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Closeout Phase II Qualification of the Thermal Conductivity Microscope for IMCL

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SUMMARY

The FY18 milestone for this work involved the completion of qualification testing of the Thermal Conductivity Microscope (TCM) in preparation for installation in the Irradiated Materials Characterization Laboratory (IMCL). In FY17, the TCM was entered into stage one mockup at the Materials and Fuels Complex (MFC) and initial testing was completed. This initial testing at the MFC centered on the ability to operate the equipment remotely using manipulators and functional testing of the instrument. During FY18, issues noted during initial testing were addressed, a sample heater indexing system was developed, software enhancements were integrated, and a sputter coater for use in the glove box was procured. Additional testing was performed to ensure instrument accuracy and reproducibility. The Phase II Equipment Qualification Plan, MFC-EQP-0267, completed approval on August 14, 2018.

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ACRONYMS

EDMS	Electronic Document Management System
EQP	Equipment Qualification Plan
IMCL	Irradiated Materials Characterization Laboratory
INL	Idaho National Laboratory
IRC	INL Research Center
LFA	laser flash
MFC	Materials and Fuels Complex
TCM	Thermal Conductivity Microscope
TPC	Thermal Properties Cell

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CLOSEOUT PHASE II QUALIFICATION OF THE THERMAL CONDUCTIVITY MICROSCOPE FOR IMCL

1. INTRODUCTION

The Thermal Conductivity Microscope (TCM) is an instrument designed to measure the thermal properties of irradiated samples. It is being developed for installation in the Thermal Properties Cell (TPC) in the Irradiated Materials Characterization Laboratory (IMCL) at the Materials and Fuels Complex (MFC), which is part of the Idaho National Laboratory (INL). The TCM simultaneously measures the thermal diffusivity and conductivity using a modulated thermoreflectance technique. This measurement approach involves measuring the temperature field spatial profile of samples excited by an amplitude modulated, continuous-wave laser beam. A thin gold film is applied to the samples to ensure strong optical absorption and to establish a second boundary condition that introduces an expression containing the substrate thermal conductivity. The diffusivity and conductivity are obtained by comparing the measured phase profile of the temperature field to a continuum-based model. The TCM has been designed to operate in a radiation hot cell environment. It can be controlled remotely via the software interface, and sample loading is compatible with hot cell manipulators.

2. FY18 MILESTONE

The title of the FY18 milestone was "Closeout Phase II Qualification of the Thermal Conductivity Microscope for IMCL." The FY18 milestone builds on the FY17 milestone that was documented in the report "Enter TCM and Associated Equipment into Stage One Mockup" (INL/EXT-17-43232). During FY17, the TCM was transported to the MFC mockup shop, assembled, and tested. The TCM was operated using remote manipulators, and tests were run on standard samples. Lessons learned during this initial testing were used in FY18 to address issues, improve the TCM, and prepare the instrument for installation in the IMCL. The following tasks were completed as part of meeting this milestone:

1. Design/implement indexing system for TCM furnace
2. Resolve issues identified in Phase II mockup trials
3. Complete TCM instrumentation software development
4. Procure installation-ready sputter coater for specimen prep.

These tasks are discussed in detail in the following sections.

2.1 Design/Implement Indexing System for TCM Furnace

Thermal property measurement of samples at elevated temperatures using the TCM was tested early in the TCM development process. The tests were conducted using an off-the-shelf Linkam model TS1000 heating stage. In order to accommodate this heating stage, the TCM sample stages had to be manually reconfigured. The object of this tasks was to find a way to integrate the heating stage onto the TCM sample stage such that it could be inserted or removed using the manipulators that will be available in the IMCL TPC glove box.

The TS1000 heating stage and controller allow automated temperature control and have a number of desirable features for use with the TCM. The heating crucible will accommodate a 17mm diameter x 3mm high sample, and heating rates and temperatures can be programmed using a control pad. However, sample loading requires the heating stage lid to be unscrewed which would be difficult to perform using manipulators. Integration with the TCM sample stage would also require an adaptor plate that would provide marginal clearance between the objective lens and the stage lid. To resolve these issues, a new heating stage base and lid were designed that could incorporate the standard TS1000 heating crucible, windows, o-ring seals, and connections, and use the Linkam stage controller. The redesigned base incorporates indexing pins that mate with the TCM sample stage top plate. This allows the standard sample plate to be removed and replaced with the heating stage using remote manipulators. The TCM software has been revised such that the presence of the heating stage will be detected during the crash prevention scan (seesSection 2.3). Sample stage motion is then limited to prevent the stage from running into the objective lens. The redesigned lid now uses manipulator-friendly levers to clamp the lid in place rather than a threaded connection that requires the lid to be unscrewed. With the lid removed, sample placement in the heating crucible will require manipulator compatible tweezers or a suction type lifting apparatus. The left pane of Figure 1 shows an image of the sample heater in position on the TCM sample stage located in the sample load position. The right pane of Figure 1 shows a sketch of the sample heater with the lid removed. The heating crucible is located in the center of the base cavity.

