalum is found to decrease the mechanical loss significantly.⁶

To understand the mechanism of the elastic dissipation that causes the thermal noise and to improve mechanical properties, a more complete view of the local atomic structure of tantalum coatings would be very helpful. However, this type of information is not readily available, since conventional structural tools, such as diffraction methods, generally provide information only on average structure. For example, an X-ray diffraction (XRD) study of anodic amorphous tantalum films concluded that (i) their average structure, overall, was similar to that of crystalline Ta₂O₅, but (ii) mean Ta–O distances in the latter were slightly longer.⁷ This would probably correspond to a slightly higher mean Ta coordination number and density in the crystal. A variety of methods, including XRD, Fourier transform infrared spectroscopy (FTIR),⁸ X-ray photoelectron spectroscopy (XPS) and electron microscopy,⁹ and micro-Raman spectroscopy¹⁰ have been applied to assess crystallinity and homogeneity of thin-film amorphous tantalum, but little is known about atomic structure. A very recent report used electron diffraction by transmission electron microscopy (TEM) to obtain the averaged reduced density function for tantalum films, which were analyzed by reverse Monte Carlo refinement and molecular dynamics (MD) simulations to obtain insights into short-range structure.¹¹

NMR has been successfully used to study many different types of amorphous solids, especially silicate and borosilicate glasses,¹² because this method is inherently sensitive to local structure, even in such disordered systems. However, very little NMR work on the structures of amorphous transition-metal oxides has been reported. This, in part, is because thin-film synthesis methods generally yield relatively small amounts of material (tens of milligrams) that must be removed from the deposition substrate for typical NMR experiments. Even more importantly, many such materials as normally prepared (including tantalum) lack a convenient NMR-active nuclide whose spectra are as well-resolved and informative as those typically studied in lighter-element oxide glasses (e.g., ¹¹B, ²³Na, ²⁷Al, ²⁹Si, and ³¹P). However, the common element in all amorphous oxides is oxygen, for which the NMR-observable nuclide (¹⁷O) is present at its low natural abundance of only 0.037%. If ¹⁷O can be enriched in such materials, either during or after deposition, high-resolution NMR spectra may be obtainable that can provide unique and quantitative information about local structure around the oxide anions. Here, we present the first such study of amorphous pure and titania-doped tantalum oxides.

EXPERIMENTAL SECTION

Pure and titania-doped amorphous tantalum oxides were initially synthesized by the sol–gel method. Tantalum ethoxide (Ta(OEt)₅) and titanium ethoxide (Ti(OEt)₄) were dissolved in ethanol, with 0.5 M cation concentration. These were then mixed with 0.5 M water in ethanol solution to yield a cation:water ratio of 1:4. A small amount of HCl(aq) was also added to the solution to catalyze the oxide-precipitating reaction. The solutions were stirred in air until they were dry, usually requiring one to two days. The dry samples were subsequently heated with 40% ¹⁷O-enriched oxygen gas in an open gold tube inside of a sealed silica glass tube at a temperature of 500 °C for one day; portions of the 50% titania-doped tantalum were heated at 500 °C or at 700 °C for one day. Amorphous tantalum oxide was also prepared by hydrolysis of tantalum ethoxide, Ta(OEt)₅, with 46.3% ¹⁷O enriched water with a cation:water ratio of 1:4. The sample was dried in air at ambient temperature for about a

week and 120 °C for 12 h. The sample then was annealed at 400 °C for 2 h in air and then at 500 °C for 2 h in an argon atmosphere.

Thin-film Ta₂O₅ was also prepared by ion-beam sputtering (IBS) by MLD Technologies (Mountain View, CA). The films were deposited with thicknesses of 3 μm on 2.5-cm-diameter NaCl substrates. The films separated spontaneously from the substrates after coating. Film fragments were collected and enriched with ¹⁷O by heating with 70% ¹⁷O-enriched oxygen gas as above, at 450 °C for 3 days.

Powder XRD was used to confirm whether the resulting compounds were amorphous or crystalline using a Rigaku X-ray diffractometer, comparing the XRD patterns with the International Center for Diffraction Data files.

