On Measuring Material Changes at Molten Salt Reactor Temperatures in a Thermal Convection Loop with Guided Ultrasound Waves

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ABSTRACT

Molten salt reactors (MSRs) for nuclear power generation contain liquid halide salts that may be highly corrosive at operating temperatures. The safety and economic viability of MSRs rely upon the continuous, *in-situ* characterization of the structural integrity of their conduits. Off-line nondestructive inspections of components are simply too expensive, making structural health monitoring in this harsh environment a critical technology. High-temperature capable transducers, such as fused silica optical fibers with a softening point of about 1600 °C, can be utilized to continuously monitor MSR containers. We are using a thermal convection loop (TCL) at Oak Ridge National Laboratory (ORNL) to develop structural diagnostic technology for continuous, autonomous in-situ monitoring of MSR conduits with corrosive molten salts. High temperature- and radiation-resistant fused silica fiber optic sensors will measure the effects of chloride salt corrosion on alloy 600 tubing. Structural material changes in alloys containing the molten chloride salts will be measured via guided ultrasound waves in alloy tubes, detected by high-temperature capable fiber optic sensors. Distributed fiber optic sensors can localize structural changes with position-sensitive measures.

The described tasks are:

- launching guided wave modes at elevated temperatures in alloy 600 tubing in a thermal convection loop, and
- detecting tube ultrasound responses at corrosion-prone locations in the TCL piping with fiber optic sensors. TCL studies at MSR operating temperatures could provide a basis for diagnostic systems that measure material changes in more complex structures.

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INTRODUCTION

Intermittent renewable energy sources, such as solar- and wind-based, have undergone accelerated development. In the wake of aiming to reach net zero carbon emissions by 2050, continuous energy sources based on nuclear have gained renewed momentum as well. A new generation of higher efficiency nuclear fission based reactors operating at atmospheric pressure and having higher fuel efficiency per unit energy include molten salt reactors (MSRs). The salts concerned as the primary coolant, either chlorides or fluorides, remain liquid without pressurization from ~ 500 °C to 1400 °C (MSR schematic [1] is shown in Figure 1). A variety of MSR designs are being developed as they offer a higher efficiency energy source than traditional reactors with larger power capability, to 2400 MW.

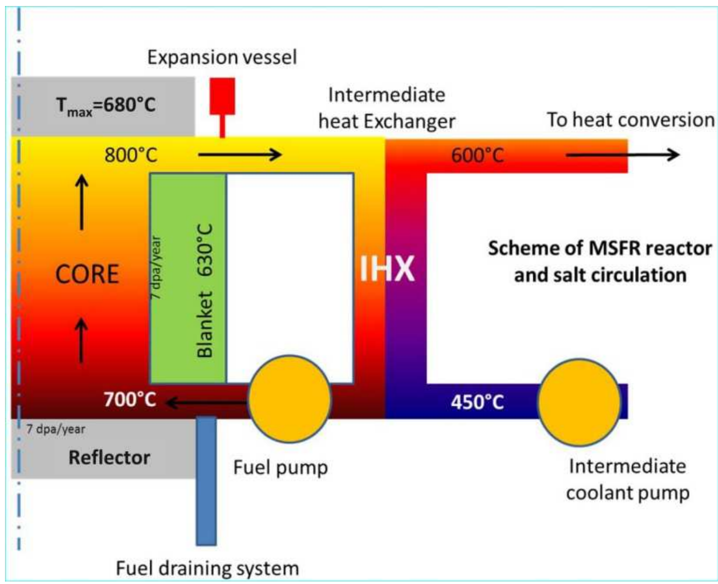

Figure 1: MSR Schematic: showing expected temperatures [1]

Molten halide salts can be highly corrosive. However, combinations of purified salt and corrosion resistant alloys (CRA) have demonstrated good compatibility [2] over a few ten thousand hours showing negligible effect at prototypical MSR temperatures. Nevertheless safety cannot be assumed for reactor structural integrity as their predicted operational lifetimes are up to 50 years (438,000 h), and practical continuous structural monitoring with reliable corrosion diagnostic capability would provide immense value.