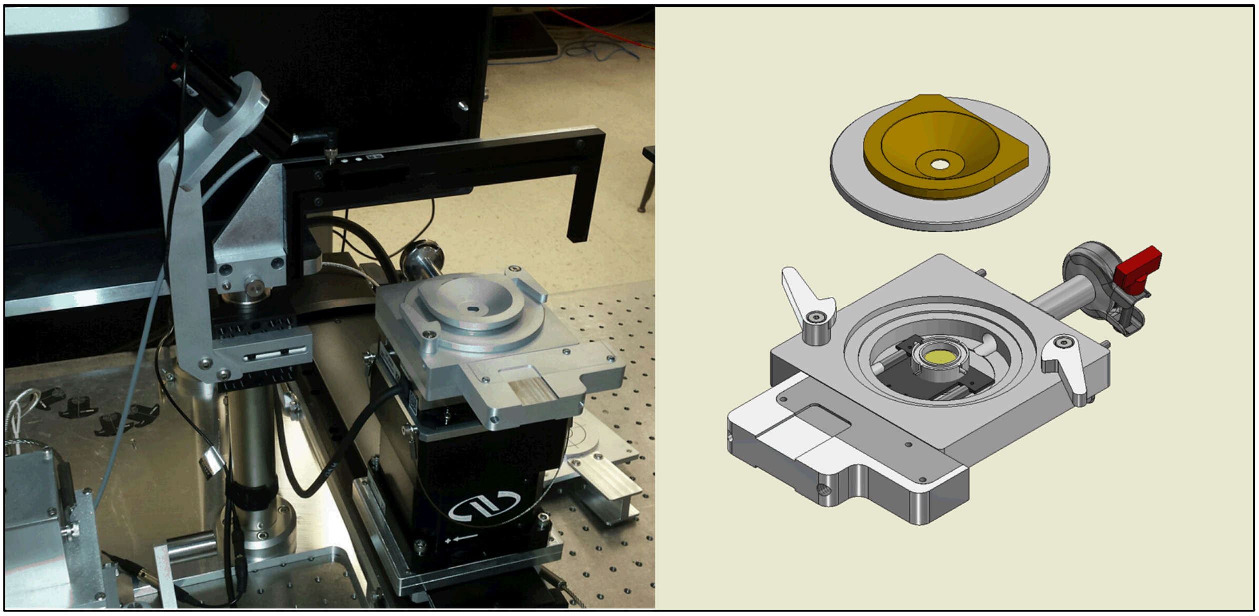

Figure 1. TCM sample heater in position on the sample stage (left). Exploded view of the sample heater (right).

2.2 Resolve Issues Identified in Phase II Mockup Trails

The TCM was placed in mockup in September 2017. Initial testing verified the ability of the manipulators to place samples and operate the thickness monitor. Operation and accuracy were verified by taking measurements on standard samples. There were, however, issues noted and ideas presented by the manipulator operators that would aid in sample placement. These are discussed in this section.

The manipulator operators suggested etching reticle type lines on the sample stage and thickness monitor drawer surface to make it easier to place the sample. These changes were incorporated, and the rim of the thickness monitor sample pocket was also chamfered to make sample placement easier.

The mockup shop personnel also raised a concern about the operability of the stage motors in an argon environment. Standard graphite brushes in DC motors require oxygen to maintain their lubricity. In an inert environment, the brushes can quickly deteriorate, resulting in failure of the motor. Some DC motors will function normally in an inert environment while others will fail in the first few hours of operation. The stages used in the TCM were procured from Newport Corp. The company was contacted, but they could not confirm the suitability of the stages for use in an argon environment. In order to verify the suitability of the stages, a duplicate motor was procured and tested in an argon glove box at MFC. No issues with the motor were noted during the 54.5 hour test. The test results were documented in INL/INT-17-44259.

