Investigation for Radioactive Deposition Source Terms of AP1000 Units

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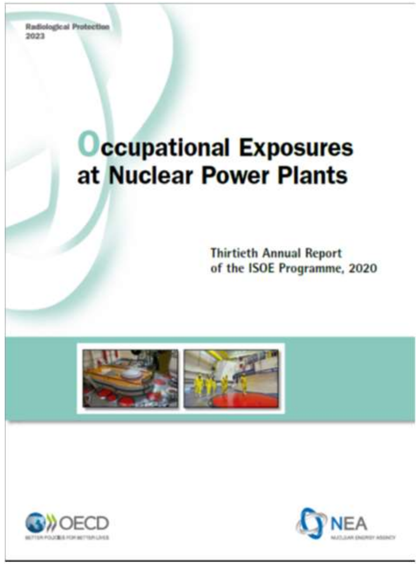
1. Introduction

- Throughout the world， occupational radiation exposure at NPP has steadily decreased since the early 1990s.
- However, some major challenges are still exist to RP professionals, for ensuring that occupational exposures are kept ALARA continuesly, because of：
  - the continued ageing and life extension of NPPs
  - regulatory, economic, social and other factors
  - new units built



- In normal operation of PWR, almost about 80%-95% of occupational exposure comes from deposited source terms (DSTs) during refueling outage.
- Through the investigation of DSTs. we can find the origins and variation trends, types and amounts of nuclides deposited on the inner surface of equipment, their contribution to the radiation field.

2. Formation Mechanism of Deposition Source Terms

- In PWR, corrosion products formed by the release of metal-oxide ions, colloids, and particles generated from the general corrosion of materials contact with the coolant, comprised of iron and nickel oxides with small amounts of manganese, chromium, and traces of cobalt.
- Some of corrosion products are released and transported by the coolant and redeposit throughout the primary system.

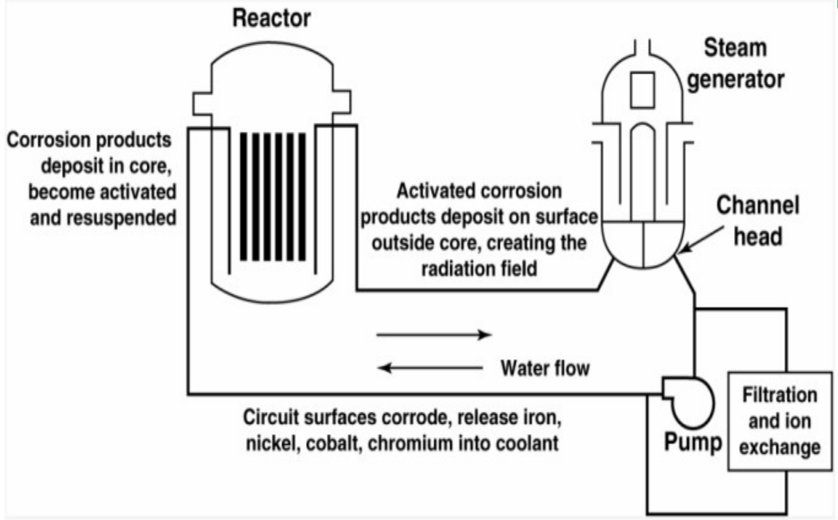


Fig. 1 Transport process of corrosion products activation in PWR primary circuit

- At the same time, some of them will be activated under neutron irradiation to form activated corrosion products (ACPs) . ACPs then redeposit on ex-core components forming DSTs, which directly result in increased radiation fields and the collective occupational radiation dose during refueling outage. The transport processes are showed in Fig. 1.

3. Method for Investigating the DSTs

3.1 Measurement Method for DSTs in Pipeline

- The DSTs have been investigated during the refueling outage.
- The related system are:
  - Reactor coolant system (RCS)
  - Chemical and volume control system (CVS)
  - Passive core cooling system (PXS)
  - Residual heat removal system (RNS)
- For this investigation, the high-purity germanium (HPGe) gamma spectrometer are setup directly outside the measured pipeline, as shown in Fig. 3.

