Ultra-rapid, physics-based development pathway for reactor-relevant RF antenna materials

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***Abstract*—This paper presents a rapid, atomistically-informed, experimental development pathway for fusion reactor-relevant radio frequency (RF) antenna materials in the Cu-Cr-(Nb,Al,Zr) composition system, with the goal of improving upon GRCop-84. RF antennas in a tokamak fusion reactor will face a unique set of challenges as both structural and functional materials. The desired material must simultaneously achieve and maintain high electrical conductivity, high strength, high thermal conductivity, resist high temperatures, possess low nuclear activation, and incur low damage due to neutron bombardment. The GRCop-84 alloy serves as a starting point for iterative improvement, with the desire to reduce or eliminate Nb from the material to minimize nuclear activation.**

The rapid development pathway makes use of a multi-target combinatorial thick film sputtering process to produce full ternary phase diagrams on a Si wafer substrate. Transient grating spectroscopy (TGS), a laser-ultrasonic method, will determine spatially-varying thermo-elastic properties, while four terminal electrical conductivity measurements will map out the best performing regions of the sample for in-depth study at larger length scales. High energy proton and self-ion irradiation emulates the effects of neutron damage on the thermal/electric properties. With rapid turnaround time (∼days) in terms of mapping radiation damage-induced material property changes in the full ternary system, these techniques allow rapid iteration towards an optimal material, testing hundreds of nearby compositions in the time it took to test one. Focused testing of larger, single composition samples (produced in an arc furnace or by laser sintering) provides data on structural and high power RF properties, and validates our thick-film based workflow.

***Index Terms*—Fusion reactor materials, microwave, radio frequency, transient grating spectroscopy, sputtering.**

I. Introduction

FUSION reactors present a unique set of challenges for functional materials. Components close to the fusion plasma are subject to inescapable, severe neutron and heat fluxes. Plasma disruptions subject plasma-facing components to large $J \times B$ forces. For radio frequency (RF) antennas there are additional constraints related to the effective operation of the antenna: high electrical conductivity to reduce resistive power losses, and low susceptibility of arcing at high power. Furthermore, these components must endure the harsh environment for months or years at a time without maintenance, and then be disposed of as low-level nuclear waste and/or recycled into new fusion reactor components.

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A recent study [1] found that no currently available material simultaneously meets all criteria of high electrical conductivity ($> 6 \times 10^7$ S/m), high thermal conductivity (> 300 W/m·K), high strength at elevated temperature (> 150 MPa at 700°C), and low activation in a 14 MeV neutron environment (waste disposal rating (WDR) < 1 after 1.7 full-power-years (FPY) in the FESS-FNSF [2]). Table I summarizes the desired material properties for comparison with available materials. The GRCop-84 alloy [3] (Cu-8 at.%Cr-4 at.%Nb) is a promising candidate material which meets nearly all of the requirements [4]–[6] and can be used to additively manufacture complex geometries with high resolution [7]. However the 4-at.% Nb content presents a challenge for hazardous nuclear waste generation when used in components that face the full 14 MeV neutron flux (WDR $= 2.4 \times 10^3$ after 1.7 FPY). The stable ^{93}Nb isotope undergoes neutron capture to become radioactive ^{94}Nb (half life ∼20,000 y), which then β^- decays to stable ^{94}Mo. The end goal of this project is to create a new alloy that best retains the desirable properties of GRCop-84 by replacing Nb with other elements that do not transmutate into high level waste after neutron bombardment.

The remainder of this paper details a new development pathway for reactor-relevant RF antenna materials. The rapid development pathway utilizes a physical vapor deposition (PVD) chamber to create novel alloys with composition varying in two dimensions on a substrate wafer, followed by ion-beam irradiation to simulate neutron damage. The resulting thick combinatorial films can then be tested for retention of thermal and electrical properties using transient grating spectroscopy (TGS) and four-point resistance measurements following irradiation, to identify regions of interest in composition space for closer examination. Our central hypothesis, that "islands of stability" are waiting to be uncovered in this five-dimensional compositional design space, rests upon a faster materials discovery workflow to enable rapid compositional searching and testing. Such "islands of stability" have been seen in many other combinatorial materials studies, such as searching for stable ternary metal nitrides [8]. This process enables evaluation of thousands of potential alloy compositions at one time, rather than the time-intensive process of creating and testing individual alloy samples. After an optimized alloy composition is determined, macroscopic samples undergo testing of structural and RF properties.