Additional issues concerning the thickness monitor were noted and addressed. The thickness monitor is used to determine the thickness of a gold film applied to a reference sample that is coated simultaneously with the sample of interest. The technique involves measuring the transmission of a 488nm beam of light through the coated standard. The amount of light reaching the detector is measured first with a clear path and then with the coated standard inserted prior to the detector. The film thickness can then be determined from the ratio of the light transmission. The maximum film thickness that can be measured depends on the power of the light delivered to the thickness monitor. A small 488nm laser is used as the light source. The light is delivered via optical fiber to the thickness monitor. Initially, single mode fibers and a single mode glove box feedthrough were used. This resulted in a significant loss of beam power between the laser and the thickness monitor. After initial mockup testing, multimode optical fibers and components were procured and installed. This increased the light power at the thickness monitor from 450 µW to 13.7 mW, and increased the maximum film measurability from about 125µm to about 170µm.

Two other thickness monitor issues were also addressed. First, it was noted that the thickness monitor would move while opening the drawer using the manipulators. This was resolved by attaching the thickness monitor to the TCM base. The second issue was the possibility of the sample becoming dislodged inside the thickness monitor. The sample is placed in a shallow pocket in the thickness monitor drawer plate which rotates into measurement position. As the drawer rotates, the sample moves within a slot to prevent lateral movement. However, if the drawer were aggressively closed, the sample could be dislodged and settle with its side instead of its face resting in the pocket. This would prevent it from exiting the opening at the front of the thickness monitor. To resolve this issue, a cover was placed over the slot which prevents the sample from tipping in the drawer pocket.

Another issue resolved after mockup concerned the mounting of the 532nm probe laser. Prior to mockup, it was noted that the noise floor of the TCM signal had increased from the prototype instrument. After a significant diagnostic effort, it was determined that the increased noise floor resulted from mounting the probe and pump lasers in close proximity. For initial mockup testing, the probe laser was temporarily mounted in the back of the equipment rack. When the TCM was returned to the INL Research Center (IRC), a separate box for the 532 laser was procured and mounted in the equipment rack.

2.3 Complete TCM Instrumentation Software Development

The software code and graphical user interface for operation of the TCM was initially described in a status report issued in August 2016 (see INL/LTD-16-39645). Since that time, additional features have been added and bugs fixed. The most significant additions were the crash prevention and automated operation of the thickness monitor.

The crash prevention is a method of preventing an operator from inadvertently moving a sample stage and driving the sample into the microscope objective. The system consists of a slot detector along with software to control and limit the sample stage range of motion. The slot detector is a “U” shaped device that transmits a beam of light between the legs of the “U” and detects when the beam is broken. The TCM control software allows three main stage locations: load position, wide view position, and scan position. When the stages are moved from the load position, the crash prevention routine is invoked. The stage will initially move to a start positon in front of the slot detector. The stage is then slowly moved through the slot detector. If the beam is broken, the position is recorded, and the stage is lowered until the beam is no longer blocked. The scan continues until the beam is again broken or the entire stage surface has been scanned. In this manner, the maximum height of the sample on the stage is determined. The software then limits the maximum height of the stage to prevent running the sample into the objective. A special case exists when the sample heater is in place. The heating stage requires the objective lens to be positioned below the rim of the heating stage lid, so the standard stage limits will not work. With the heating stage in place, the slot detector beam is broken at the starting point of the scan. The software then recognizes the presence of the heating stage and invokes special stage motion limits that prevent interference between the heating stage and the objective. An added benefit of the crash prevention is that the sample is automatically moved very close to focus in either the wide angle or scan positions. A screen shot during a crash prevention scan is shown in Figure 2. The measured sample height vs scan position is shown on the screen in the foreground. The screen in the background shows the camera image as the sample is passing through the slot detector which is located on the lefthand side of the image.

