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A STUDY ON FABRICATION TECHNOLOGY OF CERAMIC OVERPACK

—A CONCEPTUAL DESIGN AND FABRICATION OF A FULL-SCALE CERAMIC OVERPACK—

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－A CONCEPTUAL DESIGN AND FABRICATION OF A FULL-SCALE CERAMIC OVERPACK－

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要　旨

本研究は，炭素鋼等のオーバーパック材料の代替候補としてより長期の閉じ込め性が期待できるセラミック製オーバーパックの開発を目的とするものであり，その成果は次の通りである。

(1) 処分条件として地下1,000mの花崗岩体中を想定し，材料は構造材料への適用実績を考慮して磁器および$A\ell_2O_3$の2種類を対象として設計した。

(2) 形状は，円筒状の胴部と半球状の底/蓋部の構成とした。設計肉厚は，強度上の肉厚と腐食代の合計であり，解析の結果，強度上の必要肉厚は磁器：119mm，$A\ell_2O_3$：40mmとなった。腐食代に関する予備試験等の結果を考慮し，設計肉厚は磁器：150mm，$A\ell_2O_3$：50mmに設定した。

(3) 製作技術の比較的確立した磁器材料により実寸大オーバーパック（800mm O.D.×2200mmH×150mm t)を試作し今後の開発課題を摘出した。

(4) 外圧が不均一荷重の場合は，磁器材料では製作限界を超えるため，材料として$A\ell_2O_3$等のファインセラミックスが必要となることがわかった。

(5) 長期寿命評価上重要な劣化モードである遅れ破壊に関して評価を実施した。

本論文はMaterial Ressarch Society 第13回国際シンポジウム(1989年11月，ボストン）での発表をまとめたものである。

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CONTENTS

A STUDY ON FABRICATION TECHNOLOGY OF CERAMIC OVERPACK
- A CONCEPTUAL DESIGN AND FABRICATION OF A FULL-SCALE CERAMIC OVERPACK -

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ABSTRACT

The conceptual design and fabrication test of a full-scale ceramic overpack were performed from the viewpoint of structural barriers as a part of program to evaluate their potential use as overpack under conditions of deep geological disposal.

Materials investigated were porcelain (used for insulators) and Al_2O_3 with high purity of 99.7 %. The selected design consisted of a cylindrical body with hemispherical heads at each end. The design thickness of overpack is the sum of the structural thickness and corrosion allowance. The thickness required to resist the lithostatic pressure was estimated by the basic cylinder buckling formulas and finite element stress analyses in both case of uniform and non-unifom external pressure conditions. These analyses showed that structural thickness of 119 mm was necessary for overpack of porcelain and 40 mm for Al_2O_3 under the predicted maximum uniform pressure. In addition, fracture probability of delayed failure, one of significant degradation mode, was estimated for overpack of porcelain.

A full-scale overpack of porcelain, of dimensions 800 mm outer diameter x 2200 mm length x 150mm wall thickness, was fabricated under the ordinary level of fabrication technology.

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(Boston, November 27- December 2).

INTRODUCTION

One of the significant subjects in geological disposal of high-level radioactive waste is to assure the long term reliability and integrity of overpack. The overpack is expected to isolate the waste from the disposal environment for more than several hundreds years. In the disposal system in Japan, the primary candidate materials for overpack are carbon steel and titanium (and its alloy) [1], selected on the basis of the preliminary survey on literatures from many countries and various corrosion tests.

Ceramic materials are being considered as possible alternatives to metallic materials for long-term containment because of their high chemical stability and strength. Limited studies, however, have been performed on ceramic materials for applications to overpack [2,3,4,5,6,7], which focused mainly on corrosion behaviors in the predicted repository conditions.

Recently for industrial use, ceramic materials are applied widely to the structural components because of improvements in the reliability of ceramic components and its excellent properties (for example gas turbine engines and ceramic turbo-charger rotors). These wide applications depend on the progress of optimized design method, studies on behaviors of significant failure modes and life time prediction method, and establishment of fabrication process. The similar progress should be carried out to apply ceramic materials to overpack.

This paper describes the conceptual design, life time prediciton for the delayed fracture and fabrication of a full-scale ceramic overpack.

DESIGN CONDITIONS

The designs of overpack in this study were considered mainly in the view point of structural barriers. The following external pressure loading on overpack should be considered in the repository.

