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N.B.: When citing this work, cite the original article.

Original Publication:
http://dx.doi.org/10.1063/1.4921167
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Postprint available at: Linköping University Electronic Press
http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-119249
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Citation: Journal of Applied Physics 117, 195301 (2015); doi: 10.1063/1.4921167
View online: http://dx.doi.org/10.1063/1.4921167
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/117/19?ver=pdfcov
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Growth and oxidation stability of cubic \( \text{Zr}_{1-x}\text{Gd}_x\text{N} \) solid solution thin films

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(Received 27 March 2015; accepted 28 April 2015; published online 15 May 2015)

We report \( \text{Zr}_{1-x}\text{Gd}_x\text{N} \) thin films deposited by magnetron sputter deposition. We show a solid solubility of the highly neutron absorbing \( \text{GdN} \) into \( \text{ZrN} \) along the whole compositional range, which is in excellent agreement with our recent predictions by first-principles calculations. An oxidation study in air shows that \( \text{Zr}_{1-x}\text{Gd}_x\text{N} \) with \( x \) reaching from 1 to close to 0 fully oxidizes, but that the oxidation is slowed down by an increased amount of \( \text{ZrN} \) or stopped by applying a capping layer of \( \text{ZrN} \). The crystalline quality of \( \text{Zr}_{0.5}\text{Gd}_{0.5}\text{N} \) films increases with substrate temperatures increasing from 100°C to 900°C. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4921167]

INTRODUCTION

Gadolinium compounds and their alloys are of interest due to the high thermal neutron absorption cross-section of \( \text{Gd} \) and due to its magnetic properties. Among all naturally occurring elements, \( \text{Gd} \) has the highest thermal neutron absorption cross-section of 49 700 b. One of its stable isotopes, \( ^{155}\text{Gd} \), with a natural abundance of 15.65% has an absorption cross-section as high as 259 000 b and is commercially available. \( \text{Gd} \) is ferromagnetic at temperatures below 289 K, which makes it a common element in ferromagnetic superconductors. At higher temperature it is strongly paramagnetic, resulting in \( \text{Gd(III)} \) chelates being the leading contrast agents in magnetic resonance imaging.1

The high probability of \( \text{Gd} \) to absorb thermal neutrons enables new types of neutron detectors that could raise detector resolution and detection efficiency to a completely new level. A second application is found within thermal neutron shielding, where the nowadays very large and heavy materials could be replaced by micro-engineered and miniaturized solutions.2 New large-scale neutron scattering facilities like the European Spallation Source (ESS)3 and target station 2 at the Spallation Neutron Source (SNS)4 are striving to push neutron detectors, which can be an alternative to MSGCs.33 Both technologies have the potential to be used in an instrument like NMX, but require long-term stable thin films containing high amounts of \( \text{Gd} \). \( \text{GdN} \), which has a higher \( \text{Gd} \) content per unit volume than \( \text{Gd}_2\text{O}_3 \), is used.34 Such a detector would fulfill the efficiency and resolution requirements of 20% and <0.2 mm, respectively, and allow for parallax corrections in the varying sample-detector distance setup in the Neutron Macromolecular Crystallography (NMX) instrument that will be built at the ESS.3 Initial studies on the potential of solid neutron converters in combination with micro pattern gaseous detectors have been reported.30 Schulz and co-workers developed micro-strip gas chambers (MSGC)31 that contain neutron converting layers of \( \text{Gd} \) and in parallel Sauli developed gas electron multiplication (GEM) neutron detectors, which can be an alternative to MSGCs.33

The reduction of experimental backgrounds for neutron spallation sources is a very topical issue and instrument performance is typically defined by signal-to-background ratios. While the signal is defined by the intensity of the source, the background can be improved with more advanced instrument designs, resulting in improved performances. Additionally, large volumes of radiation shielding are required and, therefore, significant cost savings are expected for advances in the search for new effective materials.2,36

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Whilst the work this far has concentrated on reducing fast neutron and gamma ray background, there is also an effort needed in reducing the background from thermal neutrons preventing “cross-talk” between local neutron detector elements. Using Gd compounds, neutron shielding can be engineered to be both compact and very efficient. At the moment, this is typically solved by using Gd in the form of Gd$_2$O$_3$ that is mixed with an epoxy and painted onto surfaces to be shielded, but epoxy is not ideal due to outgassing and does not allow for something mechanically precise. Therefore, stable Gd-rich compounds that are precisely deposited as thin films present a potential application niche.