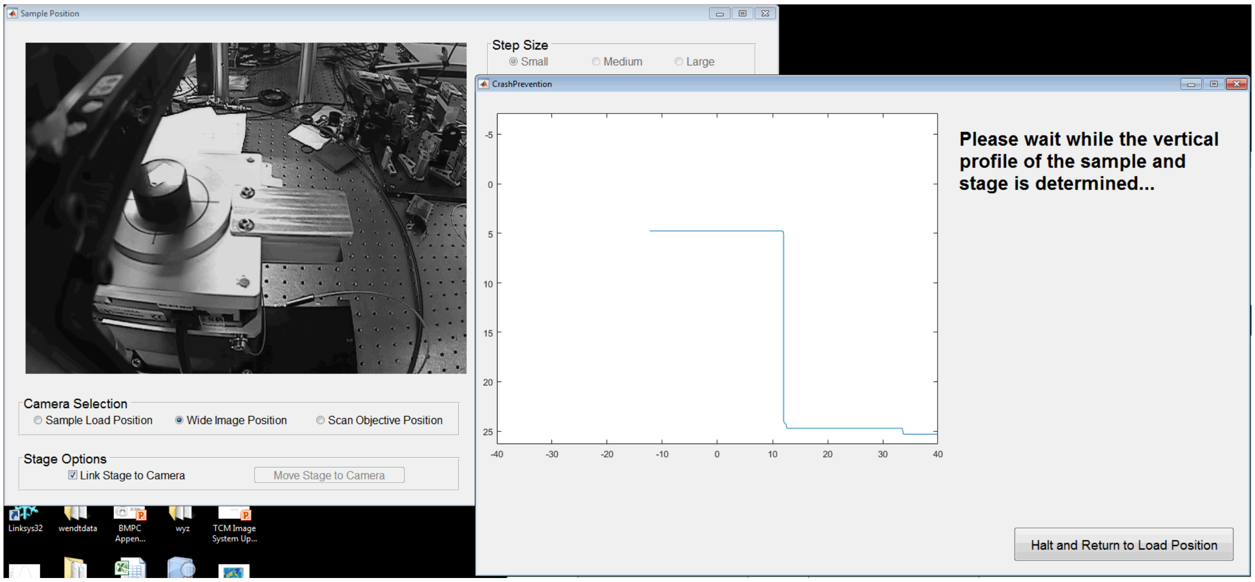

Figure 2. TCM crash prevention. Cash preventions progress (foreground), sample passing through slot detector (background).

Samples measured using the TCM require a thin gold coating, and the thickness of the coating is an important parameter used in the data analysis routine. As noted above, the thickness monitor was developed to measure the coating thickness of a BK7 glass standard which is coated alongside the sample. To simplify the use of the thickness monitor, software was developed to automate the process. A new button was added to the TCM software main page for the thickness measurements. When clicked, a new screen appears that allows the measurement parameters to be adjusted, and buttons are provided to take readings with and without the standard in the light beam. When both readings have been made, the thickness is automatically displayed. The thickness monitor screen is shown in Figure 3.