- Lithostatic pressure
- Water pressure
- Swelling pressure of buffer materials, etc.

Although equilibrium pressure between the lithostatic pressure and the swelling pressure is predicted to load on overpack, the initial lithostatic pressure was selected for conservative estimation. The following design conditions were assumed in this study.

Disposal depth : 1000 m in crystalline bedrock

External pressure for analyses

- Vertical pressure (P_V) : 280 kg/cm^2
  The pressure by overlying rock (density of rock = 2.8 g/cm^3).
- Horizontal pressure (P_H) : 280, 420 and 560 kg/cm^2
  The actual measurements in the deep rock indicate that the average horizontal pressure is equal to or slightly larger than the vertical pressure [8,9]. The measurements also show that horizontal pressure value is not uniform and the distribution depends on the directions. Therefore, analyses were carried out both uniform and non-uniform external pressure conditions.

MATERIALS INVESTIGATED

Porcelain (used for insulators), one of the traditional ceramics, and Al_2O_3 with high purity of 99.7 %, one of the specialized ceramics, were selected as candidate materials with reference to wide applications to industrial use and the following reasons.

Porcelain : Design method for structural components has been well-established and large components such as hollow insulators have been produced for actual industrial use.

Al_2O_3 : Al_2O_3 is one of the most widely-used ceramic material applied to structural components in specialized ceramic. In addition, the fabrication test of 1/3-scale container for disposal of spent fuel was carried out in Sweden [10].

The properties of these materials are shown in Table I.

FORM AND DIMENSIONS

The form of overpack was selected to consist of a cylindrical body with hemispherical ends (lid and bottom), ensuring that no tensile stress is present when the overpack is subjected to an external uniform pressure loading.

In this study, one waste glass canister (approximately 430 mm outer diameter × 1040 mm length) was selected to be enclosed in overpack.

The design thickness of overpack is the sum of the structural thickness and the corrosion allowance. There are, at present, no design standards which are directly applicable for the evaluation of the structural integrity of overpack. The following two steps were used to determine the structural thickness of the overpack components:

-Step 1. Preliminary wall thickness were estimated using the basic cylinder buckling formulas.

-Step 2. Detailed FEM stress analyses were performed to confirm the adequacy of these wall thicknesses.

Design by the basic cylinder buckling formulas

Calculation results of each component wall thickness obtained by the basic cylinder buckling formulas (Table Ⅱ)[11] are summarized in Table Ⅲ.

These results show that wall thickness of 119mm is neccesary for overpack of porcelain and 40mm for Al_2O_3 under the predicted maximum uniform external pressure.

Design by FEM analyses

FEM (finite element method) stress analyses were carried out to determine the stress distribution occurring in the overpack. Results of these analyses were used to confirm that the maximum stress in the overpack is lower than the allowable stress of materials.

As mentioned earlier, the intial lithostatic pressure was considered as external pressure loading on overpack. The actual lithostatic pressure measurements indicate that horizontal pressure value is not uniform and the distribution depends on the directions. This non-uniform distributed pressure causes tensile stress in overpack, severe conditions for ceramic materials.

The FEM stress analyses for uniform horizontal pressure (Figure 1) was carried out for the overpack of porcelain as the first step and non-uniform pressure (Figure 2) for each of the materials as the next step. The actual external pressure at disposal conditions will be expected to be similar to the uniform pressure due to the effect by buffer materials.

Uniform pressure

Analyses conditions are shown in Table Ⅳ. Results of FEM analyses indicate that no tensile stress occurs and the maximum compressive stress occurring in the overpack was lower than the allowable stress of porcelain.

Non-uniform pressure Results for the overpack of porcelain (used for insulators), indicated the requirement of a considerably thick wall. Therefore, additional analyses for Al_2O_3 were conducted. Analyses conditions are also shown in Table Ⅳ.

One example of maximum stress contours is shown in Figure 4. Figure 5 shows the relation between the wall thickness and the tensile stress occurring in the overpack, which were obtained by several FEM stress analyses. The wall thicknesses required for non-uniform pressure are summarized in Table Ⅴ.

Depending on the sum of the structural thickness obtained by these FEM analyses and calculated 1000 years corrosion allowance, the fabricability of the full-scale overpack is considered as follows:

Porcelain The dimension limit in present fabrication technology is 80 mm (wall thickness) at the mass production stage. Porcelain is expected as the potential material under the uniform external pressure, but is not suitable for non-uniform pressure.

