

Improvement of Fission Gas Analyzer

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1. Background

Reactor Fuel Examination Facility



A view of the facility



Operation room

OverView

- RFEF is one of the largest hot laboratories in Japan.
- RFEF performs PIEs on LWR spent fuels and materials to evaluate the safety and reliability.
- Safety research on the high burnup fuel is carried out as part of the PIEs at RFEF.

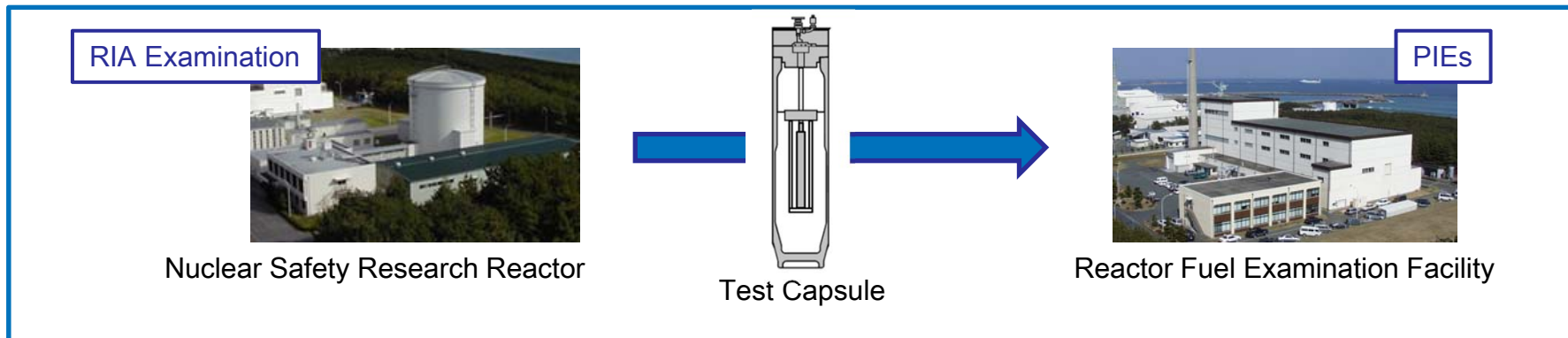
1. Background

Burnup of nuclear fuel has been extended for the beneficial utilization.

- High burnup fuel must be evaluated to keep their safety.



