Investigations of Thermite-Type Chemical Reactions to Generate Molten Core Materials for Reactor-Safety Experiments


FAST REACTOR SAFETY X: POST-ACCIDENT HEAT REMOVAL

To perform reactor safety experiments, one way to obtain mixtures that have the approximate temperatures and compositions of material produced during hypothetical core-disruptive accidents of LMFBR cores, in which mixed-oxide fuel (U, Pu)O₂ is in close contact with stainless steel (Fe, Cr, Ni, Mn), is by thermite-type reactions.

Thermite reactions involve a reactive metal and an oxide of a less reactive metal. A reaction between uranium metal powder and molybdenum trioxide to produce molten UO₂,

\[ \frac{3}{2} U + \text{MoO}_3 \rightarrow \frac{3}{2} \text{UO}_2 + \text{Mo} \]  

was used successfully in recent studies of molten UO₂ interactions with sodium. However, the UO₂ generated by Eq. (1) contains about 19 wt% molybdenum metal (not normally present in a reactor core).

Studies of other thermite reactions, which might better simulate core debris, were begun for a wide range of reactor safety experiments by producing molten stainless steel and molten steel-UO₂ mixtures. Three reactive metals were studied: Al, Zr, and U. Both pure Fe₂O₃ and a mixture of stainless-steel oxides (see Table I) were used. Approximately 75 g of oxide powders (reagent grade) were mixed with the metal powders in the desired proportions and packed into a reaction chamber made of a 6-in. length of 1-in. stainless-steel pipe (closed at the bottom with a cap that had a ½-in. central hole covered with a 3-mil stainless-steel sheet). The powder mixture was ignited using Mg ribbon and the thermite reaction propagated from top to bottom. On reaching the bottom, the steel sheet melted and the mixture, if molten, jumped into a graphite crucible located about 6 in. below the reaction chamber.

The propagation rate was measured by timing the initial rapid rises of two thermocouples located vertically 4 in. apart in the chamber. Brightness temperatures were measured by taking high-speed motion pictures of the falling molten mixtures, using several calibrated tungsten ribbon-filament lamps.

Results of the experiments are given in Table I. Propagation rates and, to a lesser extent, temperatures may depend on many factors, including particle size, packing density, etc., which were not varied in these tests. The data should be viewed, therefore, as only typical of what might be expected in an application.

The tests with aluminum gave the slowest propagation rates; however, because of the large density difference between the molten metal and alumina, the metal was effectively separated in a lower layer with the oxide on top. The tests with zirconium gave faster propagation rates and higher temperatures; however, the product was a coarse mixture owing to a decreased difference in densities. The tests with uranium gave intermediate propagation rates; however, the temperature was not high enough to melt UO₂ (MP 2840°C).

In agreement with the experimental results, thermodynamic calculations indicate that all of the reactions in Table I have enough energy to produce boiling steel (BP ≈ 2800°C), although the energy available for the uranium reaction is somewhat marginal.

Reaction (3) is well suited to produce good-quality molten steel and in two tests 1 kg was produced to observe flow and freezing of molten steel in tubes. Reaction (7) was used to simulate a hot but solid fuel rod to observe the meltdown of steel cladding. Mixtures of Reaction (1) and (7) or possibly (1), (3), and (7) may be capable of generating very representative mixtures of molten core debris. Such tests are planned.


**TABLE I**

Summary of Thermite Reaction Experiments

<table>
<thead>
<tr>
<th>No.</th>
<th>Thermite Mixture</th>
<th>Product Composition</th>
<th>Brightness Temperature, °C</th>
<th>Linear Propagation Rate, in./sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>Al-Fe₂O₃</td>
<td>48 wt% Al₂O₃, 52 wt% Fe</td>
<td>2750</td>
<td>0.2</td>
</tr>
<tr>
<td>3</td>
<td>Al-SS oxides</td>
<td>52 wt% Al₂O₃, 48 wt% SS</td>
<td>2850</td>
<td>0.2</td>
</tr>
<tr>
<td>4</td>
<td>Zr-Fe₂O₃</td>
<td>62 wt% ZrO₂, 38 wt% Fe</td>
<td>2800</td>
<td>0.9</td>
</tr>
<tr>
<td>5</td>
<td>Zr-SS oxides</td>
<td>66 wt% ZrO₂, 34 wt% SS</td>
<td>3000</td>
<td>1.3</td>
</tr>
<tr>
<td>6</td>
<td>U-Fe₂O₃</td>
<td>78 wt% UO₂, 22 wt% Fe</td>
<td>3000</td>
<td>0.6</td>
</tr>
<tr>
<td>7</td>
<td>U-SS oxides</td>
<td>81 wt% UO₂, 19 wt% Sh</td>
<td>3000</td>
<td>0.5</td>
</tr>
</tbody>
</table>

* Eight oxides indicate a mixture of 61.5 wt% Fe₂O₃, 25.5 wt% Cr₂O₃, 10.9 wt% NiO, and 2.1 wt% MnO₂.
* UO₂ was not melted and mixture did not flow.
* UO₂ was slightly melted and flow was incomplete.