During the past ten years, it has been shown that rocksalt-structured GdN thin films can be grown both with physical vapor deposition techniques like reactive sputtering, reactive thermal evaporation, and molecular beam epitaxy but also with metal-organic chemical vapor deposition processes. The intention has mainly been to deposit high quality films for investigations of their optical and magnetic properties. The foremost issue related to growth has been the high tendency to oxidization, which (when needed) has been solved with buffer and/or capping layers. GdN single crystals have been shown to have a metallic electrical conductivity. For use in a neutron detector, we are seeking a solution for a stable enough compound that is electrically conducting and contains high amounts of Gd. The stable oxide Gd$_2$O$_3$, therefore, has the drawback that it is an insulator, which hinders the transport of built up charges in a detector, and that the atomic concentration of Gd is only 40% of the compound, which lowers the neutron absorption efficiency significantly.

Recently, the authors presented a first-principles study on the mixing thermodynamics of GdN with the transition metal nitrides TiN, ZrN, and HfN. These binary compounds were chosen because they are thoroughly studied, chemically quite inert, thermally stable, and electrically conducting. They are also known to be good oxidation barriers and are industrially used as such. To alloy GdN with one of these compounds could result in a solid solution that is straightforward to deposit as a thin film, that is both conducting and oxidation resistant, and contains a high amount of Gd.

In this experimental study, we have chosen ZrN as the alloying compound to explore the solid solubility of ZrN and GdN and the resulting thin film properties. ZrN was preferred over TiN because the mixing enthalpy calculations in Ref. stipulate phase separation for the latter, while HfN was disregarded because it is more exotic, less explored, and considerably more expensive than ZrN. We have deposited solid solution films over the full compositional range of Zr$_{1-x}$Gd$_x$N from ZrN to GdN, thus verifying theoretical predictions of the mixing tendency of the alloy. Results for oxidation resistance and crystalline quality are also presented.

**EXPERIMENTAL PROCEDURES**

**Thin film deposition**

Deposition experiments were performed in an ultrahigh vacuum chamber at a base pressure of $4 \times 10^{-6}$ Pa. Reactive magnetron sputter deposition using unbalanced type II magnetrons with 75 mm diameter Zr and Gd elemental targets was used to grow Zr$_{1-x}$Gd$_x$N films, with $x$ ranging from 0 to 1, onto polished Al$_2$O$_3$(0001) and Si(001) substrates. The Ar and N$_2$ partial pressures were set to 0.53 and 0.13 Pa, respectively. As references for samples included in the oxidization study and as diffusion barriers at high deposition temperatures, seed and/or capping layers of ZrN(111) or ZrN(001) were deposited additionally. The deposition system is described in detail elsewhere.

The ZrN seed and capping layers were chosen because they are known to be temperature stable, efficient diffusion barriers, and serve as lattice-matched templates for epitaxial film growth, especially for the ones with the lowest GdN contents. Al$_2$O$_3$(0001) substrates were chosen as the base substrates due to their temperature stability and to avoid overlap of film peaks with substrate peaks in X-ray diffraction (XRD).

Prior to deposition, the substrates were cleaned in ultrasonic baths of trichloroethylene, acetone, and 2-propanol and blown dry in dry N$_2$. The substrate heater was slowly ramped up to the chosen deposition temperature, which was controlled by a thermocouple positioned behind the substrate and calibrated by pyrometry. For seed and capping layers, the same substrate temperatures were used as during the film depositions.