¹⁷O MAS NMR spectra of sol–gel-synthesized and ion-beam-sputtered tantalum were collected at 81.29 MHz, using a Varian Unity/Inova 600 spectrometer with a Varian T3MAS probe with 3.2-mm zirconia rotors (22 μL sample volume). Given the high density of tantalum (~7.7 g/cm³),¹¹ sample weights were typically 60–80 mg. Single-pulse experiments were carried out with a radio-frequency (rf) pulse length of 0.30 μs at spinning speeds of 20 kHz. The pulse length corresponding to an rf tip angle of π/18 for the liquid reference was used to ensure that the peak intensities would be quantitative, regardless of the size of the quadrupolar coupling constants. With a similar probe and a Varian 800 spectrometer, data were collected for some samples at 108.4 MHz. To obtain better separation of spinning sidebands in the more-complex spectra of sol–gel-synthesized titania-doped tantalum, data were also collected at 54.19 MHz, using a Varian Infinity Plus 400 spectrometer, with 0.28 μs pulses (corresponding to π/18 for the liquid reference) at spinning speeds of 20 kHz. All spectra were referenced at 0 ppm to ¹⁷O enriched water. Delays between pulses were typically 0.1–1 s; no differential relaxation among different resonances was observed. The relatively well-resolved resonances for oxide ions with varying first-neighbor cations had Gaussian peak shapes at all three fields, indicating a dominance of chemical shift distribution over quadrupolar broadening. Peak areas were thus determined by fitting the central resonances of the spectra with Gaussian line shapes. To take into account the contribution of satellite transition to the center peak intensity, 20% and 10% intensities (based on the first sideband intensity) were subtracted from two- and three-coordinated oxygen intensities.

RESULTS AND DISCUSSION

Pure Tantalum. Portions of the sol–gel-synthesized amorphous tantalum (normal isotopes) were heated at 300, 400, 500, 600, and 700 °C for 2 h and then examined by powder XRD (see Figure 1). At temperatures lower than 500 °C, no peaks from crystalline tantalum were detected. At 500 °C, the crystalline peaks start to appear and become clearly visible at 600 °C. The crystalline peaks dominated the XRD pattern of the 700 °C sample. In general, sol–gel-synthesized oxides must be heated at an elevated temperature to remove the residual water and organic residue after initial drying. It appears that sol–gel-synthesized tantalum remains amorphous until ~500 °C and starts to crystallize rapidly at higher temperatures.

When the tantalum and titania-doped tantalum samples were enriched with the ¹⁷O isotope for NMR experiments, the enrichment was done by heating in ¹⁷O₂ gas at 500 °C. Powder XRD confirmed that samples remained mostly amorphous. The ¹⁷O MAS NMR results (below) show that this enrichment process was successful, and that the spectra are consistent with previous results¹³ for which the amorphous tantalum was enriched with ¹⁷O-enriched water during the initial sol–gel reaction.

In the ¹⁷O MAS NMR spectra for both crystalline and amorphous tantalum, two broad resonances were found (widths

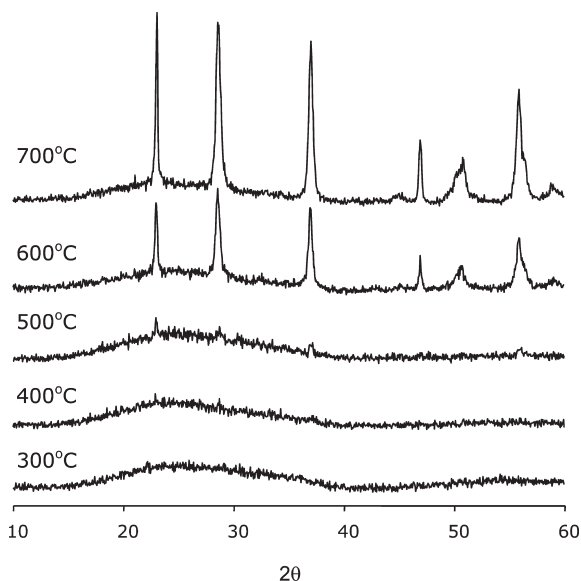


Figure 1. Powder X-ray diffraction (XRD) results for pure tantalum oxides annealed at various temperatures.