Al_2O_3 Wall thickness of 140 mm is sufficient for even non-uniform pressure conditions and the corrosion rate is expected to be negligibly low ($<$ 1mm/1000y [5]). The fabrication of the full-scale overpack will be achieved by the future development of fabrication technologies for large structural components.

LIFE TIME PREDICTION FOR DELAYED FRACTURE MODE

In the repository conditions, overpack will not be subjected to significant cyclic loading, and the overpack temperature will be well below the creep range. Therefore, corrosion and delayed fracture [SCG (Slow Crack Growth) failure] should be estimated as dominant degradation modes. At present, the preliminary corrosion tests for several ceramic materials are carried out in Power Reactor and Nuclear Fuel development Corporation. In this paper, the fracture probability by delayed failure was calculated for the overpack of porcelain, of which the properties of its failure mode are well established compared to the other materials.

Life time prediction method

Fracture probability of ceramic components exhibiting delayed failure can be calculated by means of Weibull's theory. Based on the results of FEM stress analyses and properties of material, the failure conditions are exhibited in the following equation for delayed fracture [12,13].

$$\ln\ln[1/(1-F)]=[m/(n-2)](\ln Tf-\ln B+n\ln\sigma f)-m\ln Stp + \ln[Ve(comp)/Ve(tp)]+m\ln\Gamma(1+1/m) \qquad [1]$$

where F : failure probability.

m : Weibull modulus.

B,n : SCG parameters.

Tf : average life time.

σ_f : maximum stress in the overpack component.

Stp : strength of test specimen.

$Ve(comp), Ve(tp)$: effective volume of a component and test specimen.

The analyses conditions for life time prediction are selected as shown in Table VI based on the following two reasons.

- Only the tensile stress leads the SCG failure.
- FEM stress analyses mentioned above show that the critical tensile stress occurs in the cylindrical body under non-uniform pressure loading.

Results of life time prediction

Maximum stress occurring in overpack as a function of ratio α(Pmin/Pmax) is presented in Figure 6. In the range of $\alpha > 0.7$, no tensile stress occurs in overpack, indicating that SCG fatigue failure is not dominant fracture mode. On the other hand, external pressure conditions in the range of $\alpha < 0.5$ cause the fast fracture of overpack due to maximum stress exceeding the allowable strength of materials. In the case of $\alpha = 0.6$, delayed fracture should be considered as degradation mode. Figure 7 shows that calculated failure probability with time for three Weibull modulus values and failure probability of this ceramic overpack [porcelain (used for insulators),m = 20] was neglegibly small, (i.e. 9.8×10^{-6} for 10^3 years and 3.7×10^{-4} for 10^6 years).

These results indicates that the delayed fracture of this type of overpack should be considered only in the narrow range of non-uniform pressure conditions.

FABRICATION OF A FULL-SCALE OVERPACK

Porcelain was selected as the material for fabrication test of a full-scale overpack in consideration of the present fabrication technology level, although Al_2O_3 will be the promising material in the near future. The following designs were selected for fabrication test.

- Material : Porcelain (used for insulators)
- Form : A cylindrical body with hemispherical ends
- Dimensions : One waste glass canister was selected to be enclosed.
  800 mm outer diameter x 2200 mm length x 150 mm wall thickness
  Wall thickness of 130 mm is required to resist the lithostatic pressure. Additional wall thickness of 20 mm is the corrosion allowance selected with reference to corrosion test results of porcelain [2,3], and the our preliminary corrosion tests.
- Weight : 1.7 ton
- Fabrication procedures : Ordinary fabrication procedures such as extrusion, slip casting, and firing, etc. were adopted. The outline and appearance of this ceramic overpack is shown in Figure 8.
- Sealing method : Ordinary joint method such as graze-joint was expected to apply to the sealing between body and lid.

The subjects for a future study were obtained through a series of considerations on designs and fabrications.

<u>Porcelain (Traditional ceramic)</u>

- Clarifying of applicable disposal conditions.
- Improvement of sealing method between body and lid with high corrosion resistance.

<u>Al_2O_3 (Specialized ceramic)</u>

- Development of fabrication technology for large structural component.
- Development of sealing method between body and lid.