The Zr and Gd magnetron powers were set to a total power of 300 W, with relative adjustments of the powers to obtain various compositions, $x$ between 0 and 1, in Zr$_{1-x}$Gd$_x$N. Rutherford Backscattering Spectrometry (RBS) results show that for a molar fraction of $x=0.5$, the Zr and Gd magnetron powers need to be 200 W and 100 W, respectively.

**Thin film analysis techniques**

Compositional analysis was mainly performed with RBS using a 2.0 MeV He$^+$ beam at 6° incidence and 172° scattering angle. The advantage with RBS for Zr$_{1-x}$Gd$_x$N thin films is the possibility to obtain very accurate compositional ratios between the metal atoms. The sensitivity for contaminants such as H (He$^+$ can not be backscattered on H), C, and O is low. However, C and N can be distinguished if the films contain several atomic percent of each kind. For this study, RBS was used to determine the ratio between Zr and Gd, meaning $x$. It was also a key technique to judge whether the film was oxidized or not, since that can be extracted from the data both by looking at change in the areal density of the film when oxidized and/or by determining the amount of O relative to N.

As a complementary technique to determine the composition of the light elements in the Zr$_{1-x}$Gd$_x$N thin films, we have used time-of-flight Elastic Recoil Detection Analysis (ToF-ERDA). This compositional analysis technique was performed using a 31.5 MeV $^{127}$I$^{8+}$ beam at 66° incidence and 45° recoil scattering angle. The recoil energy of each element was converted to relative elemental depth profiles using the CONTES code. As the sensitivity is good for light elements, we have used ToF-ERDA to quantify the amounts of N and impurities like Ar (from sputter gas), O, C, and H in the film.
The Gd isotope distribution in the films was measured by time-of-flight Secondary Ion Mass Spectrometry (ToF-SIMS) using a TOF.SIMS V instrument (ION-TOF GmbH, Germany). A dual-beam depth profiling procedure was applied with a 1.0 keV O$_2^+$ sputter beam, having a current of 300 nA and scanned over $400 \times 400 \text{m}^2$. A pulsed 30 keV Bi$^+$ beam was used as the analysis beam. The current was typically 1.5 pA with an analysis field of view of 100 $\times$ 100 $\mu\text{m}^2$ at the centre of the sputter crater. Positive ToF-SIMS spectra were acquired between the sputter cycles in the so-called spectroscopy mode (mass resolution $m/\Delta m \sim 8000$, beam spot $\sim 5 \mu\text{m}$).

The crystal structure was characterized by Cu Kz XRD using a Philips Bragg-Brentano diffractometer. The film thickness was measured with cross-sectional scanning electron microscopy (SEM) for films with high ZrN content using a LEO 1550 instrument, equipped with an in-lens detector operated at 5 kV at a working distance of $\sim 3 \text{mm}$.

RESULTS AND DISCUSSION

The solid solubility of GdN into ZrN was explored by depositing a series of Zr$_{1-x}$Gd$_x$N films with $0 \leq x \leq 1$ at a substrate temperature of 700 °C. The films were sandwiched between seed and capping layers of ZrN to avoid any reactions with the substrate or oxidation due to exposure to air before the characterization was done. It has been shown previously for the chemically similar Sc$_{1-x}$Al$_x$N system that a comparison between calculated lattice spacings over the whole composition range with experimental measurements is a reliable tool to reveal secondary phase formation during film growth. Applying this comparison to the present system, Figure 1 shows the calculated lattice parameters for the cubic disordered solid solutions of Zr$_{1-x}$Gd$_x$N from Ref. 50 together with the experimental curve, which is a combination of the relations between the Zr and Gd metals obtained from RBS and the lattice parameters calculated from XRD data. The measured lattice parameters follow the same increasing trend with increasing GdN content as the calculations. The calculated lattice parameters slightly overestimate our measured values, which is the usual condition found for calculations employing the generalized gradient approximation for exchange-correlation effects in nitrides. However, this overestimation vanishes for the GdN rich compositions. In Ref. 50, it was shown that the theoretical overestimation for pure GdN with respect to experiments is smaller compared to the case of pure ZrN. One should also keep in mind that the calculations were done for the ideal nitrogen stoichiometry while a slight understoichiometry is found in our measurements (see below). The effect of such understoichiometry on the lattice spacing can be different in different nitrides, possibly adding to the slight differences in slope between theory and experimental curves in Figure 1. However, the gradual increase in lattice spacing with composition almost follows Vegard’s rule and the theoretical predictions and therefore it is a strong evidence of the formation of solid solutions over the full compositional range.

