LA-UR-95 · 4076

CONF-960804--2

Title: Dynamic Characteristics of Mixtures of Plutonium, Nevada Tuff, and Water

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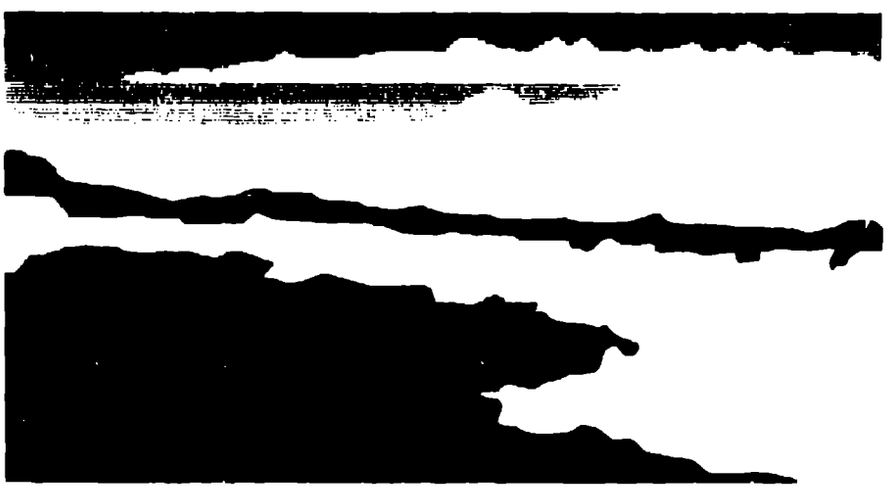
Submitted to: SPECTRUM 96

Seattle, WA
August 1996



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Dynamic Characteristics of Mixtures of Plutonium, Nevada Tuff, and Water

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Introduction

One of the technical options being considered for long term disposition of weapons grade plutonium is geologic storage at Yucca Mountain. Multikilogram quantities of plutonium [50-100 kg] are to be vitrified, placed within a heavy steel container, and buried in the material known as Nevada tuff. It has been postulated [1]-[3] that after about ten or twenty thousand years, geologic and chemical processes would have disintegrated the steel container and created the possibility for plutonium to form mixtures with Nevada tuff and water that could lead to a nuclear explosion in the "range of kilotons" or "hundreds of tons TNT equivalent". Sanchez et. al. [4] completed a survey and description of critical homogeneous mixtures of plutonium, silicon dioxide, Nevada tuff, and water and identified the mixture regimes where autocatalytic dynamic behavior is possible. This study is a follow up of that work and the major objective is to examine the dynamic behavior of the "worst case" critical and supercritical configurations of plutonium, Nevada tuff, and water. The indicators for a damaging power transient and the kinetic energy or "TNT equivalent" yield from an excursion for the "worst case" scenarios are examined.

This paper does not consider the details and probabilities of the geologic and chemical processes occurring that lead to the formation of the mixtures. The calculations are to serve as an upper limit of the consequences if such configurations were to be created.

Energy Generating Systems and Explosions

The study first examines some characteristics of explosions, both in chemical and fissioning systems and the accepted conversion factor between chemical explosions and fission energy release. The conventional conversion is total fission yield equals the equivalent chemical explosion yield, which is acceptable for large nuclear explosions, but fails completely for small or slow fission energy release. It is shown by examples, e.g. the Godiva I assembly [5]-[6], that a measure of a small explosion should be the kinetic energy created during the fission power transient, not the fission energy per se.

Conditions Necessary for an Explosion

The characteristics of fissioning systems that have the appearance of explosions are identified and comparisons are made with critical and supercritical configurations of plutonium, silicon dioxide, Nevada tuff, and water. These include the neutron spectrum, the initial alpha or reciprocal period, and a measure of the rate of propagation of pressure waves in the assembly. More importantly, a relationship defined as [7]:

$$F = \frac{\alpha_i r}{v_s}$$

where

α_i initial system alpha or reciprical power period

r characteristic assembly dimension

v_s speed of sound through assembly material

can be used to infer when assembly damage may occur. By comparison with the Godiva I assembly experiments, the initiation of damage occurs at around F = 3, but significant damage would not occur until values of F = 50 or greater. For the "worst case" configurations of plutonium, silicon dioxide, Nevada tuff, and water identified by Sanchez et al. [4], values of "F" ranged from 0.2 to 0.45, to small to suggest assembly damage.

Computer Program for Calculation of Energy Release

Lastly, a dynamic energy release computer program, created for the purpose of evaluating supercritical transient reactors, criticality incidents, and criticality accidents, is used to analyze excursions with critical and supercritical configurations of plutonium, silicon dioxide , Nevada tuff, and water.

The computer program created for this study to evaluate the dynamic energy release during an excursion was dubbed the MRKJ Reactor Transient Code. The MRKJ Reactor Transient Code is to serve as a modern replacement for the Los Alamos Pajarito Dynamics code [8]. The MRKJ code uses the calculational technique known generally as the "coupled neutronic-hydrodynamic method" [9] which combines together the differential equations for thermodynamics, materials motion, and neutron transport with rapidly changing reactivity to model a transient nuclear system. The MRKJ code is written in FORTRAN 77 and utilizes the one-dimensional discrete ordinance transport code ONEDANT [10] to perform the neutron transport calculations. The MRKJ code uses the time absorption calculation (alpha) option of the ONEDANT code and determines the power distribution of the system by extracting the regional fission rates from the ONEDANT output files. The library of cross sections used in the ONEDANT calculations is a Hansen-Roach 16 energy group cross section set [11] with 167 isotopes.