Ultrasound is a widely accepted methodology for accurate non-destructive evaluation of materials changes, undergoing cracks or pitting. Structural monitoring with guided ultrasound [3-8] has also been used for evaluating changes/damage in pipes and to assess the strength of structures; these are reported only for lower temperatures up to about 250 °C. Commercial piezoelectrics transducers can operate reliably up to this temperature as they are limited by their

Curie temperatures [9,10]. Extreme temperature (1000 °C) resistant optical fibers can overcome this limitation.

Materials and Methods

(i) TCL Monitoring with Molten Salt at Extreme Temperature

The validity of corrosion detection with guided ultrasound measured by FBGs technique is being tested on a thermal convection loop (TCL) structure at Oak Ridge National Laboratory. The TCL constructed from a corrosion resistant alloy (CRA) shown in Figure 2, is the test-bed for developing structural diagnostic technology for continuous, autonomous in-situ corrosion monitoring of MSR conduits. The CRA structure will be subject to molten salt up to 700 °C;

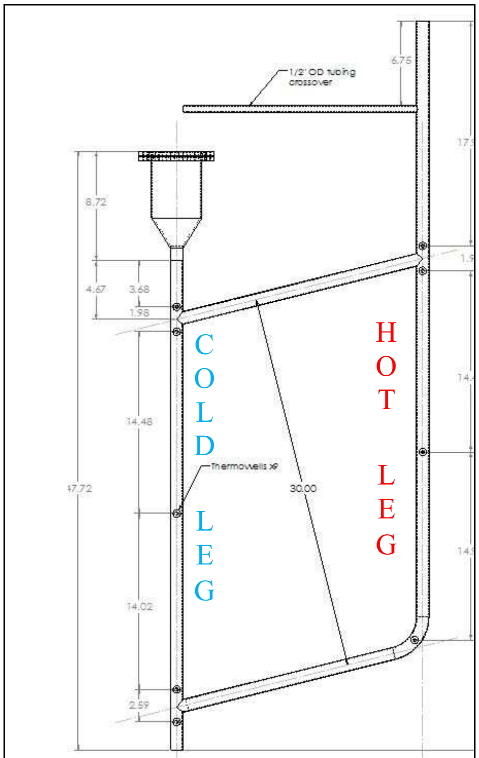

Figure 2: TCL schematic

A flowing experiment, with transducer placement, will be conducted using the harp - shaped (~0.75 m tall by 0.5 m wide) TCL, which has been described in detail else-where [11-12]. The TCL is used to test flowing salt in a temperature gradient in order to assess compatibility and, in particular, mass transfer where material dissolved by the salt on the hot leg re-deposits on the cold leg. Typically, the temperature-gradient is ~100°C, e.g., from 550 °C to 650 °C. The flowing salt experiments is conducted in the TCL made from Ni-based alloy 600 (Ni-15wt.%Cr-9%Fe) tubing (25 mm OD x 1.2 mm wall). For simplicity, a dried commercial 40:40:20 K:Mg:NaCl salt will be used. Three resistively heated furnaces heat the hot leg and temperatures and will be monitored using six type K thermocouples in thermowells in the hot and cold legs. The loop temperature is controlled at the top of the hot leg. The salt flow rate is typically ~2 cm/s.

The TCL will be instrumented with pairs of launch piezoelectric transducers (in cooler zone), and fiber optic receivers (in hotter zone) for the tests in order to measure guided ultrasound propagation time, thereby group velocity of a select guided mode. Velocity changes can thus be measured on-line to indicate changes in material properties. The guided ultrasound responses of the MSR-specific alloy conduits, when measured at several locations with fiber optic sensors for pre- and post- corrosion, could then pinpoint locations of emerging structural weaknesses in the conduits. Diagnostics from these measurements in high-temperature MSRs would be helpful in the selection of (i) less corrosion-prone alloys and/or (ii) novel molten salt preparations that curtail corrosion.

ULTRASOUND TRANSDUCERS

This ultrasound detection system for structural corrosion monitoring is based on:

- piezo-electric launch transducers placement far enough from hot surfaces so as to avoid depolarization due to excessive heating
- fused silica optical fiber's sufficiently high softening point of 1585 °C that permits detecting ultrasound at MSR temperatures to 700 °C as well as transmitting it to read-out instrumentation far enough from extreme temperature and radiation zone;

specifically, optical fiber Bragg gratings (FBGs) for ultrasound sensing are mature enough and type II femtosecond FBGs can survive to beyond 1200 °C [13]. While piezoelectric transducers will launch the selected guided ultrasound wave mode, it can be detected at multiple locations along the conduit with fused silica fiber optic Bragg grating receivers retrofitted on MSR conduits surfaces to monitor material changes.