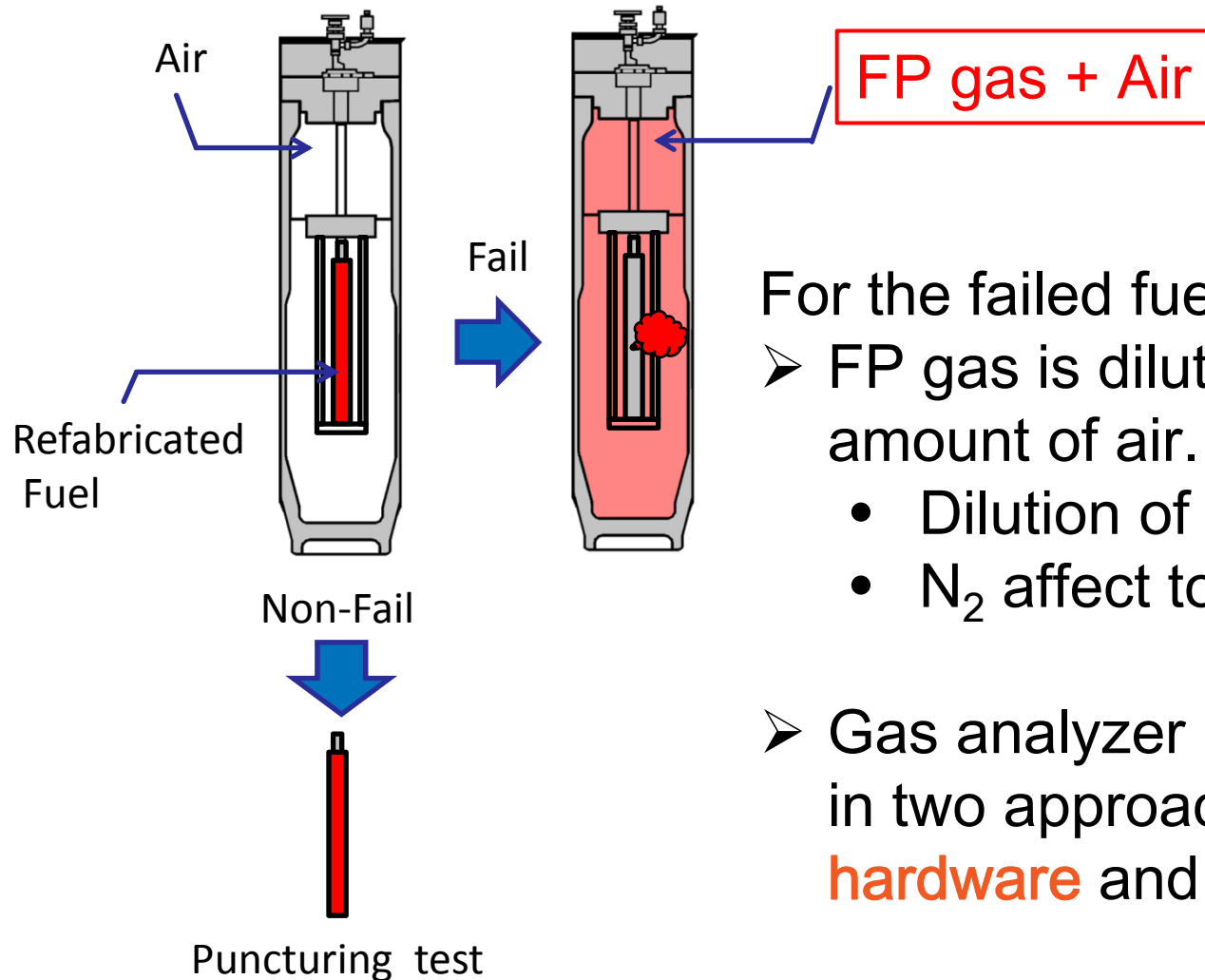
Fuel behavior during RIA condition is studied as a part of the safety research of High burnup fuel.



GC and the GC-MS were used to measure the volume and the isotopic ratio of Kr and Xe.

1. Background

FP gas is collected after RIA examination.



For the failed fuel

- FP gas is diluted by the large amount of air.
 - Dilution of Kr
 - N₂ affect to the Kr measurement
- Gas analyzer has been improved in two approaches, **hardware** and **software**.

2. Hardware approach

1. Background

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3. Software approach

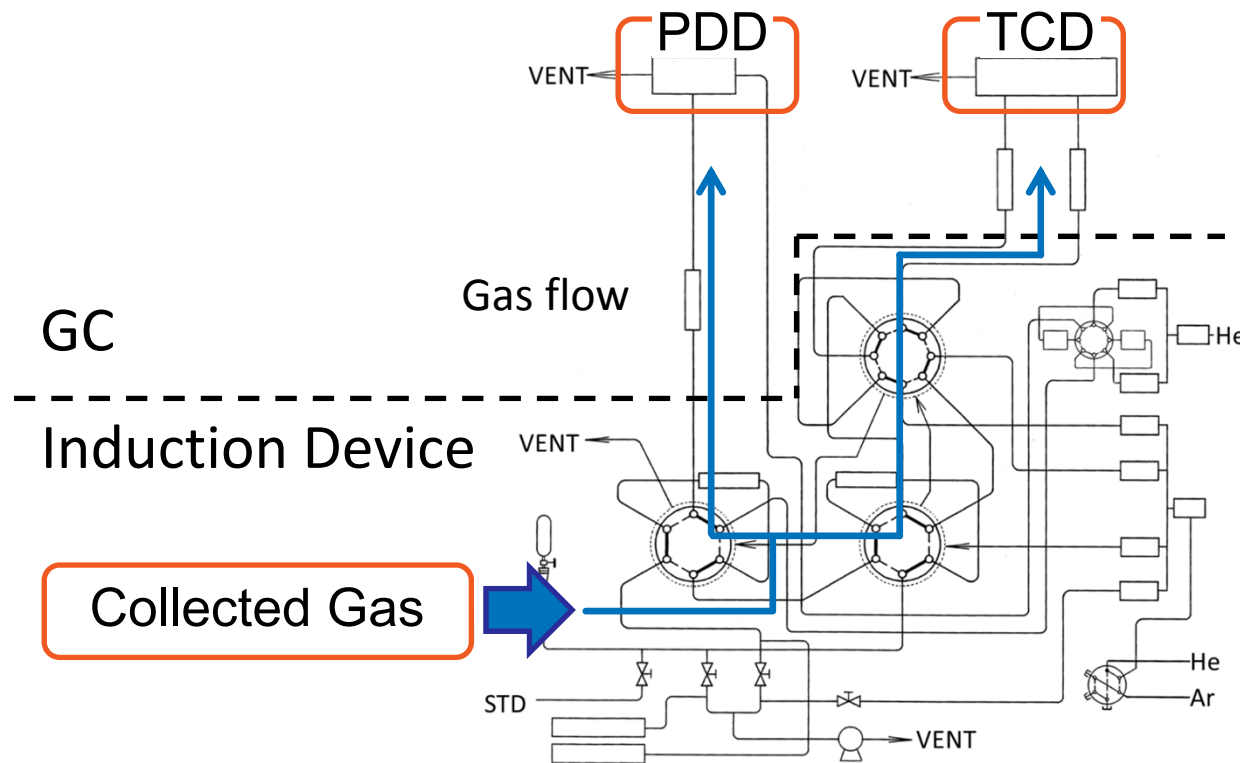
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4. Summary