The development of inspection technology is the common subject on two materials.

CONCLUSIONS

- The conceptual designs under the predicted disposal conditions of uniform and non-uniform external pressure in the range of 280 - 560 kg/cm² showed that:
  - Porcelain (used for insulators) was evaluated for their potential use as overpack material under the uniform external pressure conditions.

  - Overpack of Al_2O_3 with high purity of 99.7 % was expected to be available for predicted most severe pressure conditions (non-uniforn pressure, Pmin/Pmax=0.3).
- Delayed fracture should be considered only in the restricted range of non-uniform pressure conditions.
- Further development is necessary for the fabrication technology, such as sealing methods between body and lid and inspection methods.

ACKNOWLEDGEMENTS

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Table Ⅰ The Strength of Candidate Materials

Material	Compressive Strength (kg/cm²)	Tensile Strength (kg/cm²)	Bending Strength (kg/cm²)	Modulus of Elasticity (kg/cm²)	Poisson's Ratio (−)
Porcelain	6,200 ～ 9,600	350～800	600～1,600	$6 \sim 9 \times 10^5$	0.2～ 0.2
Al_2O_3	13,000 ～20,000	——	4,400～5,000	——	0.2～ 0.25

Table Ⅱ The cylinder buckling formulas[11]

Component	Formulas	Remarks
Cylindrical body	$t = r_2 - r_1 \geqq \left[\left(\dfrac{\sigma_c}{\sigma_c - 2PS} \right)^{1/2} - 1 \right] - r_1$	P : External pressure σ_c : Compressive stress r_2 : Outer radius r_1 : Inner radius = 250 mm S : Safety factor = 3
Hemispherical Lid/Bottom	$t = r_2 - r_1 \geqq \left[\left(\dfrac{2\sigma_c}{2\sigma_c - 3PS} \right)^{1/3} - 1 \right] - r_1$	

Table Ⅲ Wall Thickness Calculated by Buckling Formulas

External Pressure	280 kg/cm²		420 kg/cm²		560 kg/cm²	
Material	Body	Lid	Body	Lid	Body	Lid
Porcelain	43	20	74	32	119	47
Al_2O_3	18	9	28	13	40	19

Table Ⅳ FEM Stress Analyses Conditions

Computer code	:	ISAS Ⅱ (Conformity to NASTRAN code)	
External pressure	:	Vertical 280kg/cm^2 ,	Horizontal 560kg/cm^2
Properties of materials		Modulus of elasticity	Poisson's ratio
- Porcelain	:	0.65×10^6 kg/cm^2	0.2
- Al_2O_3	:	2.2×10^6 kg/cm^2	0.25
Analysis model			
- Uniform pressure	:	Three-dimensional finite element model (Fig- 3)	
- Non-Uniform pressure	:	Two-dimensional finite element model	

Table Ⅴ Wall thickness required for non-uniform pressure

Material	Tensile strength (kg/cm^2)	Safety factor	Wall thickness (mm)
Porcelain	350～800	3	290
Al_2O_3	～2500	3	140

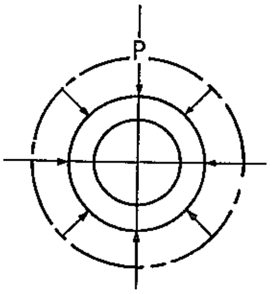

Figure 1.
Uniform External
Pressure Condition
- Profile of
Cylindrical body -

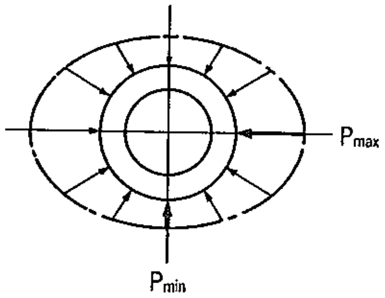

Figure 2.
Non-uniform External
Pressure Condition
- Profile of
Cylindrical body -

