

ANTIMONY TELLURIDE FORMATION HYPOTHESIZED FROM REACTOR COOLANT SYSTEM SAMPLE DATA

Recent assessments have been reported from special committees formulated by the American Nuclear Society,¹ the American Physical Society,² and a draft document undergoing U.S. Nuclear Regulatory Commission staff review,³ on their review and evaluation of the state of knowledge of how to predict a source term deriving from a severe core damage accident. These assessments were relevant and reflect the important data base information provided by the Three Mile Island Unit 2 (TMI-2) sample data.

One of the technical areas of identified uncertainty involves the behavior of the fission product tellurium. The numerous samples and analyses from TMI-2 have not provided significant information on tellurium. Furthermore, laboratory data and theoretical considerations⁴⁻⁶ have only considered the chemical interaction of tellurium with the principal metallic surfaces of components within the primary reactor vessel, i.e., stainless steel and Zircaloy cladding.

Initial material balances⁷ did not include either tellurium or antimony since sample data were unavailable for incorporation into balance estimates. The interest expressed by technical review groups and the increasing data base provided by TMI-2 sample analyses and cleanup prompted our recent review of these two fission products. There is indirect evidence from TMI-2 postaccident reactor coolant chemistry data that suggests the possibility of their chemical interaction during the accident.

The TMI-2 accident major events (i.e., loss of coolant, subsequent core degradation, and system stabilization) started during the early morning hours of March 28, 1979, and lasted for ~16 h. Reactor coolant system (RCS) samples were taken shortly after the accident on March 29, 1979, and later on April 12, 1979. These sample results indicated, among other pertinent data, that radioactive isotopes of tellurium were detected but radioactive isotopes of antimony were conspicuously absent. The short half-life of radioactive tellurium isotopes (i.e., ¹³²Te, 78.2 h) and the unavailability of additional early samples minimized this radionuclide from serious consideration for source term assessment. However, the longer lived antimony radionuclide (¹²⁵Sb, whose half-life is ~2.73 yr) was not routinely detected until after the TMI-2 RCS was opened to atmospheric conditions and began oxygen saturation. Prior to this the RCS had been purposely kept at <1-ppm dissolved oxygen content.

Once the RCS had been opened to atmospheric conditions, two phenomena were noted and reported.⁸ These phenomena involved the rapid accumulation rate for cesium radionuclides in the coolant for a short time period followed by a lower long-term accumulation rate and the appearance of ¹²⁵Sb for the first time.⁵ While both phenomena were attributed to the increasing oxygen chemical potential of the RCS, corresponding experimental data were unavailable to verify them. These coincident observations have led to the following hypothesis regarding the antimony/tellurium source term and suggest corresponding sample analyses to ascertain the consistency of this hypothesis.

The general chemistry for both antimony and tellurium at equilibrium in a light water reactor fuel rod during operation prior to accident conditions indicates that the majority of both would be released in elemental form; hence, they would be available for chemical reaction. The thermody-

amic stabilities⁹ indicate that chemical interaction for antimony telluride compound formation is favorable. The thermodynamics for steam oxidation of antimony telluride is unfavorable. Consequently, antimony telluride could be formed and with its relatively high specific gravity (6.5) it could have potentially migrated toward the lower reactor vessel region. The shutdown inventory mass of tellurium at TMI-2 is ~20 times larger than the inventory mass of antimony. If antimony telluride were formed, then the RCS solution chemistry of antimony would be initially dictated by the insolubility of antimony telluride. However, as the ratio of ¹²⁵Sb/¹³²Te that was formed increases, the antimony should become oxidized as the tellurium decays away. The Sb(III) oxide is only slightly soluble; however, Sb(V) antimonate anion is quite soluble. The increased dissolved oxygen caused by opening the RCS to atmospheric conditions could have increased the oxidation state of Sb(III) oxide to the Sb(V) antimonate anion, Sb(OH)₆⁻, which is thermodynamically preferred under basic conditions, i.e., pH = 7.6. Removal of ¹²⁵Sb from the RCS coolant has confirmed its anionic character.

Therefore, in reviewing the initial RCS sample data for these two elements, only tellurium was reported.¹⁰ This observation could be consistent with the hypothesis that essentially all of the antimony reacted with the tellurium and remained insoluble in the reactor vessel, whereas the excess tellurium would be soluble in the RCS liquid and was detected. Subsequently, with the decay of the tellurium and introduction of increased dissolved oxygen after atmospheric conditions had been established for the RCS, the antimony was oxidized to the soluble antimonate anion that was then detected in RCS samples. This hypothesis can neither be experimentally verified at TMI-2 nor separated from the other antimony sources (i.e., fuel). However, the hypothesis suggests the following:

1. The extensive research efforts (experimental and theoretical) being conducted on source terms should include antimony telluride evaluations.
2. Core debris samples from the reactor vessel lower region should be analyzed for antimony concentrations (grams of antimony/grams of UO₂) to ascertain if this ratio is significantly greater than the calculated burnup value.
3. Nuclear reactor safety tests should define as one of their experimental analysis goals to obtain samples and analyze them for this compound shortly after any loss of coolant test in order to ascertain if antimony telluride is pertinent to the source term evaluations.

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