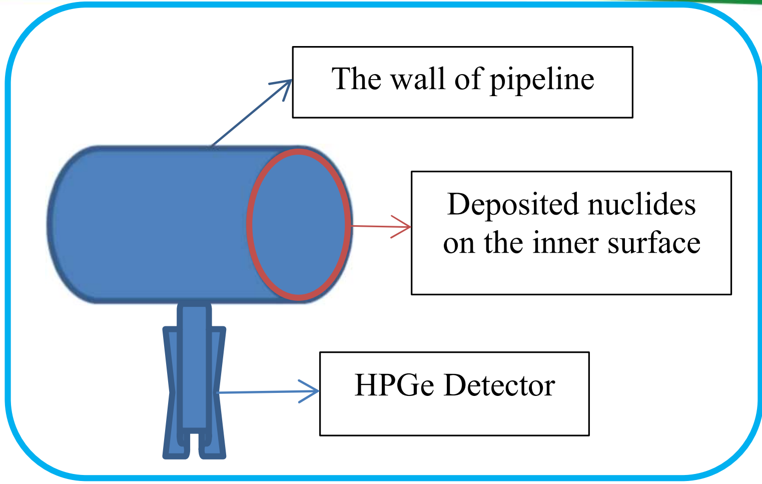


Fig.3 Layout Diagram of In situ High-purity Germanium (HPGe) Gamma Spectrum Measurement

Layout photograph of hot leg on site

3. Method for Investigating the DSTs

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3.1 Measurement Method for DSTs in Pipeline

- In this investigation, two kinds of data obtained:
  - gamma spectrum,
  - external contact dose rate of the corresponding piline.
- After gamma-ray spectrum analysis, we can get the type of nuclides and activity in DSTs.
- Masurement condition are determined：
  - parameters of the pipeline, detector
  - relative geometric parameters of the detector and pipeline, etc.

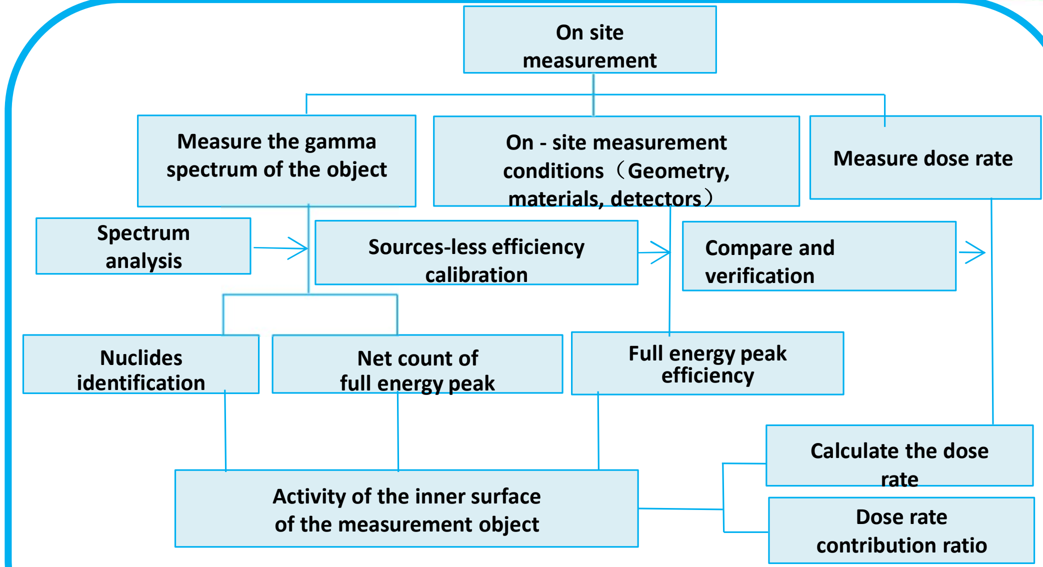

Fig. 4 Schematic Diagram of Deposited Source Term Investigation Method

Parameters of the primary coolant pipeline

measuring ststem	HPGe	name of pipeline	Loop 1 Hot Leg
insulation thickness, mm	135.00	outer protection layer thickness, mm	0
inner radius, mm	393.65	outer radius, mm	476.25
wall thickness of pipeline, mm	82.60	If there is any water in the pipeline	yes
materials of pipeline	austenitic stainless steel	distance, mm	610.00

3. Method for Investigating the DSTs

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3.1 Measurement Method for DSTs in Pipeline

- The surface activity of nuclides can obtain based on the source less efficiency calibration and activity calculation (formula 1).
- Based on the surface activity of DSTs , the dose rate generated by the deposited nuclides on the outer surface of the pipeline can be obtained.
- The reliability of the deposition source term measurement results have verified by comparing with the actual measured values.

$$A_S = \frac{n_E}{\varepsilon_E \times S \times p_E} \qquad (1)$$

Where:

A_S = surface activity of the nuclide, Bq/m^2

n_E = full-energy peak net count rate of gamma rays， s^{-1}

ε_E = full-energy peak detection efficiency of gamma rays

S = effective internal surface area of the pipeline， cm^2

p_E = branching ratio of gamma rays.