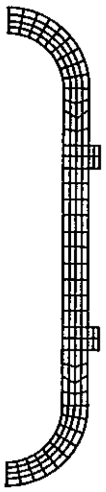

Figure 3.
Three-dimensional
Finite Element Model

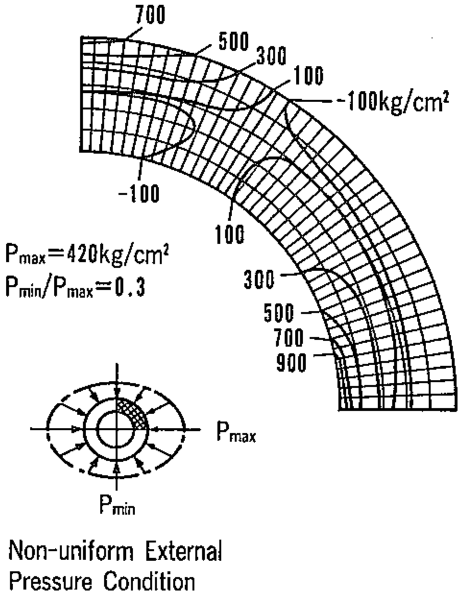

Figure 4.
Maximum Principal Stress Contours
from Two Dimensional FEM Analysis
for Profile of Cylindrical Body
- Pocelain -

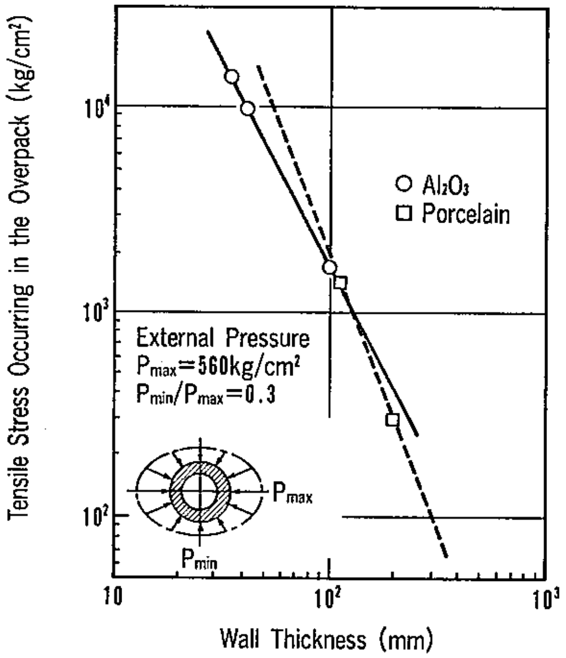

Figure 5.
Relation of Tensile Stress and
Wall Thickness of the Overpack under
Non-uniform External Pressure

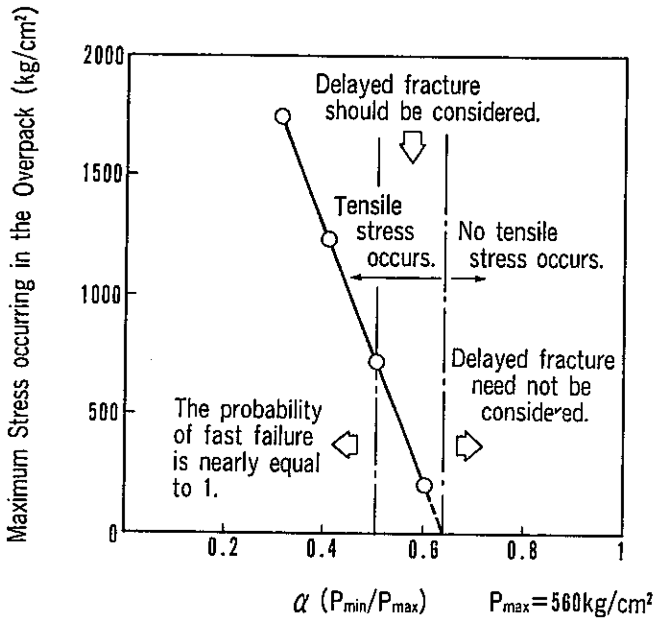

Figure 6.
Effect of Non-uniform Pressure Condition on the Maximum Stress occurring in Overpack
- Porcelain -