of ~ 30 – 60 ppm), centered at ~ 310 and 470 ppm (Figure 2). These are similar to, but slightly higher in frequency than, peaks reported in previous work on sol–gel tantala at 270 and 430 ppm,¹³ perhaps because the latter were derived from somewhat noisier spectra. Our data from multiple fields (see below) show little effect on peak position, because of small quadrupolar coupling constants, indicating that the difference between the two studies cannot be attributed to such interactions. In our spectra of crystalline tantala, these two resonances have a relative area ratio of $\sim 3:2$.

Crystalline tantala has at least two polymorphs—low and high temperature—and is known to undergo a phase transition at 1360 °C. The XRD powder pattern of the room-temperature phase is complicated, because of the slight differences in superstructure lines that are dependent on the thermal history or preparation routes. Comparing two of the available complete refined structures, L-Ta₂O₅ and T-Ta₂O₅, the overall structure seems to be quite similar, especially the relative ratio between two- and three-coordinated sites.^{14–16} We base our comparison on the L-Ta₂O₅ structure from Stephenson and Roth,¹⁶ since the XRD pattern of the crystalline tantala in this study agrees well with it. The complex X-ray structure (33 distinct oxygen sites, 12 Ta sites) shows that both three-coordinated and two-coordinated oxygen are present, in a ratio of $\sim 3:2$ when the long bonds were included (<3.0 Å). By comparing the relative oxygen site populations in the X-ray crystal structure with the NMR peak areas, the resonances at 310 and 470 can be assigned to three- and two-coordinated oxygen, respectively. This assignment, where the lower-frequency resonance is attributed to the site with higher coordination number, is consistent with the previous results for sol–gel tantala and (Ta₂O₅)_x(SiO₂)_{1-x},¹³ and related compounds such as Nb₂O₅¹⁷ and mesoporous Ta and Nb oxides.¹⁸ The peak at 310 ppm seems to have a high-frequency shoulder, which can probably be attributed to three-coordinated oxygens ($\sim 10\%$ of total oxygen) with a longer bond than average (>2.5 Å). Two-coordinated sites have fewer structural variables, where each oxygen has equal distances from coordinating tantalums with the Ta–O–Ta bond angles of 160° – 180° ,

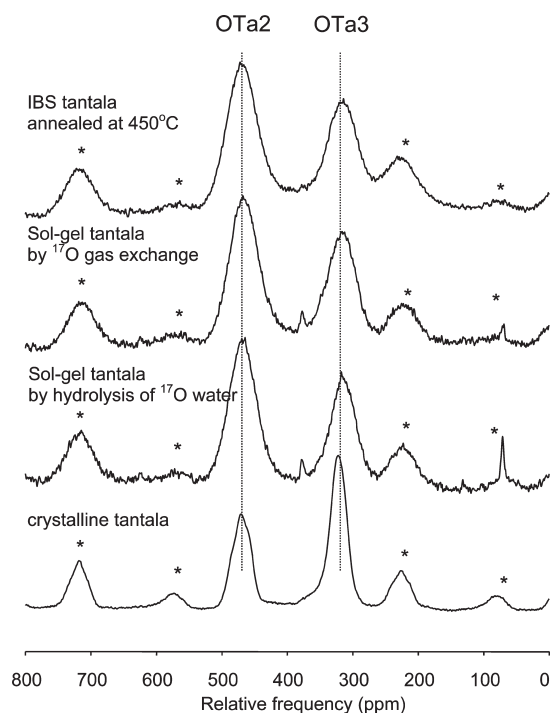


Figure 2. ¹⁷O MAS NMR spectra of pure amorphous tantala prepared via the sol–gel method (enriched with ¹⁷O by gas exchange), tantala prepared by hydrolysis with ¹⁷O water, and ion-beam sputtered thin film and crystalline tantala samples at 14.1 T. The spinning speeds were 20 kHz; spinning sidebands are marked with an asterisk (*). The narrow resonance at ~ 70 ppm is due to a small amount of an unknown contaminant and the small, sharp peak near 375 ppm is the natural-abundance ¹⁷O background signal from the zirconia rotor.