3. Method for Investigating the DSTs

3.2 Measuring Equipment

- As showed in Fig.5， the HPGe spectrumeter consists of a high-purity germanium detector, collimator, multi-channel analyzer, mobile trolley etc.
- The detector has a high measuring efficiency and good energy resolution.
  - Energy range: 50 keV - 2.2 MeV, which can cover the gamma-ray energy of most corrosion activation products in NPP (generally 300 keV - 2.0 MeV).
  - Dead time: less than 10%,
  - Full-energy peak count: more than 10,000 counts.

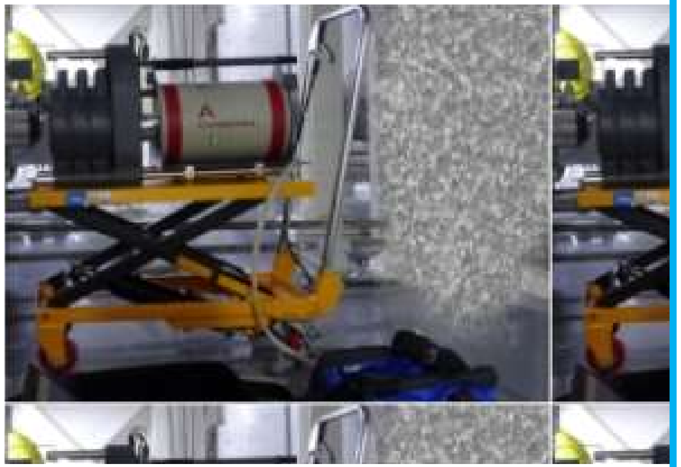


Fig. 5 Deposition Source Term Measuring Equipment

3. Methods for Investigating the DSTs

3.3 Selection of the Measuring Points

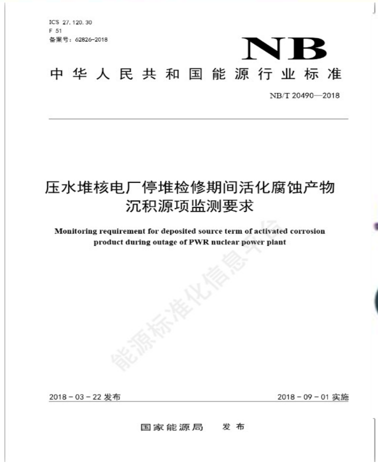
- Principle for selecting points:

① **The space around the system pipeline should be as large as possible.**

② **Hotspots should be avoided as around the measurement pipeline.**

③ **The structure of the pipeline should be as simple as possible.**

- According to NB/T 20490, combined with the system equipment layout of AP1000 units, 12 measurement points have selected, as showed in table 1.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中华人民共和国能源行业标准

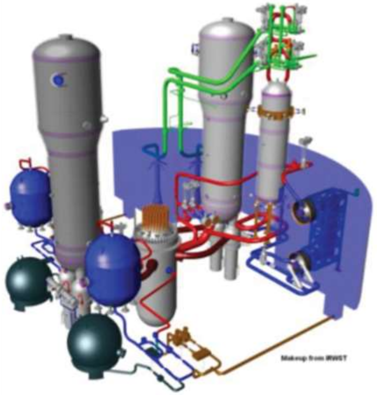
NB/T 20490—2018

压水堆核电厂停堆检修期间活化腐蚀产物沉积源项监测要求

Monitoring requirement for deposited source term of activated corrosion product during outage of PWR nuclear power plant

