MASTER

MAXIMUM ACCEPTABLE TEMPERATURES
OF WASTES AND CONTAINERS DURING
RETRIEVABLE GEOLOGIC STORAGE

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August 17, 1977

**UNION
CARBIDE**

OFFICE OF WASTE ISOLATION
OAK RIDGE, TENNESSEE

*prepared for the U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
under U.S. GOVERNMENT Contract W-7405 eng 26*

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by
G. H. Jenks

Abstract

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Estimates of maximum acceptable temperatures of waste and containers during retrievable geologic storage were needed for use in evaluating and comparing conceptual designs for repositories for wastes from reprocessing and for spent fuel in several different rock types. Estimates of these temperatures and discussions of the bases for the estimates are presented.

Introduction

Estimates of maximum acceptable temperatures of waste and containers during retrievable geologic storage are listed in Table 1. These were made for use in evaluating and comparing conceptual designs for repositories in several different rock types. General assumptions regarding the Office of Waste Isolation concept of retrievability, the length of the period of retrievability, and the container materials are discussed below. These formed partial bases for the estimated temperatures. More specific comments on the bases for the estimates for each of the waste types are given in the subsequent text as notes to Table 1.

Retrievability Concept

The wastes that are emplaced in a repository will be encapsulated in suitable metal canisters. These canisters must be readily retrievable for predesignated periods of time after emplacement. "Readily retrievable" means that a given canister can be removed and transported to surface facilities with approximately the same effort and time that were required for the emplacement. It also implies that the canister can be safely stored in appropriate surface facilities for several months after retrieval.

The canisters will be protected from contact with any vapors which might lead to failure of the containment during the period of retrievability. In order to achieve this protection, it may be necessary to use an overpack comprised of a more corrosion-resistant metal than that used in the canister. For example, if the corrosion environment is such that austenitic stainless steel high-level-waste canisters would undergo stress corrosion cracking, it would be necessary to use an overpack of a metal that does not crack in the environments, e.g., carbon steel in an aqueous chloride environment. On the other hand, if the waste form itself comprises an adequate containment in the environments, it would not be necessary to assure that the canister remains intact. For example, low-level TRU wastes that are incinerated and then made into a glass would probably be adequately protected by the glass alone.

Period of Retrievability

The assumed length of the retrievability period is five years for all wastes except spent fuel. The assumed period for the spent fuel is 25 years.

Container Material

Austenitic stainless steel, 304L, is the assumed container material for high-level waste, and either austenitic stainless steel or carbon steel is assumed for the other wastes.

Table 1. Maximum Acceptable Temperature of Waste and Container during Retrievable Geologic Storage

Item	Maximum acceptable temperature (°C)	Note number
High-level waste		
Canister	375	1
Solid waste		
Glass	500	2
Calcine	700	3
Sintered glass ceramic	800	4
Cladding and hardware		
Canister	375	1,5
Solid waste		
Compressed	200	6
Fused	400	7
Intermediate-level transuranic waste		
Canister	375	1,5
Solid waste		
Glass	500	2,5
Low-level transuranic waste		
Canister	375	5
Solid waste		
Glass	500	2,5
Cement	110	8
Spent fuel element	200	9
PuO ₂	500	5,10

Notes to Table 1

1. Austenitic stainless steel undergoes changes in structure during long-term exposures in air at temperatures in the range of 400 to 900°C. The observed effect is an increased susceptibility to stress cracking upon subsequent exposure to water at atmospheric temperatures.¹ Pacific Northwest Laboratories workers have stated¹ that precautions should be taken to maintain the canister wall temperatures below 400°C during handling and storage after filling, in order to avoid this sensitization. Argonne National Laboratory workers have quoted a maximum temperature of 343°C (650°F) for 100-year retrievable storage in air.² Atlantic Richfield Hanford Company workers³ have recently recommended maximum temperatures of 277°C (530°F) and 427°C (800°F), respectively, for normal and abnormal conditions in long-term (100-year) retrievable, sealed cask storage. The abnormal conditions could presumably prevail for an indefinite period. I have concluded that the limit of 375°C for 5-year retrievable geologic storage is reasonable, especially since the canister will probably be at the maximum temperature for not more than about 1 year.

Carbon steel is probably less susceptible to significant changes in structure and corrosion susceptibility during long-term heating at temperatures $\geq 400^\circ\text{C}$, but there is no information on this. Accordingly, I have specified the same temperature limit as that for stainless steel.

2. The transition temperature for typical borosilicate waste glasses is about 500°C.⁴ The softening temperature is a few degrees higher.⁴ Significant increases in cracking and in leach rates have been observed in test glasses that were heated for periods of a few months in the temperature range 500 to 800°C.^{1,5} Migration of heavy, separate phases in the glass might take place above the softening temperature. This could be of special concern with that part of high-level waste from uranium-only-recycle which contains most of the plutonium that was generated during reactor operation. The solubility of plutonium in a waste glass may be only about 2% by weight,⁶ in which case about 50% of this plutonium would occur as a separate phase in the glass. Hoskins and Berreth⁷ have suggested 550°C for the maximum temperature of waste glass during storage.

Considerations of the above information have led me to propose 500°C for the maximum temperature to which borosilicate waste glass should be exposed during retrievable geologic storage.

3. This is the limit value suggested by Hoskins and Berreth⁷ on the basis that significant cesium volatility occurs at temperatures greater than 750°C.

4. This is the value suggested by Hoskins and Berreth⁷ on the basis of preliminary evidence that this material is stable to at least 800°C.

5. These wastes will have relatively low heat production rates, and the maximum temperatures will probably be less than this limit value.

6. This limit value is based on the following considerations: The surfaces of the Zircaloy hulls may become very reactive during storage within a sealed canister as a result of the degradation of the protective

oxide film which will occur during heating in the absence of an oxidant.⁸⁻¹⁰ No complete analyses have been made of hazards that might result from the above effects and how these would depend upon the temperatures in the contained wastes.

My rough guess is that 200°C is a reasonably conservative maximum temperature for these cladding and hardware wastes. Additional work is needed to arrive at firmer estimates of reasonable temperature limits.

7. The surface area-to-volume ratio of these fused wastes will be much less than that for the compressed wastes, and, accordingly, the possibilities of encountering significant hazards from rapid surface reactions will be much less for the fused wastes. I believe that the 400°C limit is reasonably conservative. Additional work is needed to arrive at firmer estimates of reasonable temperature limits.

8. I do not have any good basis for estimating acceptable temperatures for concrete. Atlantic Richfield Hanford Company workers³ have recently recommended maximum temperatures of 110 and 165°C, respectively, for normal and abnormal conditions in structural concrete in long-term (100-year) surface storage of high-level waste. The abnormal conditions presumably could prevail for an indefinite period. I have assumed their value for normal operation. Additional work is needed to arrive at better estimates of reasonable temperature limits, if there is any incentive for use of higher temperatures in the concrete.

9. The considerations which led to this temperature limit are the same as those set out under note 6 above. Additional work is needed to arrive at firmer estimates of reasonable temperature limits for the spent fuel.

10. High-fired (1000°C) PuO_2 is very refractory and is difficult to dissolve by normal techniques.¹¹ Also, its vapor pressure is very low ($<10^{-10}$ atm at 500°C).¹¹ Accordingly, the stated limit of 500°C in Table 1, which will not be reached in practice with emplacement in a geologic repository, appears conservative.

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