To determine the most favorable substrate temperature for high quality epitaxial Zr$_{1-x}$Gd$_x$N films, Zr$_{0.5}$Gd$_{0.5}$N was deposited with a ZrN seed and capping layer onto Al$_2$O$_3$(0001) at substrate temperatures between 100 and 900 °C. As seen in the different XRD scans in Figure 2, it is mainly the orientation of the ZrN that determines the orientation of the Zr$_{0.5}$Gd$_{0.5}$N. The preferred growth orientation for the highest (900 °C) and lowest (100 °C) applied temperatures is (111), while temperatures between 300 and 700 °C allow for a mixture of (111) and (001) oriented growth. No other growth orientations were seen in longer overview scans. While the hexagonal symmetry of the Al$_2$O$_3$(0001) substrate surface will favor (111) oriented nucleation at high temperatures, the in-plane lattice mismatch of Al$_2$O$_3$(0001) compared to ZrN(111) is as large as 19.4%, which reduces the possibility of forming continuous epitaxial layers, in particular, at limited growth temperatures. On the other hand, Petrov and co-workers have previously shown that increased adatom mobility, as induced by increased substrate temperature, generally promotes a (001) preferred growth orientation. Thus, the mixture of ZrN (111) and (001) oriented growth at intermediate growth temperatures may be explained by a competition between the driving forces for (111) oriented epitaxy and for (001) oriented growth due to

![FIG. 1. Measured (this work) and calculated (Ref. 50) lattice parameters for the cubic rocksalt solid solution of Zr$_{1-x}$Gd$_x$N with $0 \leq x \leq 1$.](image)

![FIG. 2. XRD data from (111) and (001) oriented Zr$_{0.5}$Gd$_{0.5}$N films on Al$_2$O$_3$(0001) and (111) and (001) oriented ZrN seed and capping layers grown at substrate temperatures between 100 and 900 °C.](image)
increased adatom mobility. At the highest temperature though, the (111) oriented preferred growth indicates that the interaction with the substrate becomes the determining factor. An additional observation is the shift of the Zr$_{0.5}$Gd$_{0.5}$N 111 peak towards higher angles, meaning smaller out-of-plane lattice parameter, with increasing temperature. This behavior is consistent with an increasing influence by the substrate surface lattice as the temperature is increases, as evidenced by the trend towards (111) oriented growth. Also a nitrogen under-stoichiometry, as is often observed for reactively grown transition metal nitrides, may have a similar influence on the lattice parameter evolution. While it is beyond the scope of this work to clarify the reason for this temperature effect, we note that the coefficients of thermal expansion (CTE) of most transition metal nitrides (e.g., CTE$_{ZrN}$ = 7–9 $\times 10^{-6}$ K$^{-1}$) and sapphire perpendicular to the C-plane (CTE$_{Al_{2}O_{3}}$ = 7–10 $\times 10^{-6}$ K$^{-1}$) are very similar and cannot explain the large observed peak shifts.