which may be grouped with bond distance and angles and may result in some structure of the peak at 470 ppm, while three-coordinated sites may have a more complex distribution of bond distances and angles. Data from lower field (9.4 T, not shown) do not show any significant difference in resonance positions (<3 to 5 ppm) and linewidths for either the crystalline or amorphous tantala. This indicates that the quadrupolar interaction (C_Q) is small in these compounds (<2 MHz), which suggests that Ta–O bonds have a predominantly ionic character. This also means that variations in C_Q from the amorphous to the crystalline state are not the most likely explanation for observed large differences in the relative areas of the two resonances.¹³

¹⁷O NMR spectra of the amorphous tantala samples also show two peaks, at positions similar to those of the crystalline phase (Figure 2). However, the ratio between three-coordinated and two-coordinated oxygen sites is $\sim 2:3$, while that of crystalline tantala is $\sim 3:2$. The similarity in resonance positions suggests that the coordination environments in the amorphous material are similar or closely related to those of the crystalline phase. However, there are clearly considerably more two-coordinated oxygen sites than three-coordinated sites in amorphous tantala, while the reverse is the case for crystalline tantala. Spectra of amorphous tantalas enriched by exchange with ¹⁷O gas for various times and at various temperatures are all very similar in peak positions and relative intensities, as is that of amorphous tantala enriched by hydrolysis with ¹⁷O water, when the final heating temperatures were 500 °C. Therefore, there is no evidence for differential rates of exchange of ¹⁷O between the three- and

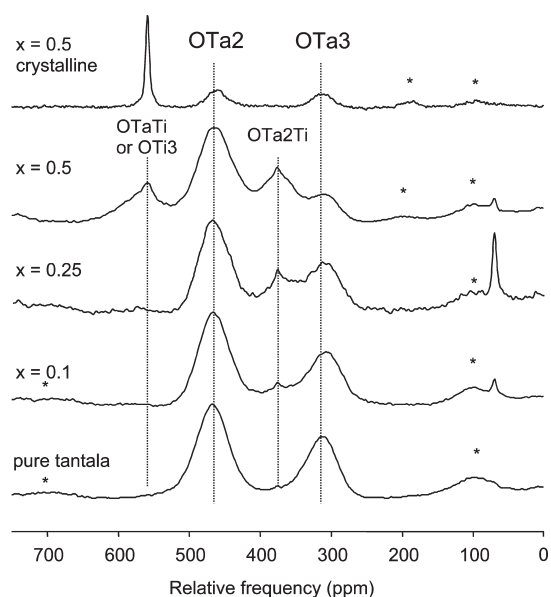


Figure 3. ^{17}O MAS NMR spectra of Ti-doped tantalum samples at 9.4 T. The spinning speeds were 20 kHz; spinning sidebands are marked with an asterisk (*). The narrow resonance at ~ 70 ppm is due to an unknown contaminant and the small, sharp peak near 375 ppm is the natural-abundance ^{17}O background signal from the zirconia rotor.

two-coordinated oxygen sites, and the peak areas in the spectra do record the true sites populations. This is the expected result, given that oxygen diffusion into solid oxides most likely occurs by random exchanges between adjacent oxygens, and therefore is not coordination-number-specific.

The average oxygen coordination, estimated from the ratio between two- and three-coordinated oxygen, is ~ 2.4 (± 0.1) in the amorphous material, while that of the crystalline tantalum is ~ 2.6 (± 0.1). This probably also corresponds to a somewhat lower mean Ta coordination number by oxygen in the amorphous phase, which leads to a lower overall density, and also somewhat shorter mean Ta–O distances, as is commonly seen in comparisons of low- and high-pressure polymorphs of oxides, e.g., the quartz (low cation and anion coordination numbers, shorter Si–O distances, lower density) versus stishovite (higher cation and anion coordinations, longer distances, higher density) phases of silica. This result is consistent with previous conclusions from XRD measurements of the averaged radial distribution function.⁷ The crystal is known to contain both TaO_6 and TaO_7 groups,¹⁶ so our results may also imply a lower proportion of the latter in the amorphous phase.