2018－03－22 发布　　2018－09－01 实施

国家能源局　发布



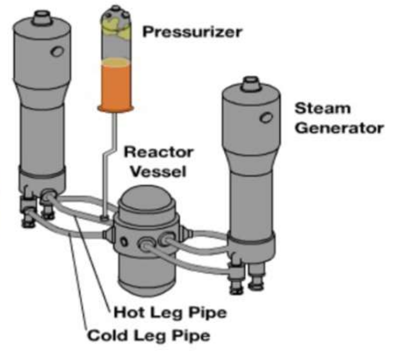


Table 1 Deposition Source Term Investigation Measurement Points

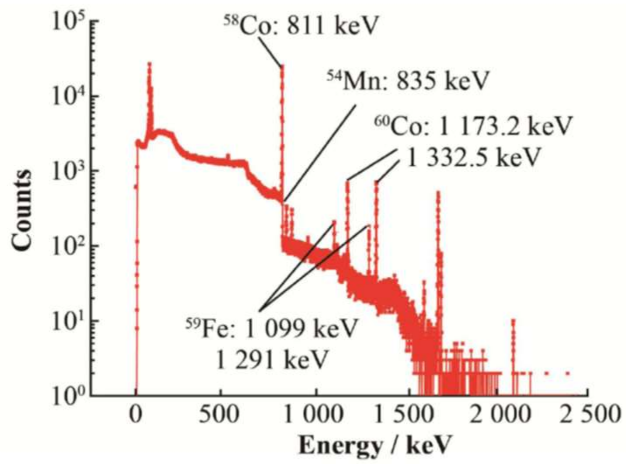
serial number	system	measurement point	serial number	system	measurement point
1	RCS System	Loop 1 Hot Leg	7	CVS System	Purification Return Pipeline
2	RCS System	Loop 1 Cold Leg	8	RNS System	RNS Header Inlet Pipeline
3	RCS System	Loop 2 Hot Leg	9	RNS System	RNS Header Outlet Pipeline
4	RCS System	Loop 2 Cold Leg	10	RNS System	RNS Heat Exchanger A Inlet Pipeline
5	RCS System	PZR Surge Line	11	PXS System	DVI B Pipeline
6	CVS System	Purification Outlet Pipeline	12	PXS System	PXS PRHR Inlet Pipeline

4 Results of the investigation

4.1 Reactor Coolant System (RCS)

Table 2 Surface Activity of Main Nuclides Deposited on the Inner Wall of Pipelines of RCS System

investigation point	surface activity of main nuclides deposited on the inner wall of the pipeline (Bq/cm²)									
	Co-58	Co-60	Fe-59	Mn-54	Nb-95	Zr-95	Zn-65	Cr-51	Ag-110m	Sb-124
Loop 1 Hot Leg	6.56E+04	1.43E+05	6.15E+04	5.82E+03	4.26E+03	2.27E+03	7.40E+03	3.92E+07	1.57E+03	2.01E+03
Loop 1 Cold Leg	3.02E+05	4.99E+04	3.18E+04	4.52E+03	5.43E+03	1.57E+03	2.12E+03	2.35E+06	4.48E+03	8.38E+02
Loop 2 Hot Leg	1.08E+05	1.56E+05	7.06E+04	1.30E+04	7.30E+04	4.51E+04	1.85E+04	2.96E+07	7.83E+03	6.14E+03
Loop 2 Cold Leg	1.62E+05	5.25E+04	1.47E+04	2.81E+03	6.36E+03	1.44E+03	1.33E+03	4.03E+06	6.50E+03	5.14E+02
PZR Surge Line	2.80E+04	6.50E+03	6.55E+02	8.25E+02	1.13E+03	1.94E+02	2.23E+02	9.29E+02	3.04E+02	4.54E+01
Average	1.33E+05	8.16E+04	3.59E+04	5.39E+03	1.80E+04	1.01E+04	5.91E+03	1.50E+07	4.14E+03	1.91E+03



HPGe spectrum for Loop 1 Hot Leg

From the table, it can be seen that:

- **Activity of Cr-51 is relatively high. Except for the PZR surge line, the activity of Cr-51 in other pipelines of the RCS system ranges from 10^6 to 10^7 Bq/cm².**
- **Activity of Co-58 is in the order of 10^4~10^5 Bq/cm², with average of 1.33×10^5 Bq/cm².**
- **Average surface activities of Co-60, Fe-59, Nb-95, and Zr-95 are above 10^4 Bq/cm². the surface activities of the remaining deposited nuclides range from 10 to 10^4 Bq/cm².**