The XRD data for the intermediate growth temperature 500 °C, which exhibits the highest 002 peak intensity, exhibits an inconsistency in the Zr$_{0.5}$Gd$_{0.5}$N 002 peak shift direction. We find this interesting, as it is an indication of a material with anisotropic mechanical and/or structural properties, as might be for the case in Ref. 50 predicted ordered structure of ZrGdN$_2$. Nevertheless, the formation of that phase could not be found in any of the measured XRD scans or X-ray pole figures (not shown), even though the compositional ratios were correct and the in-plane lattice mismatch was expected to be only 3.9% according to the calculated lattice parameters in Ref. 50. Instead, the substitutionally disordered Zr$_{0.5}$Gd$_{0.5}$N solid solution was observed for all applied substrate temperatures. It was noted in Ref. 50 that the predicted critical ordering temperature, $T_{c} = 1020$ K (747 °C), is in the same range as where bulk diffusion becomes extremely slow in nitrides. Since several of our samples were grown at lower temperatures, the absence of an ordered phase illustrates that surface diffusion alone, which is active at all the considered temperatures, is insufficient to create a long-range metal-site ordering in this nitride system.

ToF-ERDA depth profiles were recorded for films that, according to RBS, contain x = 0.30 in Zr$_{1-x}$Gd$_{x}$N, both with and without ZrN capping layers. Both films show compositional levels of H, C, F, and Ar below the ToF-ERDA detection limit of 0.03 at.%. The level of O is also close to the detection limit in the film that was covered with a capping layer, while the film without a capping layer shows higher O levels close to the surface. This indicates that the films do not contain any O before they are exposed to air and start to oxidize. It also shows that the sputtering targets are pure and the vacuum conditions good enough to provide films without contaminations. We further conclude that no Ar has been incorporated during the sputter deposition. The N content in the as-deposited films has been determined by combining ToF-ERDA and RBS results and is found to be in the region of 45–50 at.%

A ToF-SIMS depth profile of a typical GdN film sandwiched between seed and capping layer of ZrN was recorded and an important result for future neutron detector applications is that the natural abundance of the five most common isotopes in Gd, $^{155}$Gd = 14.8%, $^{156}$Gd = 20.6%, $^{157}$Gd = 15.7%, $^{158}$Gd = 24.8%, and $^{160}$Gd = 21.8% could be confirmed.

To follow the oxidation of pure GdN without capping layer, ~400 nm of GdN was deposited onto an Al$_2$O$_3$ substrate and then exposed to air at room temperature. The oxidation was followed by the surface color change, recorded every 4 s with a digital camera. The continuous oxidation process is illustrated by a selection of these images of GdN films on transparent Al$_2$O$_3$, exposed to air up to 19 h as shown in Figure 3. The as-deposited sample had a silver-metallic color that within the first minutes changed to be more golden and then brownish. The rapid color change indicates that the oxidation starts immediately. After the brown appearance and within the first hour, all the colors of the rainbow brightly appeared after each other. During the following few hours, the film appeared to be purple and green with some interference color gradients indicating a slight thickness gradient of the oxide and remaining film. After less than 19 h, the film has become fully transparent and is completely oxidized. RBS measurements on a completely oxidized film show that the majority of N has been replaced by O. A similar but much faster color change, probably due to a GdN film thicknesses of only ~100 nm, has been reported in Ref. 41.

To investigate the oxidization stability of Zr$_{1-x}$Gd$_{x}$N, a series of films with 0 ≤ x ≤ 1 were deposited with and without ZrN capping layers onto Si and Al$_2$O$_3$ substrates at 400 °C. These films were measured with RBS within two weeks after they were deposited. A second measurement was done 3–4 months after deposition, for those films that were not fully oxidized already in the first measurement. All samples were stored in room temperature and in air. The results are presented in Figure 4 and show that films with a high amount of GdN oxidize within a very short time. Increasing the amount of ZrN significantly slows down the oxidization and films with x ≤ 0.3 were just slightly oxidized even after several months. The idea to alloy GdN with a chemically more stable transition metal nitride with the purpose to prevent it from oxidization is thus working, although oxidation can not fully be avoided even with GdN contents as low as x = 0.2.

