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ANALYSIS OF FISSION PRODUCT REVAPORIZATION IN A BWR REACTOR COOLANT SYSTEM DURING A STATION BLACKOUT ACCIDENT*

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ABSTRACT

This paper presents an analysis of fission product revaporization from the Peactor Coolant System (RCS) following the Reactor Pressure Vessel (RPV)failure. The station blackout acci-dent in a BWR Mark I Power Plant was considered. The TRAPMELT3 models for vaporization, chemisorption, and the decay heating of RCS structures and gases were used and extended beyond the RPV failure in the analysis. The RCS flow models based on the density-difference or pressure-difference between the RCS and containment pedestal region were developed to estimate the RCS outflow which carries the revaporized fission product to the containment. A computer code called REVAP was developed for the analysis. The REVAP code was incorporated with the MARCH, TRAPMELT? and NAUA codes from the Source Term Code Package (STCP) to estimate the impact of revaporization on environmental release. The results show that the thermal-hydraulic conditions between the RCS and the pedestal region are important factors in determining the magnitude of revaporization and subsequent release of the volatile fission product into the environment.

INTRODUCTION

The retention of **eadionuclide** in the Reactor Coolant System (RCS) of a light water reactor is a major concern in severe accident studies. Many analyses have indicated that a large fraction of material released during core heatup and degradation can remain in the RCS. For example, calculations for the Peach Bottom staticn blackout transients showed retention of 54% and 70% for CsI and CsOH, respectively, as reported in the RCS are deposited either as aerosol or condensed vapor on the internal surfaces of the RCC. It is generally acknowledged that the decay power of the deposited material can increase the temperatures of the deposites

*Work performed under the auspices of the U.S. Nuclear Regulatory Commission. and the structural surfaces. The raise of temperature will result in the revaporization of the volatile fission products. The magnitude and timing of the revaporization after the RPW failure are important for the release of radionuclide through the containment to the environment. The revaporization is particularly important for BWRs under certain severe accident conditions. For example, the revaporization following reactor vessel failure releases the volatile fission product directly into the drywell regin without scrubbing in the pressure The importance of fission suppression pool. product revaporization is acknowledged in Draft NUREG-1150.³ It is stated that the two factors governing the revaporization issue are the chemistry of retained radionuclides and the RCS thermal-hydraulics following the vessel failure.

The chemistry involves the decay of Te-132 and reactions between the deposited fission products and structure surfaces. The continuous decay of Te-132 retained in the RCS into I-132 could result in a late source of I-132 to be released to the environment. The surface reaction could result in the formation of stable fission production compounds that have lower vapor pressure which, in turn, will affect the revaporization process. Since the chemical processes are still under investigation and are not fully understood yet, they are not included in the present work. The RCS thermal-hydraulics following the reactor vessel failure involves the predictions of decay heating of structures and gases, natural convection and the inflow and outflow of the RCS. The transport of the revaporized fission product into the containment is carried by the RCS outflow.

 material
 A review of major activities related to ther as fission product revaporization is given in Draft sur

 nal sur NUREG-1150 Appendix J.³ The review included owledged works performed by Stone and Webster, IDCOR, NRC material

 Severe Accident Sequences Analysis (SASA) Prodeposits
 gram and New York Power Authority. Because of the complexity of the revaporization phenomena

 the U.S.
 and the lack of sufficient data for model

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these studies. The dependence of the revaporization on specific accident sequence and on RCS/containment design make the comparison of these studies difficult.

The NRC STCP⁵ has fully coupled fission product transport and thermal-hydraulic analyses through its MARCH/TRAPMELT3 codes. However, the STCP does not include any in-vessel thermal-hydraulics after vessel failure due to the lack of post-vessel failure core condition. Hence, no fission production revaporization is available in STCP. With the above background, a study of the revaporization within the scope of STCP was undertaken at BNL. A simplified thermal hydraulic model was developed for the RCS after the reactor vessel failure. The thermal hydraulics is coupled with the fission product transport model of TRAPMELT3 to provide an estimate of the revaporization of three volatile fission product groups (i.e., CsI, CsOH, and Te) and their release to the containment. A computer code called REVAP was developed. The output of the PEVAP, a new source due to revaporization, was used in the NAUA code to estimate the impact of revaporization on environmental release of fission products.

