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INTRODUCTION

Many advanced nuclear reactor designs require new fuel, cladding, and structural materials. Data are needed to characterize the performance of these new materials in high temperature, oxidizing, and radiation conditions. To obtain this data, robust instrumentation is needed that can survive proposed test conditions. Traditional methods for measuring temperature in-pile degrade at temperatures above 1080 ºC. Hence, a project was initiated to develop specialized thermocouples for high temperature in-pile applications (see Rempe and Wilkins, 2005). This paper summarizes efforts to develop, fabricate, and evaluate these specialized thermocouples.

BACKGROUND

Types of instrumentation that might be employed for in-pile, high temperature applications were first evaluated. For temperatures above 1100 ºC, previously investigated in-pile instrumentation methods are limited to thermocouples (with thermoelements consisting of molybdenum, niobium, or zirconium, or their alloys), Johnson Noise Power Thermometers (JNPT), and ultrasonic thermometers (UTs). UT and JNPT techniques weren't considered because of the cost and complexity associated with their probe and signal processing equipment. Optical pyrometer techniques were eliminated because viewing ports are not typically available. Likewise, considerably more development is needed before optical fiber methods overcome difficulties associated with signal degradation in radiation fields. Hence, specialized thermocouples were deemed to be the simplest and most economic approach for in-pile high temperature measurements. However, such thermocouples have not been used for nearly a decade, and improved versions were needed.

APPROACH

Table I lists materials initially considered for thermocouple components. Available data indicate that these materials are suitable for high temperature irradiation conditions.

TABLE I. Candidate Thermocouple Component Materials

Component	Candidate Materials
Thermoelement	Molybdenum,* Zircaloy-4, Titanium-45% Niobium, Niobium-1%Zirconium
Insulator	Aluminum Oxide, Hafnium Oxide
Sheaths	Titanium, Zircaloy-4, Niobium-1%Zirconium

* Evaluations considered several types of Molybdenum: undoped Mo, Mo-1.6% Nb, KW-Mo (doped with Tungsten, Silicon and Potassium), and ODS-Mo (containing Lanthanum Oxide).

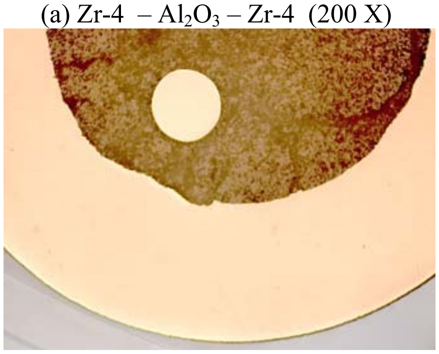
Thermocouples were fabricated and evaluated at INL's High Temperature Test Laboratory (HTTL), which has key equipment and trained staff required for such activities. Several types of evaluations were performed. Using representative thermocouple samples, tests evaluated the potential for materials interactions between insulation materials and candidate sheath and thermoelement materials. Then, mandrel-wrap tests on wires exposed to high temperatures were completed. Last, calibration tests were performed for candidate thermocouples.

RESULTS

Materials Interaction Tests

Materials interaction tests were completed by heating representative thermocouple samples in gettered argon at 1300 and 1600 °C. As shown in Figure 1(a), 1300 °C tests showed that significant materials interactions occurred with samples containing Zr-4 thermoelements, Al_2O_3 insulators, and Zr-4 sheaths. However, 1600 °C results for Nb-1%Zr and Mo thermoelement wires and Nb-1%Zr sheaths indicate that no discernible materials interactions occurred between these materials and HfO_2 insulators (see Figure 1(b)). In summary, tests indicate that several thermoelement wire materials (Mo, Nb-1%Zr, and Ti-45% Nb) appear viable with HfO_2 insulation and Nb-1% Zr sheaths. Other sheath and insulator materials may also be viable if temperatures remain below 1300 °C.

(a) Zr-4 – Al_2O_3 – Zr-4 (200 X)



(b) Mo – HfO_2 – Nb-1%Zr (100X)

Fig. 1. Results from 1300 ºC materials interaction tests using representative thermocouple samples.

Ductility

Mandrel-wrap tests on wires exposed to temperatures up to 1600°C provided insights about thermoelement embrittlement. Samples were wrapped on mandrels of two, five, ten, and twenty times the wire diameter. Those metals that wrap without damage on a small-diameter mandrel after high-temperature exposure are better candidates from the standpoint of embrittlement. Most thermocouple wires exhibited suitable ductility. The one exception, undoped Mo wire, recrystallizes at 1200 °C. This wire was brittle after heating at 1300 °C. The other tested Mo wires (e.g., KW-Mo, ODS-Mo, and Mo-1.6%Nb) remained ductile, even after heating at 1600 ºC.

Thermoelectric Calibration

Calibration tests were completed for candidate thermocouple combinations. Results (see Figure 2) indicate that the thermoelectric response is single-valued and repeatable for the candidate thermoelements considered. In addition, results indicate that the high temperature resolution is acceptable for all thermocouple element combinations considered (although some combinations are limited due to materials interactions at temperatures below 1600 ºC). The selection of the thermoelement wire combination will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1% Zr or Mo-1.6% Nb / Nb-1% Zr combination is recommended.

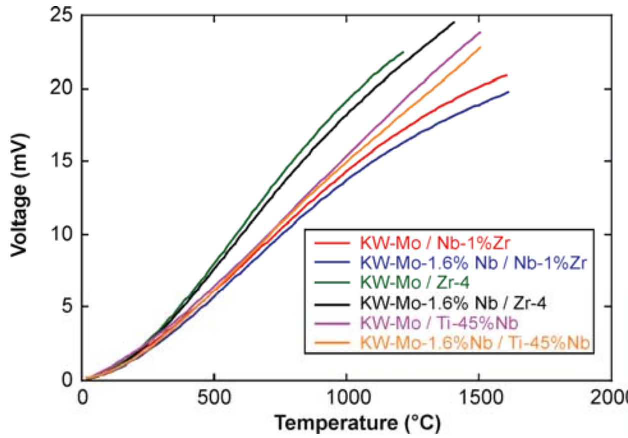


Fig. 2. Calibration curves for candidate thermocouples.

SUMMARY

Results from efforts to develop, fabricate, and evaluate specialized high-temperature thermocouples for in-pile applications suggest that several material combinations are viable. Tests show that several low neutron cross-section candidate materials are resistant to material interactions and remain ductile at high temperatures. In addition, results indicate that the thermoelectric response is single-valued and repeatable with acceptable resolution for the candidate thermoelements considered. The selection of the thermocouple materials will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1%Zr and Mo-1.6% Nb / Nb-1%Zr combinations are recommended. However, these results are preliminary. Additional testing is underway to evaluate the effects of heat treating, thermal cycling, and long duration testing.

ACKNOWLEDGEMENTS

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