II. Thick film sputtering

Combinatorial synthesis [9] creates a wide spectrum of potential alloy compositions in an automated procedure. An

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Material	Thermal Conductivity [W/m/K]	Electrical Conductivity [S/m]	Waste Disposal Rating	Temperature Window [°C]
Specification	> 300	$> 3 \times 10^7$	< 1	< 700
F82H	30	2×10^6	0.05	< 550
CuCrZr	320	3×10^7	0.36	< 500
Glidcop-AL15	365	5×10^7	1.30	< 500
GRCop-84	300	6×10^7	2,400	< 700
Tungsten	175	2×10^7	0.63	700-900

TABLE I

RELEVANT MATERIAL PROPERTIES OF SELECTED METALS FOR THE FNSF LHRF ANTENNA STRUCTURE [1]. THERMAL AND ELECTRICAL CONDUCTIVITY VALUES ARE REPRESENTATIVE VALUES FOR UNIRRADIATED MATERIALS. WASTE DISPOSAL RATING (WDR) IS BASED ON 1.7 FPY OF OPERATION ON THE HIGH FIELD SIDE WALL OF FNSF [2]. TEMPERATURE WINDOW INDICATES OPERATING TEMPERATURE WHERE ULTIMATE TENSILE STRENGTH EXCEEDS 150 MPA WHILE REMAINING DUCTILE.

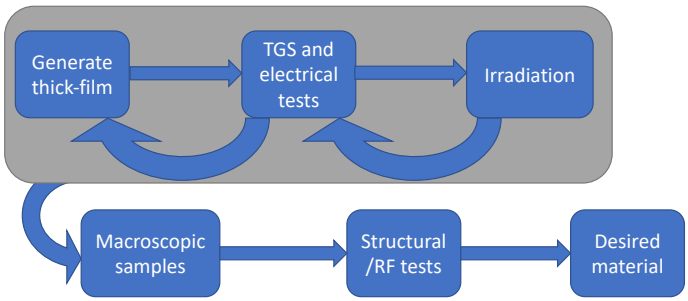

Fig. 1. Flowchart of iterative materials creation process. The grey region represents the novel rapid iteration workflow. Subsequent analysis of a reduced number of macroscopic samples occurs after promising alloy compositions are determined through the rapid iteration workflow.

upgraded five-target PVD chamber (Fig. 2) deposits atoms from each target onto the substrate wafer in a manner that varies with distance from the target, size of the target, and power applied to the target. Figure 3 shows the sputtering target layout relative to the central wafer. The spatial variation in material laid down by each sputtering target creates a sample with compositional gradients in two dimensions across the wafer surface. Adjusting the angle of each target with respect to the wafer changes the gradient for each component of the alloy. Three-component alloy compositions using three-inch, two-inch, and one-inch sputtering targets arranged at 120° angles (sources 1, 2, and 5 in Fig. 3) naturally have a dominant, secondary, and tertiary component based on the size of each sputtering target, with the gradient of each component aligned at 120° angles to the other two components. This creates a "color wheel" pattern of alloy composition on the wafer (Fig. 4, left), which is then automatically measured using TGS (center) to create maps of thermoelastic properties tightly correlated to radiation damage resistance [10]. The values of these critical properties, and their relative changes before vs. after irradiation, constitute the fitness function by which we deem a particular material to be better than others. It should be noted that sputtering targets may be made from multiple principal elements themselves, for example, a fixed amount of (Cu, Cr, Nb) with varying amounts of Al and Zr as the other two targets.

Fig. 2. Interior view of the 5-target PVD chamber. The new fifth sputtering target arm in the rear-center of the image has a clean, mirrored stainless steel finish. The wafer upon which the thick films grow sits at the top of the image. The protective shutter of one sputtering target is retracted on the left side of the image.

