Containment materials for liquid tin at 1350 °C as a heat transfer fluid for high temperature concentrated solar power

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Abstract

One pathway for reducing the cost of concentrated solar power (CSP) is to increase the system efficiency by operating a heat engine with a higher hot side (inlet) temperature. If a turbine is used, then a system utilizing a combined cycle could potentially reach upwards of ~60% efficiency, provided that the turbine could be operated with an inlet temperature > 1300 °C. Such high temperatures place severe limits on heat transfer fluids; that is, such fluids would need to remain chemically stable, and be compatible with containment materials, at such extreme temperatures. One potential class of such fluids are liquid metals, such as molten tin. While possessing low melting and high boiling points (232 °C and 2600 °C, respectively) for a high operational range, molten tin also tends to be highly corrosive towards common structural metal alloys used as components for the containment and controlled flow of liquids (i.e., for pipes, tanks, valves, pumps, etc.). Thus, it would be useful to identify materials that are compatible with molten tin at ≥1300 °C. The purpose of this paper is to evaluate three candidate high-temperature materials, possessing a range of thermal conductivities, for the containment of molten tin: graphite (C), silicon carbide (SiC), and mullite (Al6Si2O13). The corrosion and penetration of these materials by molten Sn or doped Sn liquids at 1350 °C for 100 h were evaluated via local electron microscopic analyses and global weight change measurements. Under appropriate conditions, all three of these materials exhibited minimal to no reaction with tin-based liquids at 1350 °C and were not penetrated by these liquids at this temperature. This work indicates that graphite, silicon carbide, and/or mullite can serve as effective containment materials for the use of tin-based liquids as heat transfer fluids operating at 1350 °C in CSP plants.

1. Introduction

The cost of concentrated solar power (CSP) with thermal energy storage (TES) has been estimated to be 13.5–20 €/kWh (Hernandez-Moro and Martinez-Duart, 2013; Margolis et al., 2012; Kolb et al., 2011). While current flat plate photovoltaic solar technologies have a lower levelized cost of electricity (LCOE), their output is not dispatchable. If electrical batteries are added to such photovoltaic systems to provide dispatchability, then the combined cost can be greater than for CSP/TES systems (Dunn et al., 2011; Rastler, 2010). While CSP/TES systems can be cost-effective options for dispatchable solar power, direct competition with fossil fuel-based power generation requires that the CSP/TES LCOE (i.e., for state-of-the-art power tower configurations with molten salt TES) be further reduced by > 50% (U.S. Energy Administration Information, 2015; Kolb et al., 2011).

The LCOE for CSP may be significantly reduced by increasing the overall system efficiency via use of more efficient heat engines. However, current turbomachinery-based heat engines already operate near their thermodynamic limits (Henry and Prasher, 2014). Thus, one of the only strategies for major increases in turbine efficiency would be to operate with a much higher turbine inlet temperature (TTI) (Henry and Prasher, 2014). It should be noted that several prior system analyses have indicated that operating at higher temperatures is of critical importance for realizing major LCOE reductions (Margolis et al., 2012; Melhos et al., 2016). However, depending upon the peak temperature used, there could be other efficiency trade-offs in the optics, receiver, or storage. Nonetheless, it is worthwhile to identify potential materials that could allow for a wider range of peak CSP operational temperatures.

The highest-efficiency, long-life heat engine systems utilize combined...
cycles and would require TITs in the range of 1300–1500 °C to achieve efficiencies up to ~60% (Huck et al., 2016; Brooks, 2000). An increase in heat-to-electricity conversion efficiency from 35 to 40% (current heat engines) to 60% corresponds to a 50–71% increase in relative efficiency. If all other plant costs remained fixed, then such an enhancement in relative efficiency would result in a cost reduction of 33–42%, which would be a major step towards parity with fossil fuel-derived electricity. Hence, approaches enabling the operation of CSP systems at such high temperatures should be considered.

A variety of liquids (e.g., molten salts, glasses, metals) have been considered as high-temperature heat transfer agents (Elkin et al., 2014; Pacio and Wetzel, 2013; Forsberg et al., 2006; Misra and Whittenberger, 1987). Liquid metals can be particularly attractive, due to their relatively high values of thermal conductivity (usually ~10–100 W m⁻¹ K⁻¹), low values of viscosity (typically ~1 cP), and (for certain metals) relatively high ranges of thermal stability (i.e., low melting points and high boiling points) (Pacio and Wetzel, 2013; Sobolev, 2012, 2010; Ghoshal et al., 2005; Regel et al., 1971). When considering the parasitic loads associated with pumping various heat transfer liquids at the MW scale, these attractive characteristics of liquid metals can lead to notable system level performance advantages and, ultimately, cost benefits.

The use of liquid metals as heat transfer fluids has been recently reviewed for nuclear and CSP systems (Pacio and Wetzel, 2013; Pacio et al., 2014). Among potential liquid metals for heat transfer are group 13 and 14 elements, such as Al, Sn, and Pb. These metals have low melting points (660 °C, 232 °C, and 328 °C, respectively) and high boiling points (2518 °C, 2600 °C, and 1746 °C, respectively) for wide operational temperature ranges (Barin, 1995). Molten Pb is unattractive from a health and environmental perspective (Goyer, 1990), and operational temperature ranges (Barin, 1995). Molten Sn and Al are quite corrosive towards Ni-based and Fe-based alloys (and numerous other metal alloys) at elevated temperatures (Emmerich and Schroer, 2017; Heinzel et al., 2017; Dybkov, 1990; Reed, 1954). Because molten aluminum has a higher affinity for oxygen, nitrogen, and carbon than does molten tin (Pietzka and Schuster, 1997; Barin, 1995; Oden and Gokcen, 1993), molten tin is expected to be generally less reactive than molten aluminum with refractory oxides, nitrides, carbides, and carbon. Nonetheless, experimental evaluation of the chemical compatibility of molten Sn with potential high-temperature refractory containment materials at >1300 °C is needed.