MODELING OF FISSION PRODUCT REVAPORIZATION IN THE RCS FOLLOWING THE RPV FAILURE

The mcdeling of fission product revaporization following the RPV failure consists of two parts. The first part involves of computation of decay heating of RCS structures and gar -, the evaporation, condensation and chemisorption rates of three volatile fission products (Cs1, Cs0H and Te). This is done by using the existing models of TRAPMELT3.⁴ The second part involves the development of models to estimate the RCS thermal-hydraulic conditions and the transport of the revolatilized fission products into the containment. The computational procedure of the REVAP code is shown in Figure 1. The description of the related models will follow the procedures illustrated in Figure 1.

First, three input parameters are required for the revaporization calculation. They are the characteristic height of RCS (CHT), RCS rupture area size (AREA) and the RCS initial pressure following the reactor vessel failure (PIVF). Then, an input tape provided by TRAPMELT3 is required to define the parameters of the various control volumes and the final state as computed by the TRAPMELI3 code prior to vessel failure. The control volume parameters include volume size, height, surface area, structure thickness, and the mass heat capacity of each structure. The TRAPMELT3 computed hydrogen mass, steam mass, gas temperature, structure temperature and the state of fission product in each control volume at the time just prior to the vessel failure are used to define the initial conditions for the revaporization calculation. The state of fission products includes the TRAPMELT3 computed parameters of each species, namely, the suspended masses of vapors and aerosols in space, condensed vapors and aerosols deposited on structures, and those chemically absorbed by structures. In addition, the core inventory, initial fission power, and the RCS pressure are included in the TRAPMELT3 input tape. Modifications of the TRAPMELT3 data are made to properly define the initial state in the RCS immediately after the vessel failure. A new control volume is added to represent the lower head of the reactor vessel and the core volume is adjusted for the relocation of core materials into the reactor pedestal region according to the MARCH assumption. The pseudo control volume of containment used in TRAPMELT3 is discarded. The final state computed by TRAPMELT3 represents the RCS under high pressure prior to the vessel rupture. According to the STCP calculation, all materials including steam, hydrogen, fission product vapor and suspended aerosols are blown out instantly from the RCS to the containment in the form of a puff release at the time of vessel rupture. After the puff, release, no further interaction with the RCS is assumed in STCP calculation. For the revaporization calculation, the end of vessel failure is the beginning of the PCS/containment interaction. Hence, an initial state of RCS must be defined for this new phase of transient. It is assumed that immediately after the puff release, the RCS pressure is at equilibrium with that in the pedestal region of the containment which is computed in MARCH. This pressure is the input parameter (PIVF) for REVAP. Using this pressure, the given control volume size and the final gas temperature from TRAPMELT3, the mass of gases in each control volume are computed by the perfect gas law. The fractions of hydrogen. steam and fission product vapors in each cortrol volume are assumed unchanged initially. All suspended aerosols are assumed to be removed completely to the containment as modeled in the MARCH code and no further treatment of the suspended aerosol is involved in REVAP calculation.

Another input tape containing MARCH predicted time-dependent pressures and temperatures in the containment drywell region during the entire transient after the vessel failure is required. The pedestal region located below the reactor vessel is included in the drywell region according to the MARCH calculation. The MARCH tape contains the partial pressures of steam hydrogen, oxygen, nitrogen, carbon dioxide and carbon monoxide. It also contains the drywell atmospharic temperature and the corium surface temperature. The partial pressures and temperatures are used to compute the atmospheric composition and density. The total pressure and density in the pedestal region will be used to determine the inflow and outflow of the RCS.