4 Results of the investigation

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4.2 Chemical and Volume Control System (CVS)

Table 3 Surface Activity of Main Nuclides Deposited on the Inner Walls of Measured Pipelines of CVS System

investigation point	surface activity of main nuclides deposited on the inner wall of the pipeline (Bq/cm²)									
	Co-58	Co-60	Fe-59	Mn-54	Nb-95	Zr-95	Zn-65	Cr-51	Ag-110m	Sb-124
Purification Outlet Pipeline	7.51E+04	1.03E+04	9.68E+02	9.26E+02	1.31E+04	5.42E+03	6.61E+02	1.14E+04	1.60E+04	4.29E+02
Purification Return Pipeline	1.35E+04	1.68E+03	1.60E+02	2.13E+02	1.29E+03	5.74E+02	1.18E+02	1.38E+03	1.69E+03	8.02E+01
Average	4.43E+04	6.01E+03	5.64E+02	5.69E+02	7.22E+03	3.00E+03	3.90E+02	6.40E+03	8.84E+03	2.55E+02

It can be seen that:

- **The deposited nuclides on the inner walls of the CVS system pipelines include Co-58, Co-60, Fe-59, Mn-54, Nb-95, Zr-95, Zn-65, Cr-51, Ag-110m, Sb-124, etc.**
- **The activity of Co-58 is relatively high, with an average of 4.43×10^4 Bq/cm².**
- **The average activities of Ag-110m, Nb-95, Cr-51, Co-60, and Zr-95 are in the order of 10^3 Bq/cm².**
- **The average activities of other nuclides range from 10 to 10^2 Bq/cm².**

4 Results of the investigation

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4.3 Residual Heat Removal System (RNS)

Table 4 Surface Activity of Deposited Nuclides on the Inner Walls of Measured Pipelines of the RNS System

investigation Point	surface activity of main deposited nuclides on the inner wall of the pipeline (Bq/cm^2)									
	Co-58	Co-60	Fe-59	Mn-54	Nb-95	Zr-95	Zn-65	Cr-51	Ag-110m	Sb-124
RNS Header Inlet Pipeline	8.88E+03	1.57E+03	7.57E+03	1.72E+02	2.37E+03	1.22E+03	3.67E+01	1.80E+04	7.62E+02	5.05E+02
RNS Header Outlet Pipeline	1.13E+04	1.18E+03	1.52E+04	6.71E+01	1.16E+03	2.43E+02	1.01E+02	3.34E+03	1.71E+03	1.27E+02
RNS HX Inlet Pipeline	2.98E+03	8.97E+02	3.23E+03	7.25E+01	8.08E+02	3.51E+02	1.80E+01	9.42E+03	2.55E+02	1.60E+02
Average	7.73E+03	1.22E+03	8.68E+03	1.04E+02	1.45E+03	6.05E+02	5.19E+01	1.02E+04	9.09E+02	2.64E+02

It can be seen that:

- **The nuclides on the inner walls of the RNS system pipelines include Co-58, Co-60, Fe-59, Mn-54, Nb-95, Zr-95, Zn-65, Cr-51, Ag-110m, Sb-124, etc.**
- **The activities of Cr-51, Fe-59, Co-58, Co-60, and Nb-95 are relatively high, with average values in the order of 10^3 to 10^4 Bq/cm^2.**
- **The average activities of other nuclides range from 10 to 10^2 Bq/cm^2.**

4 Results of the investigation

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4.4 Passive Core Cooling System (PXS)

Table 5 Surface Activity of Main Deposited Nuclides on the Inner Walls of Various Pipelines in the PXS System

investigation point	surface activity of main deposited nuclides on the inner wall of pipeline (Bq/cm²)									
	Co-58	Co-60	Fe-59	Mn-54	Nb-95	Zr-95	Zn-65	Cr-51	Ag-110m	Sb-124
DVI B Pipeline	3.76E+04	1.34E+04	9.95E+03	1.89E+02	2.06E+03	8.93E+02	3.86E+02	1.34E+05	3.18E+03	2.04E+02
PRHR Inlet Pipeline	9.08E+04	1.79E+04	5.76E+02	1.59E+03	7.19E+02	1.79E+02	1.46E+03	9.47E+02	1.96E+03	6.19E+01
average	6.42E+04	1.57E+04	5.26E+03	8.90E+02	1.39E+03	5.36E+02	9.21E+02	6.73E+04	2.57E+03	1.33E+02