For the first iteration of the process, a wide variation of composition is used to identify promising targets for additional, more detailed examination. In contrast to the traditional "cook and look" workflow, the initial scoping can be much broader and thousands of times faster. Subsequent iterations refine the tuning parameters for each sputtering target (distance to wafer, angle of target arm, bias voltage, and duty cycle) to create samples with shallower gradients for more accurate

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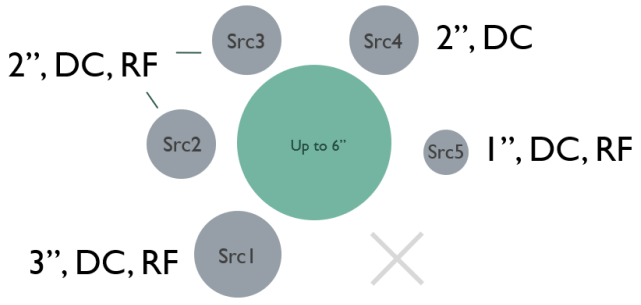

Fig. 3. Configuration of sputtering sources in the 5-target PVD chamber. The sputtering targets surround a wafer of up to six inches diameter. Sources 1, 3, and 5 create 3-component alloys with spatial gradients oriented at 120° angles.

2-D measurement of the material properties within narrower compositional spaces.

Thick film sputtering creates alloys with component atoms distributed roughly uniformly within each region of the sample, while pushing film thicknesses to their limit helps to grow them out of the epitaxially-controlled microstructure regime to more bulk-like. However, properties of the alloys as-grown may differ from their fully bulk counterparts. Heat treatment during and after sputtering can help to achieve closer to the desired microscopic grain structure within the thick film.

Regardless of microstructural issues originating in the growth of the film compared to a bulk material, we specifically seek to prove a core hypothesis on the atomistic scale about what confers radiation damage resistance. We hypothesize that potential energy landscape (PEL) heterogeneity, as quantified by higher variance in migration energy barriers (MEBs), confers primary radiation damage resistance via localization of radiation defects, promoting recombination over long-range transport and clustering. To explain more simply, we hypothesize that a rougher atomistic landscape incurs less damage, and this can be tested regardless of film quality provided that it is controlled for. Our early results in the Ni-Fe system show that PEL heterogeneity does indeed correlate better with radiation damage resistance [11] compared to the prevailing theory of higher mixing entropy [12], though our hypothesis not yet supported enough to be considered true.

This workflow is key to putting this hypothesis, and its resultant design principle, to the test. With this higher-throughput workflow, we can validate the idea of PEL engineering, encouraging heterogeneous MEBs to form by specific alloying elements, and then rapidly put them to the test nearby thousands of close compositions to validate both the hypothesis and the workflow. This could offload much of the work designing new alloys for fusion structural and functional applications to a more automatable, computational process, guiding targeted high-throughput experimental efforts with constant validation.

III. Simulated neutron irradiation

High energy ions can simulate the damage effects of 14 MeV neutrons in the absence of a high flux fusion neutron source [13], [14], particularly the recent demonstration of intermediate energy proton irradiation [15]. The high particle flux and large scattering cross sections of ions can generate a representative number of displacements per atom (DPA) on a timescale much faster than achievable in fast neutron reactors. The fast turnaround time, as well as availability of high current ion beams at the MIT PSFC, makes this workflow attractive for iterative materials development. The irradiation process can be stopped for evaluation of material properties at intermediary levels (*e.g.* 0.1, 1, 10 DPA), or even monitored *in situ* during irradiation [16], to better understand the performance of the material over its expected lifetime.

Ion beam irradiation does not simulate transmutation from neutron irradiation, but in a fusion reactor the cross sections for transmutation are small compared to the scattering events that lead to atom displacements. Our primary concern with respect to transmutation is to avoid constituent isotopes that lead to significant activation after neutron exposure, which does not require simulation of the transmutation process itself. It is possible to add simulated daughter atoms (*e.g.* ^{94}Mo in the case of neutron capture by ^{93}Nb, or ^{4}He for alpha decay) to the sample through ion implantation to investigate the effects of transmutation on materials properties, however this is beyond the scope of our rapid iteration workflow. Higher fidelity studies of radiation induced effects can be performed on a subset of promising alloy compositions, similar to the process described in Section V.