The aims of this paper are therefore: (i) to assess the relative thermochemical and thermophysical properties of three candidate commercially-available refractory materials for the containment of tin-based liquids at 1350 °C graphite (C), silicon carbide (SiC), and mullite (Al₆Si₂O₁₃); (ii) to evaluate the resistances of graphite, silicon carbide, and mullite to chemical interaction with, and penetration by, various grades (purities) of molten tin at 1350 °C; and (iii) to identify conditions under which such chemical interaction/penetration can be avoided or reduced.

2. thermochemical and thermophysical properties of graphite, silicon carbide, and mullite at 1350 °C

Graphite, silicon carbide, and mullite were selected for evaluation as candidate high-temperature containment materials for tin-based liquids at 1350 °C (the temperature selected for experimental work in this paper) after considering the following characteristics:

- thermal stability
- chemical compatibility with molten tin
- wetting and penetration by molten tin
- resistance to oxidation or reduction
- thermal conductivity
- thermal expansion
- high-temperature stiffness and creep resistance

- commercial availability and relative cost

Regarding thermal stability, graphite undergoes sublimation at 1 atm above 3600 °C (Joseph et al., 2002; Barin, 1995; Leider et al., 1973). Silicon carbide melts incongruently to form solid carbon and a silicon-rich liquid at ≥2540 °C (Kleykamp and Schumacher, 1993; Oleinski and Abbaschian, 1984b; Scace and Slack, 1959). Mullite is also reported to melt incongruently, to yield a mixture of solid alumina and a silica-alumina liquid, at 1890 °C (Klug et al., 1987; Horibe and Kuwabara, 1967).

Regarding chemical compatibility with liquid tin, graphite is not reported to form a stable carbide by reaction with tin at ambient pressure (Pietzka and Schuster, 1997) and the solubility of graphite in pure liquid tin is low. Extrapolation of the data of Oden and Gokcen (1993) from 1550 °C–2300 °C down to 1350 °C yields a carbon solubility in liquid tin (99.999% purity) of only 0.34 ppm at 1350 °C. The Si-C-Sn ternary phase diagram, and the solubility of SiC in liquid Sn at 1350 °C, do not appear to have been evaluated. The chemical interaction of mullite (Al₆Si₂O₁₃) with liquid tin at 1350 °C also does not appear to have been reported. As for other oxide/liquid systems (Kumar, 1999; Sandhage and Yurek, 1991, 1990b, 1990a, 1988), the incongruent reaction of mullite (Al₆Si₂O₁₃) with pure liquid tin at 1350 °C via the following reactions may be considered:

2Sn(l) + Al₂SiO₃(s) = 2SnO₂(s) + 2[Si] + 3Al₂O₃(s)  
4.5Sn(l) + Al₂SiO₃(s) = 4.5SnO₂(s) + 6[Al] + 2SiO₂(s)

where (Si) and (Al) refer to silicon and aluminum dissolved in molten tin, respectively. However, thermodynamic analyses (see Supplemental Information) indicate that the extent of decomposition of mullite by incongruent reduction reactions (1) and (2) at 1350 °C should be negligible. While it is expected that graphite, silicon carbide, and mullite should exhibit good chemical compatibility with high-purity liquid tin at 1350 °C, the chemical compatibility of these potential containment materials with lower purity, commercially-available liquid tin is unclear.

Regarding liquid tin wetting and penetration, pure molten tin is reported to exhibit poor wetting of graphite at up to 1200 °C. The contact angles of high-purity (≥99.99%) tin liquid on dense (≥7% porosity) graphite in high-purity (99.999%) argon or reducing (5% H₂/Ar) atmospheres at 300–1200 °C have been reported to be ≥122 degrees (Weltisch et al., 2013; Sanchez et al., 2008). The contact angles reported for high-purity (≥99.99%) tin liquid on dense SiC in argon (≥99.9% purity, 1 atmosphere) or vacuum (10⁻⁸ atmospheres) at 800–1400 °C were ≥105 degrees (Liu et al., 2010; Tsoga et al., 1997; Takahashi and Kuboi, 1996; Nikolopoulos et al., 1992). These values were similar to measured contact angles of liquid tin on silica (Harding and Rossington, 1970), which was consistent with the presence of a silica scale on the silicon carbide surface. Non-wetting behavior (contact angles greater than 90°) has also been reported for molten tin at 600–1400 °C on mullite-based refractories (Ignatova et al., 1971). While this data indicates that pure liquid tin should not wet, or exhibit appreciable penetration into, porous graphite, silicon carbide, or mullite, the influence of impurities in various commercial grades of tin on such wetting/penetration at 1350 °C remains to be examined.

Regarding oxidation or reduction by reaction with an external gas environment, graphite and silicon carbide are stable in sufficiently reducing atmospheres, whereas mullite is stable in a sufficiently oxidizing atmosphere. At 1350 °C and 1 atm total pressure, graphite can coexist at equilibrium with an O₂-CO₂ gas mixture possessing an oxygen partial pressure, pO₂, of 4.3 × 10⁻¹⁷ atm (Barin, 1995). At lower oxygen partial pressures (i.e., higher pCO and lower pCO₂ values), graphite should not oxidize at 1350 °C and 1 atm. At 1350 °C and 1 atm, SiC can coexist with graphite and an O₂-SiO₂-CO₂ gas mixture possessing a pO₂ of 4.3 × 10⁻¹⁷ atm and a carbon dioxide partial pressure, pCO₂, of 2.4 × 10⁻⁷ atm (Barin, 1995). At lower partial pressures of
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