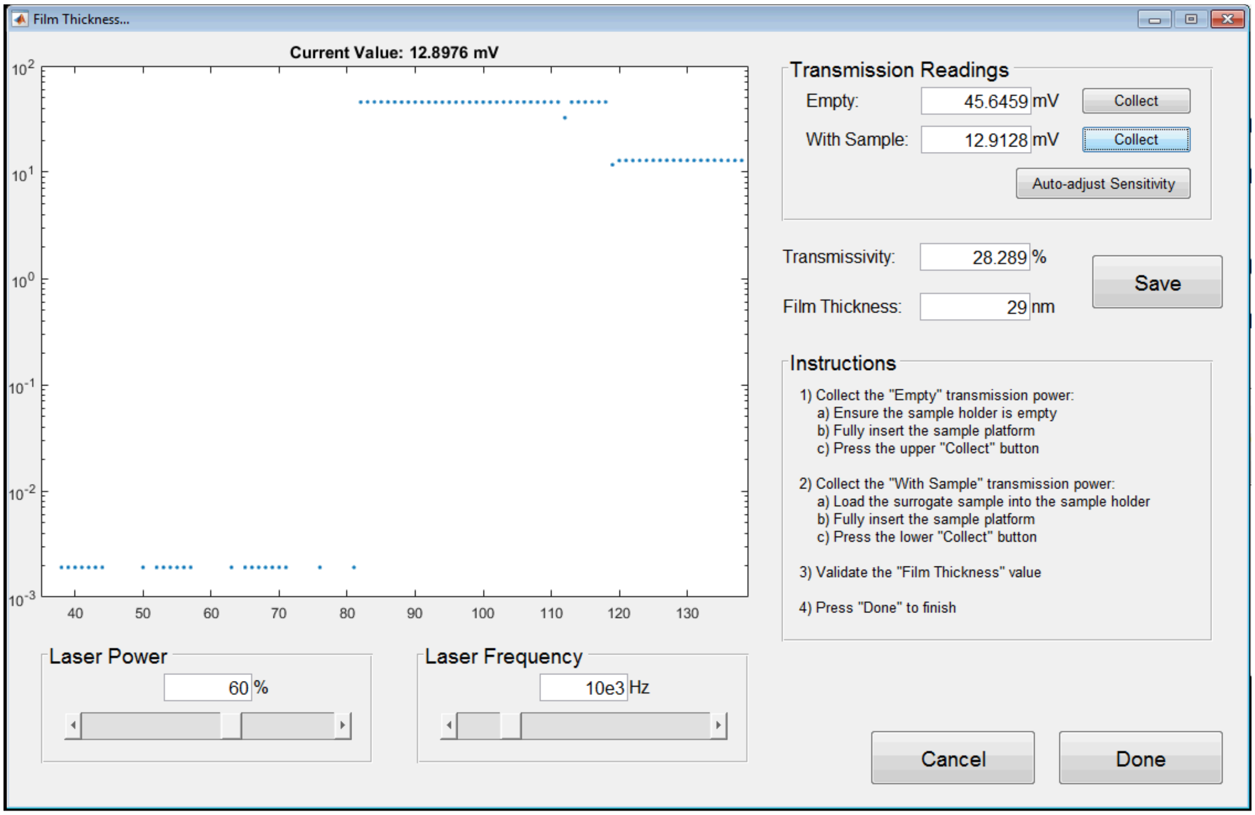

Figure 3. Thickness monitor screen.

Another software addition allows the DC voltage read by the photoreceiver in the TCM to be displayed. This feature can be accessed by clicking the Advanced Settings button on the Collect Data screen. This reading is a measure of the probe light reaching the photoreceiver and can be used as a diagnostic tool to optimize the probe laser power.

With the features discussed above added to the software along with minor bug fixes, the TCM instrumentation software is currently in a ready for operation state. It is anticipated that additional features and improvements will be made as time and funding allow.

2.4 Procure Installation-ready Sputter Coater for Specimen Prep

The thin gold film required on the sample for measurement of thermal conductivity using the TCM is applied using a sputter coater. During instrument development, a Cressington Model 208HR benchtop sputter coater was used to apply this coating. This benchtop model does not lend itself for use in a glove box using remote manipulators. Processes requiring sputter coating in other shielded glove boxes at IMCL use Quorum Technologies Q150GB sputter coaters to apply thin films. Since the feedthroughs, cabling, and connections have already been developed for this sputter coater, it was determined that a similar sputter coater should be used for the TCM. An order was placed, and the sputter coater was delivered on July 19, 2018. After initial testing, it was determined that a modified planetary stage would be required. The stage has been designed and is currently being fabricated. This sputter coater is designed for use in a glove box and will require no modifications for installation. The sputter coater as set up in the back of Lab C10 at the IRC is shown in Figure 4. Note the hand held control pad in the foreground.