The decay neating of RCS structures and gases are computed using the existing models of RAPMELIB. In these models, the decay energy is determined from the ANS decay heat standard, and the distribution of decay energy among the 10 fission product groups is based on calculations with the ORIGEN cone. All decay energy associated with gamma rays is assumed to be absorbed by the control volume structures based on their relative surface areas. The beta decay energy is distributed among the gas mixture and the structures based on an estimated beta particle range in the gas. Several subroutines in the TRAPMELT3 code are adopted directly in the REVAP code. They are:

- FISPQ For the calculation of energy addition _____ to structures and the gas mixture resulting from the decay of fission products which are airborne or deposited on structures.
- ANSG For the calculation of core decay heat as a function of the time following reactor shutdown based on standard ANS correlation.
- CESIUM For the calculation of airborne or deposited core mass fraction of cesium based on the mass fractions of cesium iodide (CsI) and cesium hydroxide (CsOH).
- DECAY For the calculation of location. species specific deposits for decay heat analysis.

The gas and structure temperatures in each control volume are determined by the following equations:

$$\frac{dT_s}{dt} = \frac{Q_r + Q_{fp} - h_c \cdot A \cdot (T_s - T_q)}{CM}$$
(1)

$$\frac{dT_q}{dt} = \frac{Q_{fpq} + h_c \cdot A \cdot (T_s - T_q)}{C_p \cdot M_t}$$
(2)

The two differential equations governing the structure temperature $\{T_g\}$ -and gas temperature $\{T_q\}$ are written in finite-difference form and are solved simultaneously by the implicit method. In the above equations,

- Jr_D = Beta and gamma for decay heat structures from FISPQ calculation,
- Gfpg = Beta decay neat for gas mixtures from FISPO calculation,
 - Qr = Radiative neat exchange between conjum in the cavity and the surface in the lower need control volume (no radiation for other control volumes),
 - A = Structural surface area (input),
 - CM = Mass heat capacity of structures (input),
 - Cp = Heat capacity of gas mixture, Mt = Total mass of gas mixture,

 - h_c = Heat transfer coefficient,

Note that the heat loss from RCS to containment is not included in the present analysis in order to have a conservative estimation of the fission product revaporization.

The heat transfer coefficient is based on the natural convection correlations given by the TRAPMELT3 code,

$$h_{c} = \frac{2 \cdot STLK \cdot HCN}{2 \cdot STLK + HCN \cdot DELX1}$$
(3)

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HCN = 0.1-K-(Pr-Gr)
$$^{1/3}$$
/HGT for Gr-Pr > 10⁹ (4

where STLK (25.0 Btu/hr-ft-F) is the stainless steel thermal conductivity used for considering the structural heat conduction effect, and DELX1 and HGT are the structure thickness and height, respectively. The Prandtl number (Pr), Grashof number (Gr), thermal conductivity (K) and heat capacity (c_p) are determined from properties of the gas mixtures. Initially, the RCS con-tains only steam and hydrogen. During the transient, gases such as O_2 , N_2 , CO_3 , and CO_2 will enter the RCS from the cavity compartment. These non-condensable gases will be included in the RCS gas mixture. The TRAPMELT3 subroutine PROP is used to compute properties of steam and hydrogen. MARCH correlations are used for properties of CO, CO_2 , N₂, and O_2 .

Since the vessel rupture may result a large opening area on the lower head, it is considered that the structure in the lower head may receive radiative heat from the hot corium in the react tor cavity below the ruptured vessel. The radi ative heat is approximated by:

$$Q_{r} = 0.173 \times 10^{-8} \cdot F \cdot A \cdot \left[(T_{rad} + 460)^{4} - (T_{s} + 460)^{4} \right]$$
(6)

where the Trad is the corium surface temperature given by the MARCH tape, A is the structure sur face area in the lower head control volume, and F is the radiation exchange factor assumed to be 0.5 in the present analysis.

Knowing the structure and gas temperature in each control volume, the condensation of evaporation of the three species (CsI, CsOH and Te) are computed by the TRAPMELT3 subroutine ADHOC and ADHOCS. The two subroutines are adopted directly in the REVAP code. The equil librium vapor concentrations of the species at these temperatures are the driving force for the evacoration or condensation process. The mass transfer coefficient used in the calculation is derived by the heat transfer analogy in the TRAPMELT3 code. The TRAPMELT3 chemisorption model" is adopted in the REVAP code. In this

model the sorption rate coefficient is given as 1 cm/s for Te and 0.012 cm/s for CSOH, independent of temperature. No chemisorption of CSI is considered. The chemisorption process is limited by the mass transfer rate and is irreversible. Once a species is chemically absorbed by the wall, it will never be revaporized.

