**Motivation**

- Solid Oxide Fuel Cells are a desirable clean energy conversion technology, due to their fuel flexibility, high efficiency, and relatively low cost. [1]
- The standard 97.5Ag-2.5CuO braze often suffers from pore formation caused by poor wetting, CuO reduction, and/or the chemical reaction of oxygen and hydrogen within the braze to form water. [2,3]
- The Ag-Cu braze fails due to pore formation during long term application of more than 10,000 hours. [4]
- Methodology is developed to computationally search for durable, impermeable, silver-free brazes for SOFCs operating at 750°C.

**Selection of Alloying Elements and Thermodynamic Study**

- List of all potential brazing alloy elements were selected from periodic table, based on braze manufacturing and operating conditions.
- A binary and ternary composition search was performed within the list of base alloy elements, based on CALPHAD approach implemented in Thermo-Calc® software.
- Compositions were selected which met the required liquidus (1000°C) and solidus (900°C) temperature for brazing.
- Predicted alloys were searched in commercial alloy databases, if they were commercially available.

**The CALPHAD Method**

- Computational prediction of multicomponent material property requires thermodynamics and kinetics data. A thermodynamic modeling technique, widely known as the CALculation of PHAse Diagrams (CALPHAD) method is implemented for design of material performances. [5]
- The CALPHAD methodology is to evaluate the change in Gibbs free energy in the multidimensional space of temperature, pressure, and compositions, and integrate them into multicomponent systems. [6]

**Predicted Alloys**

<table>
<thead>
<tr>
<th>Alloy System</th>
<th>Composition (mol %)</th>
<th>Phase composition at 750°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-Si-Cr</td>
<td>Ni: 40 – 50, Si: 45 – 59, Cr: 0 – 6</td>
<td>NiSi + (Ni,Cr)2Si + (Cr3Si)2Si + NiSi</td>
</tr>
<tr>
<td>Ni-Si-Mn</td>
<td>Ni: 34 – 53, Si: 43 – 58, Mn: 0 – 11</td>
<td>FCC(γ) + Ni3Si + Ni3(Si)</td>
</tr>
<tr>
<td>Ni-Si-Ta</td>
<td>Ni: 38 – 68, Si: 14 – 59, Ta: 0 – 34</td>
<td>Ni3Si + Ni3Si + C14_Laves, SiTa + Ni3Si + Ni5S2</td>
</tr>
<tr>
<td>Ni-Si-Mn-Ta</td>
<td>Ni: 40 – 50, Si: 35 – 60, Mn: 0 – 26, Ta: 0 – 11</td>
<td>Ni3Si + M23Si + Ni5Si3 + Ni5S2</td>
</tr>
<tr>
<td>Ni-Mn-Nb</td>
<td>Ni: 27 – 54, Mn: 35 – 65, Nb: 4 – 19</td>
<td>FCC(γ') + FCC(γ), C14_Laves</td>
</tr>
<tr>
<td>Ni-Mn-Ta</td>
<td>Ni: 28 – 47, Mn: 44 – 66, Ta: 3 – 7</td>
<td>FCC(γ') + Ni3Ta6 + Ni3Ta</td>
</tr>
</tbody>
</table>

**Phase diagrams at Required Liquidus & Solidus Temperature**

**Ab initio study of ductility**

- Density Functional Theory based elastic property calculations were performed to find ductility of the predicted alloys.
- Pugh Criterion: if G/B < 0.57 material is ductile, else brittle. [7]

**Conclusion**

- A Ni-Mn and Ni-Si-containing new brazes alloy systems were predicted based on CALPHAD approach and Density Functional Theory based mechanical property predictions.
- Due to high silicon content predicted Ni-Si alloys are brittle. [8]
- Ni-Mn based alloys are suggested to develop experimentally, in the laboratory.
- The study will be extended for four or more elements containing alloys, if required to fine tune the alloy property using Thermo-Calc.