![FIG. 3. Color change for a GdN thin film on Al$_2$O$_3$ substrate during exposure to air for up to 19 h.](image-url)
Figure 4 shows typical RBS spectra of $\text{Zr}_{1-x}\text{Gd}_x\text{N}$, recorded from the same film, both as-deposited and after 4 months. Here, we show $\text{Zr}_0.5\text{Gd}_0.5\text{N}$ on Si. Comparing the Zr and/or Gd spectra, respectively, clearly shows the increase in the areal density when O replaces N during oxidization. The complete transition is also seen in that the as-deposited film contains no oxygen (within the detection limit) and the oxidized film contains no nitrogen. The ToF-ERDA depth profile of the as-deposited film, mentioned above, confirmed that those films were free from oxygen. Even though the nitrogen completely escapes from the film and is replaced by oxygen, the ratios between the Gd and Zr content stay constant for all measured samples before and after oxidization, respectively.

For films that were oxidized half way through in the oxidization study in Figure 4, an abrupt interface between the oxidized layer on top and the not oxidized layer below can be identified in the analyses of the RBS-spectra and their corresponding simulations. We therefore conclude that within the $\sim 2 \text{ mm} \times 2 \text{ mm}$ RBS beam spot, the thickness of the formed oxide is the same within the measurement errors. We find it an interesting observation that none of all the samples in the oxidization study showed a gradient at the interface between the oxidized layer and the intact nitride layer, but always a sharp transition. This is in agreement with the observations from the surface color change in Figure 3, where a uniform interference color over the whole film surface, $\sim 8 \text{ mm} \times 10 \text{ mm}$, within the first 30 min corresponds to a uniformly thick oxide layer in the film.

For several applications, including highly efficient neutron detectors, it is possibly more beneficial to protect a film that contains a high amount of Gd with a thin (preferably conducting) oxidization barrier, than to use a Gd compound that degrades over time. We therefore tested a 260 nm $\text{Zr}_0.5\text{Gd}_0.5\text{N}$ film deposited with both 35 nm ZrN capping and seed layers. The film was measured with RBS soon after it was deposited and again after storing it in air and at room temperature for one year. Within the measurement accuracy, the compositions of the $\text{Zr}_0.5\text{Gd}_0.5\text{N}$ film and ZrN protective layers were not changed during that time. This experiment shows that applying a capping and seed layer prevents the film from oxidizing when it is exposed to air and promises a long lifetime of the nitride structure, even with higher amounts of GdN. The minimum required thickness of the protective layers remains to be determined for the different compositions. The final decision, whether to use capping layers or films with lower Gd-content for neutron detector applications depends on the detector requirements and if the films can be kept in oxygen free atmosphere.

It was also observed by RBS analysis that the interface between a $\text{Zr}_{0.55}\text{Gd}_{0.45}\text{N}$ film and a Si substrate after one year still exhibited an abrupt transition, when only a ZrN capping layer but no seed layer was used. This shows that no significant amount of substrate elements have diffused into the film, even though there is a native oxide on the Si substrate, but rather that gaseous oxygen is responsible for the oxidation. The same observation was made for all measurements included in Figure 4 that showed partly oxidized films, where the oxygen only was found in the top layers.

CONCLUSIONS

We have shown that (001) and (111)-oriented solid solution $\text{Zr}_{1-x}\text{Gd}_x\text{N}$ alloy thin films can be grown by magnetron sputter deposition over the whole compositional range from ZrN to GdN, in excellent agreement with theoretical predictions. The alloys were substitutionally disordered and no ordered alloys could be observed for deposition temperatures up to 900°C. The oxidization of the $\text{Zr}_{1-x}\text{Gd}_x\text{N}$ films is substantially reduced when increasing the amount of ZrN and can even be arrested by applying a capping layer of ZrN. Even though this study is an initial experimental work on Gd containing compounds with applications within neutron detection and shielding in mind, it adds a significant amount of information to the judgment about the potential of these materials for those applications.

ACKNOWLEDGMENTS

The authors would like to acknowledge the Tandem Laboratory at Uppsala University for giving access to their ion beam facilities, and Dr. Muhammad Junaid for his laboratory assistance. B.A. acknowledges the financial support from the Swedish Research Council (VR) and the Knut and Alice Wallenberg Foundation.
support from the Swedish Research Council (VR) through Grant Nos. 621-2011-4417 and 330-2014-6336.