The RCS thermal-hydraulics is computed based on several assumptions. All gases follow the perfect gas law. There is no pressure difference among the various control volumes, i.e., the entire RCS is under a uniform pressure. The presence of fission product vapor does not alter the tnermal-hydraulic benavior, except the den-sity of the gas mixture. The "chimney effect" associated with the LOCA-type sequences is not included. The analysis only applies to the transient-type sequence, where rupture of the vessel lower head is the only opening through which mass transfer with the containment occurs. Two types of flow are modeled in REVAP, namely, the pressure-driven flow and the density-driven flow as illustrated in Figure 2. During the transient, the density and pressure in the RCS are computed and compared with that in the containment pedestal region determined from the MARCH tape. The density is evaluated as a function of temperature and composition of the gas mixture. Fission product vapors are included in the gas mixture in the RCS but not in the containment.

A thermally stable situation is assumed if the density in the RCS is lighter than that in the pedestal region which is located below the reactor vessel. Under this situation, any flow between the RCS and pedestal region is caused by the pressure difference between the two regions. Either an inflow or an outflow from the RCS may occur. For example, the decay heating in the RCS will increase its pressure and result in an outward flow to the containment. On the other hand, high pressure in the containment induced by hydrogen burn will cause an inward flow to the RCS. For the pressure-driven flow, the mass flow rate is given by

$$W = A_0 \cdot C \cdot (\rho \approx DP)^{1/2}$$
(7)

where Ao = sive of opening,

C = loss coefficient,

- p = density of gas being transferred,
- DP = pressure difference between the RCS and containment.

Individual flow rates of the gas constituents are assumed proportional to their mass fraction. Thus, the release of volatile fission products are related to their mass fraction in the PCS.

A thermally unstable situation is assumed if the gas mixture in the RCS is heavier than that in the pedestal region. Under this situation, the heavier gas in the RCS tends to move downward into the pedestal region. Meanwhile, the light gas in the pedestal region moves upward into the RCS in order to satisfy the continuity requirement. A density-driven counter flow is expected at the ruptured opening. The counter flow is computed using the difference in potential energy compared to a stable situation with the heavier gas below. The driving force is

$$DP = CHT \cdot g \cdot (\rho_a - \rho_c)$$
(8)

It should be pointed out that Equation (8) only applies to the situation where a gross failure of the lower vessel head occurs. This implies that the flow exchange between the RCS and pedestal region is through a large open area. The present model does not apply to the situation where the lower vessel head has a small rupture area.

If one assumes that the volume flow rate of the upward flow is the same as the downward flow, it can be shown that the downward and upward mass flow rates are, respectively,

$$W_{\mathbf{d}} = \rho_{\mathbf{a}} \cdot A_{\mathbf{o}} \cdot V_{\mathbf{d}} \cdot \left(\frac{1}{1 + V_{\mathbf{d}}/V_{\mathbf{u}}}\right)$$
(9)

and

$$W_{u} = \rho_{c} \cdot A_{o} \cdot V_{u} \cdot \left(\frac{V_{d}/V_{u}}{1 + V_{d}/V_{u}}\right)$$
(10)

where V_d = downward flow velocity

$$= (2 \cdot Dp/p_a)^{1/2}$$
, and

V_u = upward flow velocity

 $= (2 \cdot Dp/p_{c})^{1/2}$.

Analm, the downward mass flow will carry the fission product vapor from the RCS into the containment according to its mass fraction in the gas mixture.

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RESULTS OF REVAPORIZATION FOLLOWING REACTOR VESSEL FAILORE

In order to demonstrate the effect of volatile fission product revaporization from the RCS, the REVAP code was applied to a short-term station blackout transient (TB) for a BWR Mark I Power Plant. According to the STCP analysis, the reactor vessel failed at 202 minutes after the initiation of the accident. Prior to the vessel failure, the volatile fission product distributions in the RCS are given below:

	wall Deposit		Chemisorption		
	Кg	ž	Kg	76	
Cs I CsOH Te	18.15 74.25 0.054	53.4 30.9 0.16	0.0 96.06 10.73	0.0 40.0 30.7	

The wall deposit includes both condensed vapors and deposit as prosols. The percentage is given in terms of initial core inventory. The initial core inventories are 33,98, 240.1 and 34.90 Kg for CSI, CSOH and Te, respectively. The revaporization of the wall deposits and chemisorption of the revaporized materials are the subject of this analysis.