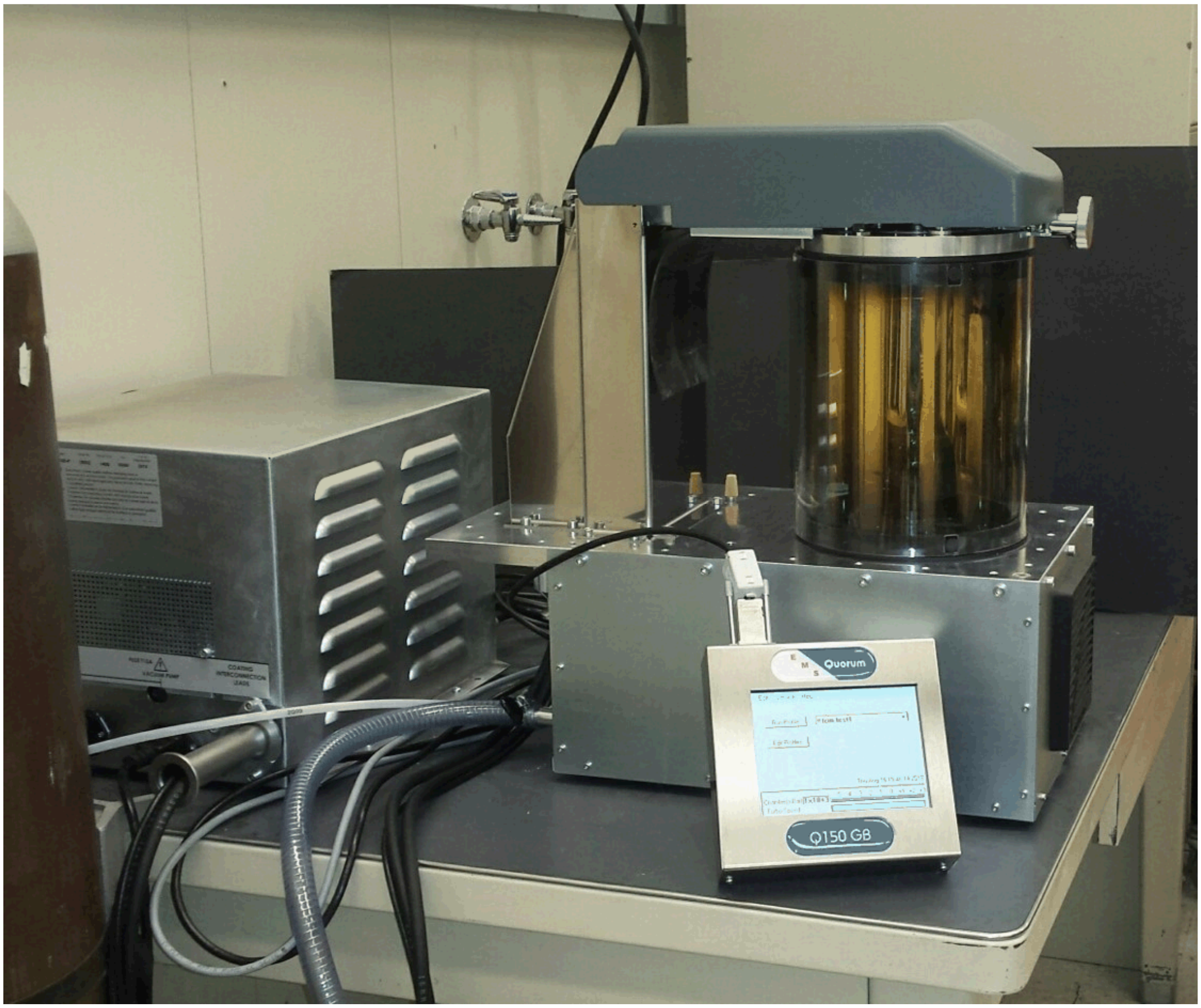

Figure 4. Glove box ready sputter coater.

2.5 Run Final Test on Standards and Present Results

Mockup testing of the TCM was initiated in the mockup shop at MFC near the end of FY17. After initial testing at MFC, the TCM was returned to the IRC for further testing. Measurements to finalize the mockup testing of the TCM using the standard samples were conducted at the IRC from August 7 - 10, 2018. As stated in the FY17 report, the inability to measure the thickness of gold films thicker than about 125 nm was an issue for the ZnSe sample. Using a multimode optical fiber as discussed in Section 2.2 increased the maximum film thickness measurement to ~ 170 nm. However, the optimized film thickness for samples with a thermal conductivity higher than ~20W/m/K is more than 200nm. Thicker films can be applied, but the ability to measure the films was lacking. To resolve this issue, a dual height sputtering method was devised. The distance from the sputtering target to the sample is one of the variables that effects the sputtering rate. By placing the sample of interest closer to the sputter target and the BK7 reference sample further away, the sample of interest will receive a thicker film than the BK7 while coated simultaneously. The thickness of the film on the BK7 reference sample can then be measured, and the sample film thickness can be determined based on a known scaling factor. This method was validated using the Cressington 208HR sputter coater and placing a few pairs of BK7 samples on posts of different heights. The film thicknesses achieved with different coating time periods are summarized in Table 1 below. Coating rates of 0.25-0.28 nm/s and 0.53-0.55 nm/s were achieved on the low and high posts, respectively, and the calculated scaling factor was ~1:2. The film thickness is used in determining the reference sample film properties, and both the film thickness and film properties are used to determine the target sample properties. Errors in film thickness are compensated in this process, so resulting target sample property errors are greatly diminished.

Table 1 Thickness monitor results of the low-high stage approach.