These system transducer components are described as follows:

(i) Launch Piezoelectric Transducers

One objective of the project is to develop dedicated hardware for launching guided ultrasound in candidate CRA tubes at MSR temperatures of 500 to 700 °C using passive rod waveguides and active transducers. The ultrasound will be generated by piezoelectric elements and propagate through the passive waveguide to the TCL tubing where it will be detected by fiber-optic sensors.

The first decision is where to place the actuators and receivers on the TCL. If the actuator is located anywhere on the TCL itself, it will have to operate in the 500 to 700 °C range, which is far too hot for commercial piezoelectric transducers. One alternative is to extend a vertical leg of the TCL far enough from the hot section of the TCL such that the tube temperature has cooled to below approximately 200 °C. If this can be accomplished, then piezoelectric transducers tolerant to service temperatures of 200 °C could be mounted on a wedge to function as an angle-beam transducer and designed by Snell's law. The wedge material could be PEEK. In this alternative the actuator would be installed on tubing where the temperature does not exceed 200 °C, and it would generate guided waves that propagate down the tube, past two T-joints and two thermowells into the hot zone of the TCL where the largest material degradation is expected to occur during operation. After propagating some distance in the TCL the guided waves will be received by the fiber-optic sensors.

In order to minimize the number of propagating wave modes (so that signal processing is not overly complicated) and to maximize the distance that the ultrasound will travel, the excitation frequency of the launched guided wave will be kept below the cutoff frequencies of higher order modes. While both longitudinal fundamental wave modes in a tube, L(0,1) and L(0,2) can propagate, the L(0,2) mode has been selected. The dispersion curves for longitudinal waves in a CRA tube are shown in Figure 3. The wave structures are not shown, but the frequency-thickness product (*fd*) of 2 MHz-mm the L(0,1) mode, shown below, has a large radial displacement component at the pipe surface, which means that a significant amount of energy will leak into the fluid inside the tube. On the other hand, the L(0,2) is dominated by the longitudinal displacement component and the radial displacement *at the surface is small*, meaning that not much energy will leak into the fluid inside the tube. Moreover, the optical fiber Bragg grating will be sensitive to the L(0,2) longitudinal displacement on the surface. This is another good reason to select the L(0,2) mode for this application.

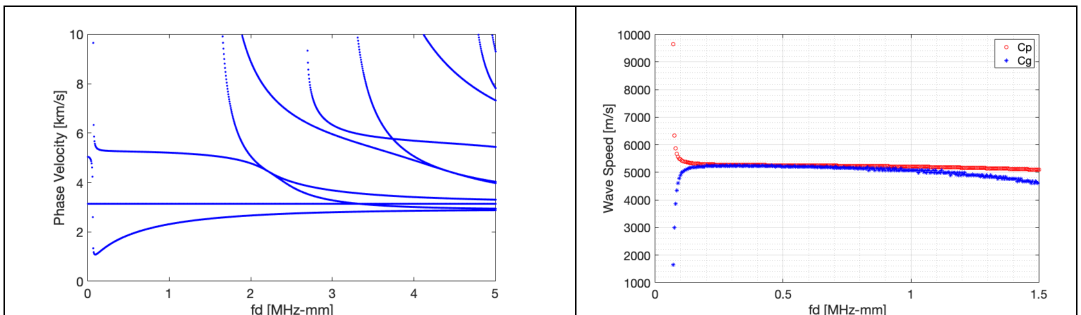

Figure 3. Phase velocity dispersion curves for axisymmetric modes in CRA tube (top) and zoomed in view of phase and group speeds for the L(0,2) mode. Alloy 600 tube parameters: $D_o = 25.4$ mm, $\rho = 8420$ kg/m^3, $c_L = 5770$ m/s, $c_T = 3138$ m/s. The frequency-thickness product is *fd*.

(ii) Fiber Optical Bragg Grating Receivers

In recent years, fiber Bragg grating (FBG) sensors have become popular for process monitoring, especially for temperature, strain, and acoustic emission. Fiber sensors require no electric power at the sensor itself or along the transmission fiber cable, only at its farthest end. FBG ultrasonic measuring applications [14], include structural health monitoring and precision medical therapy monitoring devices, and readout systems can maintain sensitivity under variable strain and temperature environments. The high temperature and radiation resistance of FBGs that renders them excellent candidates for this study are summarized in Table I.