After the vessel lower head breach, the corium entered the pedestal region underneath the reactor vessel. The contum/concrete interaction resulted in the generation of noncondensable gases and pressurization of the containment as snown in Figure 3. A drywell rupture was predicted to occur at about 317 minutes. The flow exchange between the drywell and containment ouilding deinerted the containment, and hydrogen burns were induced in the containment late in the transient. The MARCH predicted atmospheric temperature in the dryweil region and the corium surface temperature are included in Figure 3. The pressure and temperatures are important parameters to determine the inward or outward flow for the RCS. Two limiting cases were considered. In Case 1, the corium surface temperature is used to compute the pedestal atmospheric density. The high confum temperature will lead to a lower density which will produce a thermally unstable condition and induce a large density-driven flow between the RCS and pedestal region. A large release of the revolatilized fission products is expected in this case In Case 2, the drywell temperature is used to compute the pedestal atmosphere The lower drywell temperature will density. yield a neavier density which will produce a thermally stable situation. The release of fission products from the RCS will be limited by the pressure-driven flow, which is expected to be less then the density-driven flow in Case 1. The detailed results of Case 1 which represents a conservative approach will be discussed in this paper.

The predicted mass-average gas temperature of all control volumes in RCS is at about 1800 F level and is about 400 F lower than the corium surface temperature during most of the transient time. The lower RCS gas temperature results in a higher gas density as illustrated in Figure 4. Hence, a counterflow situation is created by the thermally unstable condition between the RCS and the pedestal region. Under natural convection conditions in the RCS, the structure temperature is very close to the adjacent gas temperature. The average structure temperature is also about 1800 F during most of the transient time. The revaporization of CsI, CsOH and Te determined by the wall temperature and their. partial pressures in the RCS are given in Figure 5. The calculation was ended at 680 minutes' approximately 8 hours after the vessel failure. It is seen that about 1130 g of CsI and 21,400 g of CsOH are revaporized in 8 hours. In terms of initia! wall deposits, the revaporization is only 0.6% and 2.9% for CsI and CsOH, respectively. For Te, all the 54 g initially deposited on the wall are evaporized within 92 minutes.

The revolatilized vapors are subjected to chemisorption by the structure walls according to the TRAPMELT3 model. The chemisorption of CSOH and Te are shown in Figure 6. About 202 g of the revaporized 21,400 g of CSOH and 12 g of the 54 g Te vapor are redeposited and chemically bonded on the structure walls. The high chemisorption rate of Te is caused by the high dep_sition velocity assumed in TRAPMELT3. The chemiscrption of CSOH appears to be insignificant.

The release rates of the fission product vapors associated with the RCS outflow are shown in Figure 7. The fluctuation of the release rate is largely caused by the numerical computation of the quasi-steady state outflow. The pulse-type outflow, which is based on an instantaneous comparison of the pressures and densities between the RCS and the pedestal region, is associated with a thermally unstable condition. These release rates will be used in the NAUA calculation for estimating the environmental releases. It is seen that the release rate of CsOH is one order of magnitude higher than that of CsI, and is two orders of magnitude higher than that of Te. Large releases of CsOH and CsI started at about 320 minutes after the containment failure and continued until the end of the calculation. The accumulated releases are given in Figure 8. At the end of 3 hours after vessel failure, a total of 1120, 20,970 and 42 g of CsI, CsOH and Te were released from the RCS to the drywell region without scrubbing in the suppression pool. The revaporization and release of Csl and CsOH from the RCS are long-term processes. At 8 hours after the vessel failure (i.e., the end of calculation) the CsI and CsOH are continuously released to the containment at the approximated rates of 4 g/min and 70 g/min. respectively.