IV. Evaluation of thick films

Evaluation of the thick films focuses on thermo-elastic and electrical properties using transient gradient spectroscopy (TGS) and four-point conductivity techniques. TGS provides localized measurements of the thermo-elastic properties of a thick film. The TGS system uses two counter-propagating high intensity laser pulses to create an interference pattern on the surface of the film. The shock heating of the material due to the lasers generates ultrasonic waves. A lower intensity probe laser detects the ultrasonic waves, which oscillate according to the elastic properties of the material, while the abrupt decay of the initial shock yields the thermal diffusivity of the material. An automated translation stage moves the focal point of the lasers across the sample generating a high resolution map of these properties across the graded alloy as in [17]. Multiple iterations with the simulated irradiation workflow (Fig. 1) provide DPA-dependent measurement of the material properties.

Moreover, employing energy dispersive spectroscopy (EDS) to measure elemental maps of the multi-target combinatorial thick films allows us to measure the exact composition of the substrates at each position, enabling us to correlate thermo-elastic properties with a particular composition. To know the microstructure behind each composition, scattering electron microscopy (SEM) would be used. On this point, being able to characterize the chromium precipitates or secondary phases that could appear in the copper matrix, would be determinant to explain the mechanical properties of the alloy and how they evolve with neutron damage.

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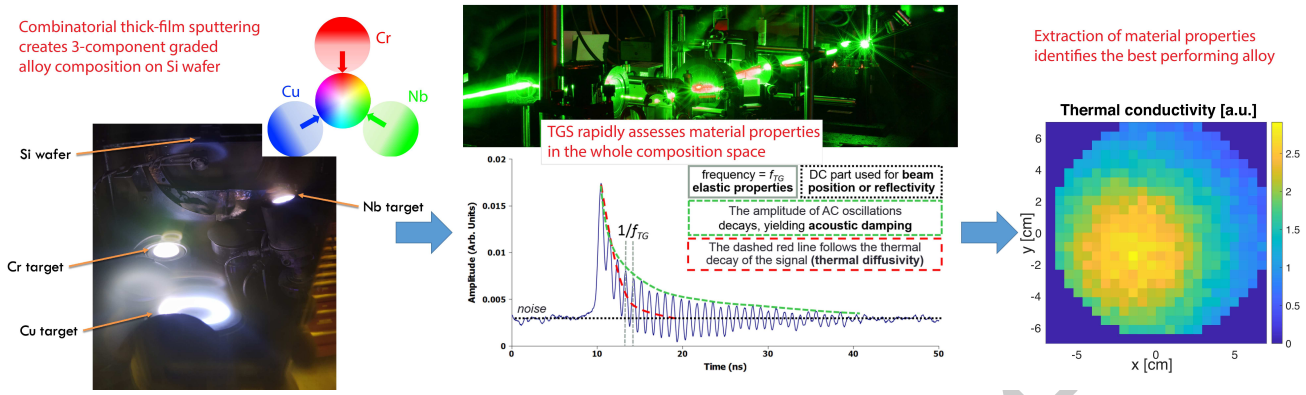

Fig. 4. Visual explanation of our proposed ultra-rapid materials discovery workflow. Co-deposition of multiple materials creates a compositional gradient (color wheel at left) will be analyzed by TGS (middle) to search for islands of stability (right) in thermoelastic material properties following irradiation, themselves correlated directly to radiation resistance. Data shown are synthetic for illustrative purposes.

V. Structural and High Power RF Testing

Analysis of the thick films provides important information on the thermal and electrical properties of the new materials, but structural and high power RF performance tests require destructive testing of larger samples. Traditional methods such as forging or casting can create macroscopic structural samples, however laser sintering is one of the best methods for producing GRCop-84 [6] with high strength at elevated temperature. In addition to tensile/fatigue testing, larger samples are also needed for high power RF breakdown characterization. Since the creation of these larger samples is a low throughput operation, traditional structural and RF test equipment is sufficient to assess the properties of the reduced number of large-scale samples.

VI. Status and Future Work

Pandemic related supply chain issues significantly delayed the implementation of this project. The target and power supply upgrade to sputtering chamber (Fig. 2) is now complete and the system is ready for generation of thick films. Development of new materials will begin from the starting point of the GRCop-84 Cu-Cr-Nb system. The Nb will be replaced first with other Group 5 elements (V, Ta) and common Cu alloying elements (Zr, Al).

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