Time(s)	**Film thickness (nm, low stage)**	**Film thickness (nm, high stage)**
60	15	32
100	27	53
150	42	82

Four standard samples were used for the final TCM testing: fused silica, CaF2, ZnSe, and ZnS, with thermal conductivities ranging from ~1W/m/K to ~30W/m/K. The fused silica and CaF2 were coated along with their BK7 reference samples. The ZnSe and ZnS samples were coated using the low-high approach described above. These samples used two BK7 reference samples. One BK7 was coated next to the sample for film property determination, and the other was coated on the lower post for film thickness determination. Five measurements at different locations were made on each sample. The details of the results are listed in Table 2 (k-conductivity, and D-diffusivity). The reference values were obtained from laser flash (LFA) measurements. Measured film thicknesses are shown in parenthesis below each sample.

Table 2 The summary of TCM results on standard samples, with the LFA results as reference.

Sample	**k [W/m/K]**	**D [mm^2/s]**	**Ref. k**	**Ref. D**	**Sample**	**k [W/m/K]**	**D [mm^2/s]**	**Ref. k**	**Ref. D**
Fused Silica (70nm)	1.415	0.911	1.41 (4.3%)	0.85 (8.9%)	CaF2 (82nm)	13.46	3.514	9.75 (11.2%)	3.59 (-10.4%)
	1.549	0.952				9.956	3.089		
	1.364	0.876				9.615	3.159		
	1.524	0.949				9.565	2.959		
	1.499	0.932				11.62	3.355		
Mean	**1.47**	**0.924**			**Mean**	**10.84**	**3.215**		
ZnSe (188nm)	16.3	8.999	17.8 (-1.4%)	9.97 (-4.1%)	ZnS (225nm)	33.2	15.22	32.7 (-12.8%)	15.5 (-10.0%)
	15.6	8.942				25.27	12.8		
	19.25	10.02				27.55	14.39		
	19.92	10.66				28.95	13.56		
	16.75	9.213				27.47	13.71		
Mean	17.56	**9.566**			**Mean**	**28.49**	**13.94**		

As shown in the table, good agreement is achieved between the TCM and LFA results. The differences between the mean measurement values and the reference values are shown in parentheses below the reference values. Differences between the mean thermal conductivities are less than 5% on fused silica and ZnSe, and ~10% on CaF2 and ZnS. Similar accuracy was also obtained on the thermal diffusivity values. The results at different locations are also very consistent for both thermal conductivity and diffusivity on all four standard samples despite measurement locations millimeters away from each other. The results are also consistent with the results obtained at the MFC mockup shop in FY17.

Two other important issues identified and investigated during FY18 testing were the possible microstructural changes of the gold film coating and the anisotropy measurement capability. During testing, it was observed that high pump powers could lead to changes in the film property measurements on the BK7 reference samples. The temperature rise during a measurement depends on the intensity of the focused laser beams, the heat capacity of the film, and the thermal conductivity of the sample. High laser powers combined with very thin films coated on low thermal conductivity samples such as BK7 could result in a temperature rise high enough to alter the microstructure of the thin film, thus altering its thermal properties. The film properties of the BK7 reference sample are used in the data analysis of the target sample, so alteration of the thin film will result in errors in the target sample measurement. To investigate this phenomenon, a test was performed on a BK7 substrate with a 56-nm gold thin film coating. The pump laser power was adjusted from 10% to 100% (the 10% setting results in an average pump laser power of about 1.5mW reaching the sample surface). The measured film thermal conductivities (k_f) with different power settings are summarized in Table 3.

Table 3 Film thermal conductivity measurement results with different pump laser power settings. The film thickness is 56 nm.

Power setting	**Measured k_f [W/m/K]**	**Signal amplitude @10kHz [mV]**
10%	99.36	2.116
20%	99.6	3.789
50%	113.11	6.329
100%	127.02	9.427

As Table 3 shows, the measured film thermal conductivity increases from ~100W/m/K with a 20% pump laser power setting to ~130W/m/K with a 100% power setting. A measurement with a 10% power setting was immediately conducted following the 100% power measurement at the same location, and the measured thermal conductivity remained at the higher level. This is an indication that the film microstructure had been permanently changed by the laser heating. If the film is being annealed, the thermal conductivity increase is expected as the annealing process can remove or rearrange the defects in the microstructure and reduce the total thermal resistance.