It can be seen that:

- **The nuclides on the inner walls of measured pipelines of the PXS system include Co-58, Co-60, Fe-59, Mn-54, Nb-95, Zr-95, Zn-65, Cr-51, Ag-110m, Sb-124, etc.**
- **The activities of Co-58, Co-60, and Cr-51 in various pipelines of the system are relatively high, with average values of 6.42×10^4 Bq/cm², 1.57×10^4 Bq/cm², and 6.73×10^4 Bq/cm² respectively;**
- **The average activities of other nuclides are in the order of 10^2~10^3 Bq/cm².**

4 Results of the investigation

4.5 Comparison of Calculated and Measured Surface Contact Dose Rates of Pipelines

- The measurement conditions are simulated using the MC method, to obtain the detection efficiency of the HPGe system under the investigantion specific scenarios.
- The activity of the DSTs on the inner walls of the pipelines, and the sum of the surface dose rates produced by each radionuclide on the pipelines are calculated.
- When γ-spectrum measurement conduct, the surface dose rate of the measured pipeline is also measured using survey meter.

Table 6: The calculated and measured surface dose rates, and relative deviation for the surveyed pipes during HY102 outage

Related system	Investigation point	Calculated dose rate (μSv/h)	Measured dose rate (μSv/h)	Relative deviation
RCS	Loop1 hot leg	114.03	99.85	14.20%
	Loop1 cold leg	134.99	120.88	11.68%
	Loop2 hot leg	117.03	100.92	15.96%
	Loop2 cold leg	104.43	97.20	7.44%
	PZR surge pipeline	49.43	32.48	47.79%
CVS	Purification outlet pipeline	348.96	285.96	22.03%
	Purification return pipeline	79.47	61.60	29.01%
RNS	RNS Header Inlet Pipeline	59.07	59.04	0.05%
	RNS Header outlet Pipeline	101.74	85.00	19.70%
	RNS HX A inlet pipeline	63.43	46.09	37.63%
PXS	DVI B pipeline	191.25	165.66	15.45%
	PRHR inlet pipeline	145.10	94.20	54.04%
SFS	SFS heat exchanger A outlet pipeline	11.61	9.47	22.55%

4 Results of the investigation

4.5 Comparison of Calculated and Measured Surface Contact Dose Rates of Pipelines

- In order to verify the accuracy of the radiation source term measurements, the relative deviation between the calculated and measured surface dose rate are compared.
- The scatter plot with the calculated dose rate on the X-axis, and the measured dose rate on the Y-axis are showed in Fig.6.
- It can be seen that the calculated and measured surface dose rates are consistent well, and the correlation coefficient is 0.98.