An amorphous tantalum thin film, produced by ion-beam sputtering and isotopically enriched after the deposition by exposure to 70% $^{17}\text{O}_2$ gas at 450 °C (see the Experimental Section), also yielded a high-quality NMR spectrum (Figure 2), indicating a substantial degree of oxygen isotope exchange under these experimental conditions ($\sim 5\%$ – 10%). The spectrum again shows two resonances at 310 and 470 ppm with a relative intensity ratio of $\sim 2:3$ and an average oxygen coordination number close to 2.4. There are no significant differences in the relative intensity ratio, the resonance positions, or the line shapes from the samples made using sol–gel synthesis. Therefore, at least after annealing at 450–500 °C, the short-range structures of samples of amorphous tantalum made by sol–gel synthesis and by ion-beam sputtering must be very similar, even though there are many

factors that can affect the NMR resonance positions and line shapes (for example, the extent of disorder and corresponding variations in distributions of bond angles and distances). Therefore, studies using sol–gel samples may provide useful preliminary structural insights prior to the more costly synthesis of ion-beam sputtered coatings.

Titania-Doped Tantalum. As shown in Figure 3, ^{17}O NMR spectra were collected for titania-doped amorphous tantalum, $(\text{Ta}_{1-x}\text{Ti}_x)_2\text{O}_{5-x}$ ($x = 0.10, 0.25, \text{ and } 0.50$). As many as four different, partially overlapping resonances are observed. The spectra at 9.4 T were used for the analysis, which are better resolved from spinning sidebands than in spectra at 14.1 T, since the quadrupolar interaction is small enough not to broaden the line at the lower field and the separation (in ppm) between sidebands is larger at a lower field for a fixed spinning frequency. The two main resonances at 310 and 470 ppm can again be assigned to oxygen with only tantalum first-neighbors, three-coordinated and two-coordinated, respectively. In addition, at least two more broad resonances are observed for the titania-doped amorphous tantalums (at 380 and 560 ppm). To begin to assign these, we note that the ^{17}O resonances for crystalline TiO_2 , in which all oxygens are three-coordinated by Ti, are shifted to considerably higher frequency (557 and 588 ppm in rutile, 560 ppm in anatase¹⁹) from the comparable peak in tantalum (310 ppm). Thus, it makes sense that, as TiO_2 is added to Ta_2O_5 , a resonance for an oxygen with one Ti and two Ta neighbors (OTa2Ti) should first appear at a frequency above 310 ppm, leading to the assignment of the 380 ppm resonance to this local structure. If two-coordinated oxygens with one Ta and one Ti neighbor also form, their resonance would be expected at a frequency above the 470 ppm peak in pure tantalum and might contribute to the broad feature that appears at 560 ppm in the $x = 0.5$ sample. However, if the titania and tantalum are not completely mixed in this composition, a resonance for anatase-like domains (oxygen with three Ti neighbors) could account for much or all of this peak, as has been reported in ^{17}O MAS NMR spectra of partially phase-separated SiO_2 – TiO_2 sol–gel products.²⁰

When the $x = 0.5$ titania-doped tantalum was heated at 700 °C, the sample crystallized completely and phase-separated to TiO_2 and Ta_2O_5 . The ^{17}O NMR shows separate sets of resonances from TiO_2 (anatase, 560 ppm) and Ta_2O_5 (320 and 460 ppm). The anatase peak is narrow and has the same frequency as that reported for the pure phase,¹⁹ suggesting that it contains little dissolved Ta; the lack of extra resonances in the crystalline tantalum for oxygens with Ti neighbors also suggests that there is little solution of Ti in this material.

From pure tantalum to the $x = 0.1$ and 0.25 titania-doped samples, the resonance at 380 ppm grows as expected if it is related to an oxygen with two Ta and one Ti neighbors. The lack (roughly $<5\%$ of the total oxygens) of a peak at 560 ppm in these samples also indicates that anatase-like domains are not abundant, and, thus, that the Ti and Ta oxides are mixed at the atomic scale. However, the lack of an obvious feature corresponding to an oxygen with one Ti and one Ta neighbor (which might also occur near to 560 ppm) suggests that the Ti–Ta mixing is not random, and that Ti cations are preferentially associated with three-coordinated oxygens (380 ppm resonance).