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Comparison of Cases 1 and 2 shown in Table 1 indicates that the driving force of the RCS outflow is an important factor in determining the revaporization and release from the RCS. A thermally unstable condition between the RCS and the pedestal region will ennance the RCS outflow which results in a high evaporization in the RCS and a large release to the containment. Un the other hand, a thermally stable condition between the RCS and the pedestal region will limit the RCS flow and innibit the revaporization process. The results of this preliminary study agree qualitatively with the analysis by Donahue et al.⁶ for a BWR Mark II plant. In Reference 6. Stone & Webster's THREED-RCS code, derived from RELAP4 MOD5 code, was used to analyze both transient-initiated and loss-of-coolant accident cases. It is reported that for the LOCA sequence, the revaporization resulted in nearly complete subsequent release. For a transient with loss of containment heat removal (TW) only 6% is calculated to be released from the reactor pressure vessel (RPV) to the drywell due to revaporization over the next 24 hours. Their analyses also snowed that the natural convection between the drywell and one interior of the RPV is an important factor in the revaporization issue. The revaporization depends highly on the accident sequence and on the reactor/containment design.

EFFECT OF REVAPOPIZATION ON ENVIRONMENTAL RELEASES

The STCP version" used for the JUASAR analysis² was adapted to accept the Col, CsOH and Te revaporized from the RCS as an additional source to the aerosol transport and release code NAUA. A tape containing the revaporized species was created by the <code>PEVAP</code> code, in a form similar to the in-vessel leak which <code>TRAPMELT3</code> computes for the NAUA code prior to vessel failure. The tape contains time, source in (g/s) and release fractions. The aerosol density was fixed at 3 g/cm 3 as in TRAPMELT3 in the STCP analysis. Since aerosol size distribution is uncertain for the revaporized elements, the total volume-weighted number of particles was calculated as in TRAPMELT3 and uniformly assigned to the three smallest size bins, because the products of revaporization, if in aerosol form at all, would most likely be released in very small sizes.

For the present analysis, the revaporized material is released from the breach opening at the reactor vessel lower head. This additional release is treated similar to the puff and exvessel releases, and is input directly to the drywell compartment of the containment bypassing the suppression pool. Feedback leaks from the drywell enter the pool also and are subject to scrubbing as the in-vessel release. However, a different decontamination factor must be calculated for these leaks, because thermal-hydraulic conditions are different than for in-vessel release. Feedback leak from the reactor building to the containment is also significant and must be included in the calculations.

Three analyses were performed: (1) base case without revaporization products, (2) high revaporization from the RCS (Case 1) and (3) low revaporization (Case 2).

Table 2 shows the results for the fraction of initial core inventory release to the environment and the additional release due to revaporization for iodine, cesium and tellurium. The refractory species are not snown because they were not appreciably affected by the additional sources into the containment. Environmental releases for these groups are given in Ref. 2. It is seen in Table 2 that the low revaporization case (i.e., Case 2) has no significant effect on the release to environment. For the case of high revaporization (i.e., Case 1) the additional release of \underline{I}_2 is about the same as that in the base case; the additional release of Cs is about threefold of that in the base case. Hence, revaporization has a significant effect on the releases of $I_{\dot{z}}$ and Cs to the environment. Since the tellurium is mostly associated with the ex-vessel release due to corium/concrete interaction, the revaporization of Te from RCS has no impact on the total release to the environment.

SUMMARY

An analysis of the revaporization of volatile fission product in a BWR reactor coolant system following the vessel failure is performed for the Mark I power plant during a short-term station blackout transient.

It appears that the thermal conditions in the reactor vessel and pedestal region play an important role on the revaporization and its release from RCS to containment. The natural convection between the containment pedestal region and the interior of the RCS is the controlling mechanism for revaporization. According to the limiting cases studied in this work the additional revaporization releases of l_2 and Cs would have a significant effect on the total releases to the environment. The revaporization of Te has no apparent effect on the environmental release, since the Te is mostly associated with the ex-vessel release due to the corium/ concrete interaction.

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Fig. 3. MARCH Predicted Pressure and Temperature

RCS/cavity compactment flow exchange





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267	C 5	6.0(-3)	1.76(-2)	2.36(-2)	6.56(-4)
10	·e	1. 3 6(-1)	5.34(-4)	2 0(-1)	7.14(-5)
	_				

total

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6.46(-3)

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