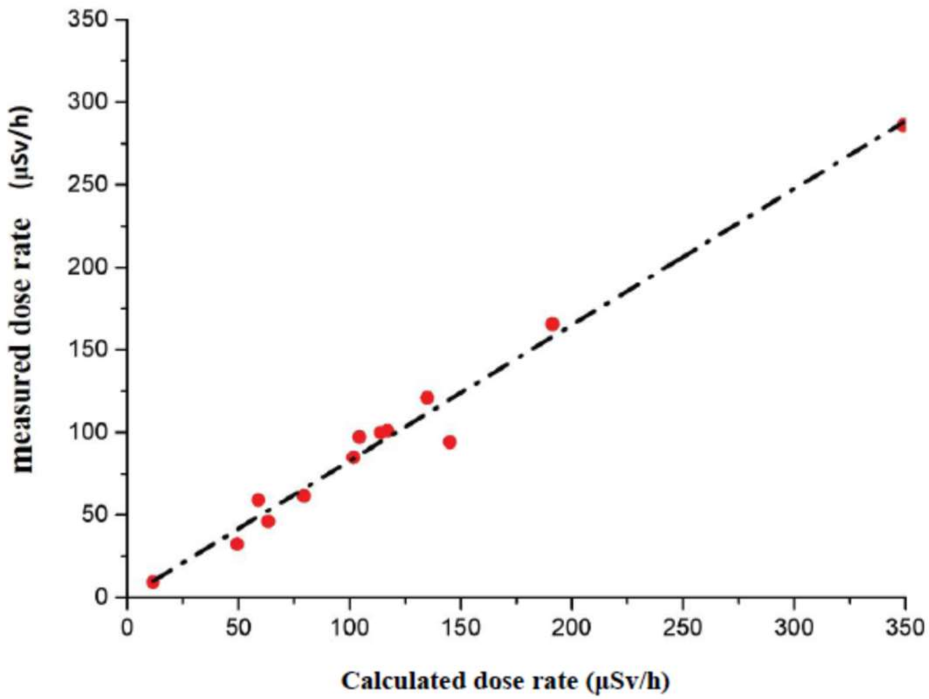

Fig. 6 Scatter plot and linear fitting curve

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

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- In order to comparise the general DSTs level of AP1000 and M310 unit, 8 units (4 are AP1000 , 4 are M310) are selected, and assumed that the average surface activity of the deposited nuclides on the inner walls of the RCS system pipes of each type of units represents the average level of DSTs of that type of nuclear power unit.
- The surface activities of Co-58, Cr-51, and Ag-110m in the deposition layer on the RCS pipelines of different type units are shown in Fig. 7.
- In general,
  - level of Co-58 surface activity on the inner walls of RCS system pipelines in AP1000 units is lower,
  - level of surface activities of Cr-51 and Ag-110m in AP1000 units is higher.

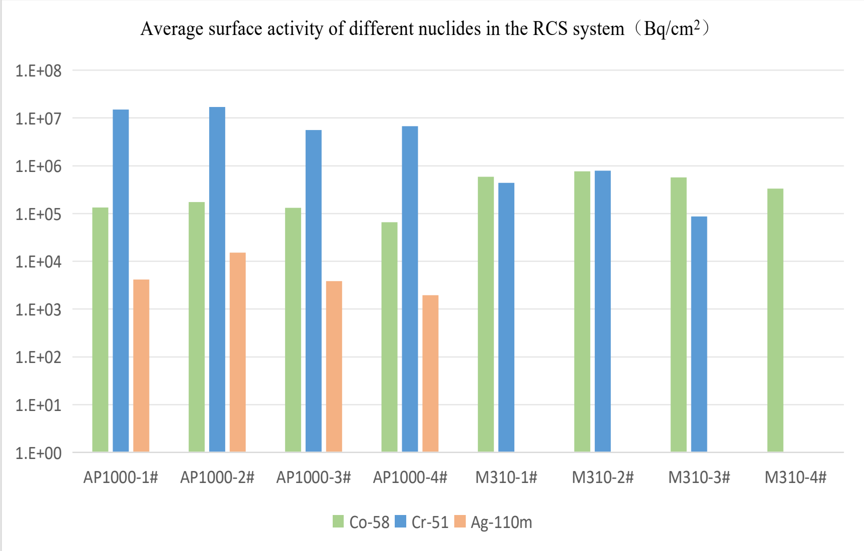

Fig. 7 Surface Activity of Main Deposited Nuclides in RCS Pipelines of Different Units

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

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5.1 Analysis for Co-58 Nuclide Deposition

- **In detail, the activity of Co-58 deposited in the RCS system pipelines of the four AP1000 units is between with 1.3×10^5~1.7×10^5 Bq/cm², and for the four M310 units are between with 3.3×10^5~7.6×10^5 Bq/cm².**
- **The main reasons why the surface deposition amount of Co-58 in the RCS system of AP1000 units is lower than that of M310 units are as follows:**

① **The steam generator heat transfer tubes of AP1000 units are made of Inconel 690 nickel-based alloy, while M310 units use Inconel 600 nickel-based alloy. As shown in Table 7, the nickel content in Inconel 690 is more than 58%, and that in Inconel 600 is 72%. Under the same contact area with the primary loop, the Co-58 nuclides caused by nickel corrosion activation products of M310 units are higher than those of AP1000 units;**

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

5.1 Analysis for Co-58 Nuclide Deposition

Table 7 Alloy Composition of Primary Loop Materials of AP1000 Unit and M310 unit

alloy	proportion of contact area with primary loop	alloy composition (%)							
		Fe	Cr	Ni	Zr	O	Sn	Mn	Co
Zr-4 or Zr (fuel rod cladding)	0.24	0.2	0.1	-	98.5	0.12	1.40	-	0.002
Inconel 600(SG heat transfer tube)	0.69	8	9	72	-	-	-	-	0.45
Inconel 690 (SG heat transfer tube)		9	29	58	-	-	-	0.5	0.15
Inconel 718	Low proportion	17	19	52.5	-	-	-	0.35	0.04
304 SS low Co	0.02	70	19	9	-	-	-	2	0.05
304 SS unlimited Co	0.05	70	19	9	-	-	-	2	0.15
Stellite （hard facing）	Low proportion	1	24.5	1	-	-	-	-	57