The MRKJ code solves for the thermodynamic and hydrodynamic state variables by treating the analysis as an initial value problem. For the kinds of transient systems that we are currently studying, the thermodynamic model assumes no heat transfer occurs between adjacent coarse mesh regions. This assumption is based on the fact that the rate of energy deposition due to fission is much, much larger than the rate of heat conduction between regions because the characteristic time scale for energy deposition from local fissions is much shorter than the characteristic time scale for heat conduction between regions.

The equations are written as a system of first order ordinary differential equations and solved simultaneously using a fourth order Runge-Kutta [12] integration scheme. The equations are defined as follows:

There is an equation that solves for the power of the system as a function of time:

$$\frac{dP_{power}}{dt} = \left[\alpha_{displ} + \alpha_{Temp}\right] P_{power}$$

where

α_{displ} displacement feedback

α_{temp} temperature dependent feedback (doppler broadening of cross sections)

At this time we are neglecting the contributions of delayed neutrons to the power level because the short time scales of the excursions being examined are much shorter than the delayed neutron precursor decay times.

There is an energy balance equation used to find the temperature of each region derived from the first law of thermodynamics. For a material in the condensed phase, this equation has the form

$$\frac{dT_i}{dt} = \frac{1}{m_i C_i}\left[P_{power_i} - \beta T_i \frac{dV_i}{dt}\right]$$

where

T_i temperature in region i

m_i mass in region i

C_i material specific heat at constant volume

P_{power_i} regional power

β material isobaric compressibility

κ = material isothermal compressibility

V_i = region i volume

An equation of state appropriate for the material is used to relate the pressure and temperature in each region. The equation of state used to model solid Nevada tuff is:

$$P_{cond_i} = \alpha B(T_i - T_{ref}) + \frac{B}{\rho_i}(\rho_i - \rho_{ref})$$

where

P_{cond_i} = condensed pressure in region i

T_i = region i material temperature (T_{ref} = reference temperature)

ρ_i = region i material nominal density (ρ_{ref} = reference density)

α = material coefficient of expansion

B = material bulk modulus

This equation of state is similar to the EOS model used for uranium metal in a godiva I excursion simulation [13] and is a function of the velocity of sound through the material by the relation.

$$B = \rho V_s^2$$

where

V_s = velocity of sound through the material

Rewriting this equation in a form that can be solved as an initial value problem we have:

$$\frac{dP_{cond_i}}{dt} = \alpha B \frac{dT_i}{dt} - \frac{B}{V_i}\frac{dV_i}{dt}$$

Equations of motion are used to determine the regional boundary movements due to pressure differences between adjacent regions. They have the form

$$\frac{dv_i}{dt} = \frac{1}{2}\left[P_{cond_i} - P_{cond_{i+1}}\right]\frac{A_i}{\left[m_i + m_{i+1}\right]}$$

and

$$\frac{dr_i}{dt} = v_i$$

where

v_i = region i outer boundary velocity

r_i = region i outer boundary radial position

A_i = region i outer boundary surface area

m_i = mass in region i

P_{cond_i} = condensed pressure in region i

The MRKJ code gives estimates of the total fission yield and the maximum kinetic energy yield of a transient. The temperature, pressure, boundary location, and boundary velocity are calculated for each coarse mesh region for each time step until a predetermined code termination criterion is met. The values of the kinetic energy yield and state variables help determine if significant damage occurs to an assembly during an excursion.

Application of MRKJ Code to Pu-Tuff-Water Configurations

The MRJK code was used to study the dynamics of three autocatalytic configurations of homogeneous mixtures consisting of plutonium, Nevada tuff, and water. These cases represented the "worst case" scenarios as identified by Sanchez et. al. [4]. Table 1.0 gives general descriptions of the initial core configurations.

Mass (kg Pu)	Core Radius (cm)	Weight Fraction of Water
63.8	150.11	0.09
108.3	180.50	0.10
404.7	282.64	0.112

Table 1.0 Critical Parameters for Homogeneous Spherical Mixtures of Plutonium, Tuff, and Water Reflected with Tuff and Water Mixtures

The silicon to Pu (239) ratio for all the cases was 1570 and the model assumes each core is reflected by 100 cm of a tuff and water mixture. The mixture in the reflector has the same weight fraction of water as the core. The water was assumed to disappear from the system once it was vaporized during boiling. Extremely conservative assumptions, such as beginning the excursions with impossibly high initial powers, were used with the MRKJ code with the result that no kinetic energy yields existed at the end of the calculations.

Conclusion

The paper examined the difference between total fission yield and kinetic energy in an assembly Characteristics from Godiva I assembly experiments were used to illustrate indications when damaging power transients would occur. A similar comparison with the characteristics of the plutonium, tuff, and water systems indicate that these systems do not meet the requirements for explosive energy release. The "worst case" autocatalytic plutonium, tuff, and water systems were analyzed using the MRKJ code with extremely conservative assumptions with the result that no kinetic energy yield existed at the end of the calculations. Critical configurations of plutonium, Nevada tuff, and water can exist. Autocatalytic behavior of such systems is possible, but no explosions would occur. Hence, should multikilogram quantities of plutonium [50-100 kg] are to be vitrified, placed within a heavy steel container, and buried in the material known as Nevada tuff, no explosion would occur even throughout geologic time.

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