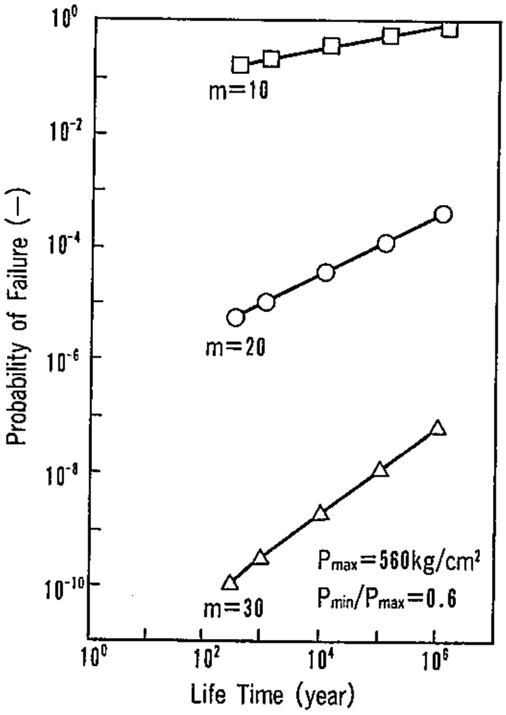

Figure 7.
SPT (Strength Probability Time) Diagram of Ceramic Overpack
- Porcelain -

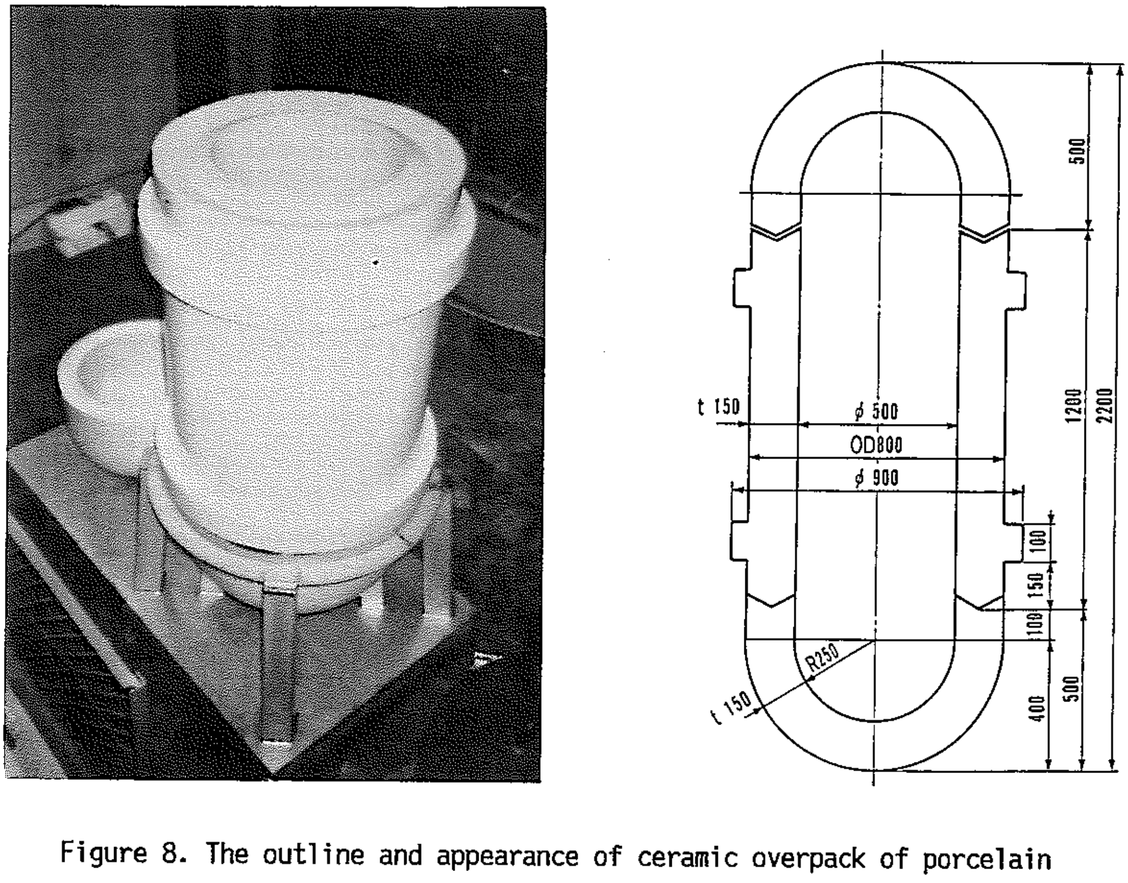

Figure 8. The outline and appearance of ceramic overpack of porcelain