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5.1 Analysis for Co-58 Nuclide Deposition

② **Simplification design is a major character of the AP1000 units, which have fewer components, cable, and seismic building volume, and the radioactive equipment and components are reduced.** Compared with other type PWR NPPs, the number of valves has been reduced by 50%, pumps by 65%, and pipelines by 20%, which reduces the generation of corrosion activation products.

③ **In terms of operation, the technology of zinc injection to the primary circuit are adopted from beginning of the hot functional test to operation stage for AP1000 units.** After the primary circuit is saturated with zinc, the anti-corrosion property of the oxide film on the inner surface of the equipment was enhanced, which alleviated the corrosion and release of system materials, and reduced the source term of corrosion products.

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

5.2 Analysis of Cr-51 Nuclide Deposition

- **From Fig.7, we can see that the surface activity of Cr-51 nuclide deposition in the RCS system pipelines of the four AP1000 units is between 6.0×10^6~1.7×10^7 Bq/cm², much higher than that of the four M310 units, which is between 8.0×10^4~7.9×10^5 Bq/cm².**
- **The main reasons are as follows:**
  - **① As shown in Table 7, the Cr content in Inconel 690 alloy used for the SG heat transfer tubes of AP1000 units (29%) is higher than that in Inconel 600 alloy of M310 units (9%);**
  - **② The outer surface of the control rod cladding of AP1000 units adopts Cr plating technology, and the control rods are irradiated by high neutron flux in the core for a long time during the entire power operation period, so Cr-50 has a higher probability of being activated into Cr-51.**

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

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5.3 Analysis of Ag-110m Nuclide Deposition

- **From Fig.7, we can see that AP1000 units have higher Ag-110m nuclide deposition compared with M310 units.**
  - **Ag-110m nuclide deposition occurs in the RCS system pipelines of the four AP1000 units, and the surface activities is between with 4.0×10^3-1.5×10^4 Bq/cm^2. It is one of the major source terms of radioactive corrosion products in the primary circuit of Haiyang NPP.**
  - **All the surface activities of Ag-110m in the DSTs of RCS pipelines of the four M310 units are below the detection limit.**

5 General Analysis for Average Surface Activity Level of Deposition Nuclides

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5.3 Analysis of Ag-110m Nuclide Deposition

- A special investigation have been conducted, targeting the equipment and components with silver-containing material in the primary circuit, and confirmed that the silver-containing gaskets on the tube side of the RNS heat exchanger are the main source of nuclide Ag-110m. Nuclide Ag-109 containing in the gaskets of the RNS heat exchanger dissolved into the coolant and was being activated in the core to become Ag-110m.
- After these gaskets of Unit 1 have been replaced with silver-free gaskets during the HY104 outage, the activity of Ag-110m decreased from 77ppb to 6ppb, and finally dropped below the detection limit (4ppb) with the continuous operation of oxidation purification. Later, same replacement work was carried out for Unit 2. The high content problem of Ag-110m in the radioactive liquid effluent of the units was solved.

6 Conclusion

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① **The average level of Co-58 surface deposition activity on the inner walls of RCS system pipelines in AP1000 units is lower, comparable to those in M310 nuclear power units, and the average levels of surface activities of Cr-51 are higher. The reason are as follows:**

  a. **the lower nickel content in Inconel 690, fewer primary loop equipment in AP1000 units, and the adoption of zinc addition technology.**

  b. **the high chromium content in Inconel 690 and the adoption of chromium plating technology for the control rods of AP1000 units;**

② **The main reason with higher Ag-110m nuclide in AP1000 units is that the silver in the gaskets on the tube side of the RNS heat exchanger enters the core with the coolant, activated, and deposited in other systems. All the silver-containing gaskets of the RNS heat exchangers in both units with silver-free gaskets have been replaced , the problem of Ag-110m was solved.**

Thank you for your attention!

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