TABLE I: EFFECTS ON FIBERS FOR MSR APPLICATIONS

FBG Parameter	**Effects on Fibers for MSR Application-Specific Sensing**
Radiation Resistance	As optical fiber refractive index changes under gamma radiation, in Ge-doped optical fiber there is an expected wavelength shift of the peak FBG wavelength, or ***Bragg wavelength*** $\boldsymbol{\lambda_B}$. There is also increased loss over the entire telecommunication band. For high frequency ultrasound (> 20 kHz) detection, under the quasi-static slow varying λ_B spectral shifts, readout systems with high fidelity are reported. The spectral shift saturates after an initial exposure [15, 16]**,** and the radiation has not been observed to change the reflected / transmitted spectral shape. ***The*** $\boldsymbol{\lambda_B}$ ***change as a result of irradiation is < 25 pm and saturates at a total dose of 0.1 MGy.*** Lowest radiation induced losses of fused silica fibers are in the 1530 to 1550 nm spectral range, at which standard telecom components are prolific. By selecting ultrasound-detecting FBGs in this spectral regime, losses can be minimized. Furthermore, pre-annealing of silica optical fiber of "type-II" **femtosecond-FBGs** reduces this increase in loss under radiation (see Figure 5).
High Tempera-ture Resistance	a) Fused silica, the primary constituent of telecommunication optical fiber and of ultrasound-detecting FBG sensors, has a melting point in the range 1710 to 1900 °C; b) Standard type-I FBGs inscribed in Ge-doped SMF-28 fiber using UV exposure do not survive beyond 500 °C. An alternate type-II FBG inscribed using femtosecond laser ablation with an 800 nm laser, survives beyond 1200 °C, and is a candidate for high temperature MSR structural monitoring [13, 17, 18]. c) Lastly, a limiting factor of standard telecom fiber is acrylate polymer coating, which does not survive beyond approximately 250 °C. Commercial carbon- and gold-coated fiber optic FBGs are rated to over 750 °C, which are being tested for this MSR material characterization.

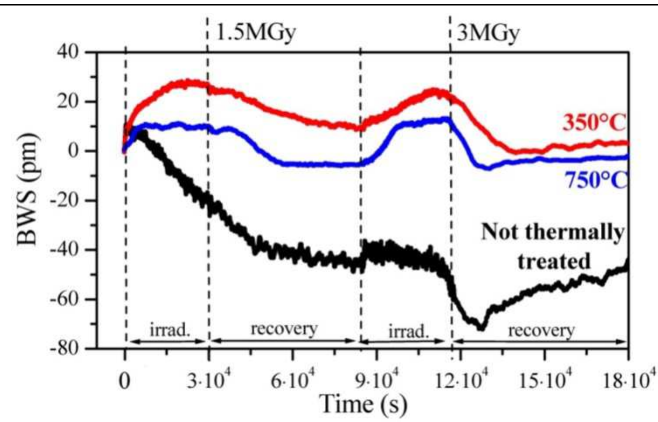

Figure 5: FBG wavelength shift under gamma-radiation with annealed and un-annealed fiber [15]

For this large temperature range (to 750 C), thermal effects on the FBG (thermal and mechanical effects of) include: (i) the MSR alloy's thermal expansion due to mismatch with fused silica, as well as (ii) the thermo-optic fused silica effect, and shall evolve as temperature increases at different locations on the TCL. This evolution can be tracked and compensated to maintain sensitivity with reliable tunable laser based optoelectronic control hardware as in other guided ultrasound monitoring applications [14].

SUMMARY

The system considerations are outlined based on the ultrasound launch and receiver transducers' extreme temperature operation and the preferred guided wave mode for measuring molten salt corrosion effects on the TCL conduit. Wave speed changes will be measured and interpreted using the L(0,2) guided mode dispersion model, and by comparing pristine vs. corroded sample data. Experiment progress and results will be reported shortly. Short duration runs of tens of hours are planned initially, followed by longer, > 100 hour, runs, as the reliability of transducers over maximum time periods needs to be ascertained. Concurrently, analytic methods to correlate corrosion extent to changes in wave speeds are also under development.

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