A quick way to test for annealing is to compare the signal amplitude vs. laser pump power. The thermal wave amplitude should change linearly with the incident heat flux but decrease with film thermal conductivity. Therefore, the signal amplitude should increase linearly with the pump laser power. As Table 3 shows, the signal amplitude at 10kHz with the 50% pump laser power setting is only ~3 times higher than the one with the 10% power setting. As a comparison, a similar test was conducted on a BK7 sample with a 265-nm gold film. The results are given in Table 4. Due to the heat capacity increase provided by the thicker film, the annealing temperature is not achieved at the 100% power setting. As can be seen, the measured film thermal conductivity varies only within the normal uncertainty range, and the linear dependence of the signal amplitude to the power setting remains consistent.

Table 4 Thermal conductivity measurement of a 265-nm film with different pump laser power settings.

Power setting	**Measured k_f [W/m/K]**	**Signal amplitude @10kHz [mV]**
10%	152.49	0.598
20%	148.87	1.143
40%	149.64	2.276
60%	149.97	3.411
80%	151.56	4.587
100%	151.47	5.788

The results of the investigation suggest that an excessively high pump power should not be used when measuring thin films on thermally slow materials (such as BK7 or fused silica). Higher pump power does increase the signal amplitude and signal to noise ratio, so a compromise must be made to achieve adequate signal amplitude without excessive laser power. When in doubt, a quick laser power vs signal amplitude test can be run to ensure the pump power to signal amplitude ratio remains in the linear region. The details of the power settings used for the standard sample testing are given in Table 5 as a reference. These values can be used as a guide, but optimal settings in the future may vary due to changes in the transmission losses in the optical fiber couplings and feedthroughs.

Table 5 Power settings used in the standard sample testing.

Sample	**Film thickness [nm]**	**Power setting (film)**	**Power setting (substrate)**
Fused silica	70	10%	10%
CaF_2	82	10%	50%
ZnSe	188	20%	60%
ZnS	225	20%	80%

The TCM has the ability to perform the spatial scan in the direction specified by the user. This capability provides the potential to probe thermal properties vs grain orientation. For isotropic materials, scans in orthogonal directions should yield similar results while anisotropic materials would show a relationship between the measured properties and the scan direction. The ability of the TCM to measure anisotropy is limited by the precision of scans performed in orthogonal directions. This was tested by measuring several isotropic materials in orthogonal directions and comparing the results. Several sets of measurements were performed at different locations on gold film, ZnSe and ZnS, and the results for each sample were averaged. Differences in the results from orthogonal scan directions were below the measurement uncertainty (conductivity 0.1% - 5.7%, and diffusivity 0.7%-1.6%), as shown in Table 6. Materials with anisotropy lower than the uncertainty could not be accurately measured. An issue observed while taking measurements in orthogonal directions must be noted: a systematic error can occur when the laser beams are not perfectly centered. It is possible to have the pump and probe beams centered in one scan direction but offset in the orthogonal direction. In this case, the pump beam will scan directly over the probe beam in one direction, but the pump beam will be offset from the probe beam when scanned in the orthogonal direction. Such a scenario will yield a significant difference in the results of the orthogonal scans. In extreme cases, the difference can be over 20%. The user must verify beams are well centered to minimize errors in orthogonal scans. One method to check beam centering is to perform multiple scans and alternate the order of the scan direction. Well-centered beams would yield consistent results in each direction.

Table 6 Results of the anisotropy testing.

Sample	**k (0/90) [W/m/K]**	**D (0/90) [mm^2/s]**	**Difference**
Gold film (130nm)	129.12/124.05	N/A	4.1%/-
ZnSe	24.13/24.10	11.81/11.62	0.1%/1.6%
ZnS	46.54/49.35	18.74/18.88	5.7%/0.7%

3. CONCLUSION

The FY18 milestone of closing out Phase II qualification of the TCM was completed in August 2018. The Phase II qualification was initiated in September 2017 when the TCM was delivered to the mockup shop at MFC. The instrument was assembled and the ability to load samples and operate the thickness monitor using remote manipulators was verified. Measurements on standard samples were performed to verify accuracy and operability of the instrument. The TCM was returned to the IRC, and testing continued during FY18. Recommendations made by the mockup shop operators were incorporated, and issues noted during initial testing were resolved. New software features were added and software bugs were fixed. A glove box compatible sputter coater was procured, and a sample heating stage was modified for compatibility with the TCM. Final qualification testing on standard samples was completed in early August 2018. The qualification plan MFC-EQP-0267, "Remote Equipment Qualification Plan (Phase I and II) Testing of Thermal Conductivity Microscope," was approved and entered into EDMS on August 14, 2018.