Also, in the $x = 0.1$ and 0.25 Ti-doped tantalums, the relative intensity of the OTa3 resonance relative to that for the OTa2 decreases somewhat, again as expected if Ti is preferentially sited next to three-coordinated oxygens. However, both peaks are smaller percentages of the total oxygen than in the pure tantalum,

Table 1. Relative Intensities of Resonances in Ti-Doped Amorphous Tantalum, $(\text{Ta}_{1-x}\text{Ti}_x)_2\text{O}_{5-x}$

x	Relative Intensity of Resonance						average coordination number
	OTa2	OTaTi or OTi3	total 2-coordinated	OTa3	OTa2Ti	total 3-coordinated	
0	57		57	43		43	2.43
0.1	53	<1	53	32	15	47	2.47
0.25	47	<5	47	27	26	53	2.53
0.50	46	17		13	25		

To take the contribution of satellite transition to the center peak intensity, 20% and 10% intensities (based on the first sideband intensity) were subtracted from two- and three-coordinated oxygen intensities. The uncertainties are $\sim 5\%$, because of the overlap between resonances and uncertainty of the contribution of satellite transition to the intensity.

suggesting that some OTa2 is also converted to OTa2Ti. In these samples, the fitted peak areas (Table 1) show that the total fraction of three-coordinated oxygen (OTa3 + OTa2Ti) increases with titania content, leading to a net increase in the overall average oxygen coordination. This observation is sensible from simple crystal chemical considerations: an oxide ion coordinated by one Ti^{4+} (likely to be in six-coordination) and one Ta^{5+} (in six- or seven-coordination) would be highly “underbonded.” The increase in mean oxygen coordination on addition of titania to tantalum means that, in some sense, the structure has a greater degree of overall connectivity, which, in turn, may have important implications for the mechanical properties.

In the $x = 0.5$ titania sample, the OTa3 peak appears to be reduced in relative area more dramatically as the OTa2Ti peak grows even more, although there is an additional complicating possibility of a new resonance for OTaTi2 underneath the OTa2 peak (at ~ 470 ppm). The prominent new peak at 560 ppm could represent the development of OTiTa sites, or it more likely shows the presence of anatase-like domains (OTi3) as the limit to solid solution of titania and tantalum is exceeded. Because of these ambiguities in peak assignments, we cannot accurately estimate further changes in overall oxygen coordination above $x = 0.25$.

CONCLUSIONS

Amorphous pure and titania-doped tantalum can be prepared via sol–gel methods and enriched with ^{17}O by gas exchange at a temperature low enough to retain the amorphous structure, as can thin-film pure tantalum initially synthesized by ion-beam sputtering as used to make optical coatings. ^{17}O MAS NMR proves to be very sensitive to the coordination environments of the oxide ions in tantalum (i.e., coordination number and type of cation neighbors) and is thus useful in investigating the local structure. The average oxygen coordination number was estimated to be 2.4 in both amorphous sol–gel and thin-film tantalum, while that for crystalline tantalum was 2.6, the latter in good agreement with crystal structure data.

The increase in the average coordination number also implies an increase in cation coordination number and may be related to an increase in density upon crystallization. ^{17}O MAS NMR spectra of titania-doped tantalum show additional resonances due to oxide ions with mixed Ti and Ta neighbors, and they suggest increases in the mean oxygen coordination number with titania content. This may affect mechanical properties and could possibly be related to observed improvements in mechanical loss properties, which is a key issue for high-performance interferometer mirrors.

The results presented here provide some of the first clues regarding the short-range structure of amorphous tantalum, and they point the way for future applications of ^{17}O NMR to a better

understanding of structure–property relationships. Future studies may include, for example, the preparation of IBS films of more-complex compositions for comparison with sol–gel materials, and the measurement of mechanical properties of the same films as produced for spectroscopic studies to more finely optimize mirror performance in optical systems such as LIGO.

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