2.1 Improvement of apparatus

For the measurement of dilute Kr gas.

- High sensitivity detector **PDD** has been added.

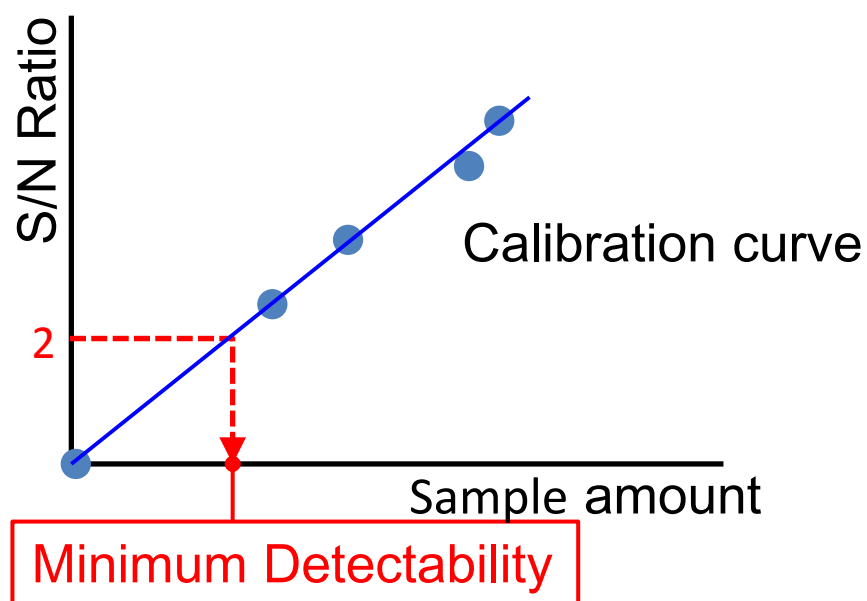


Schematic Diagram of the Fission gas Analyzer

2.2 Evaluations

Determination of the Minimum Detectability^[1]

- Standard gas of 0.5 Kr% is measured.
- Varying gas volume and calibration curve of S/N ratio is calculated.
- Determine the Minimum detectability from calibration curve on $S/N = 2$.



[1] IUPAC Analytical Chemistry Division 9.2.4.4

2.3 Results

Results of the determinations

Component	Detector	Minimum Detectability (ppm)
Kr	TCD	5.27×10^{-1}
	PDD	1.05×10^{-3}

Minimum Detectability of TCD and PDD.



PDD has **500 times higher sensitivity** compared with TCD.

3. Software approach

1. Background

2. Hardware approach

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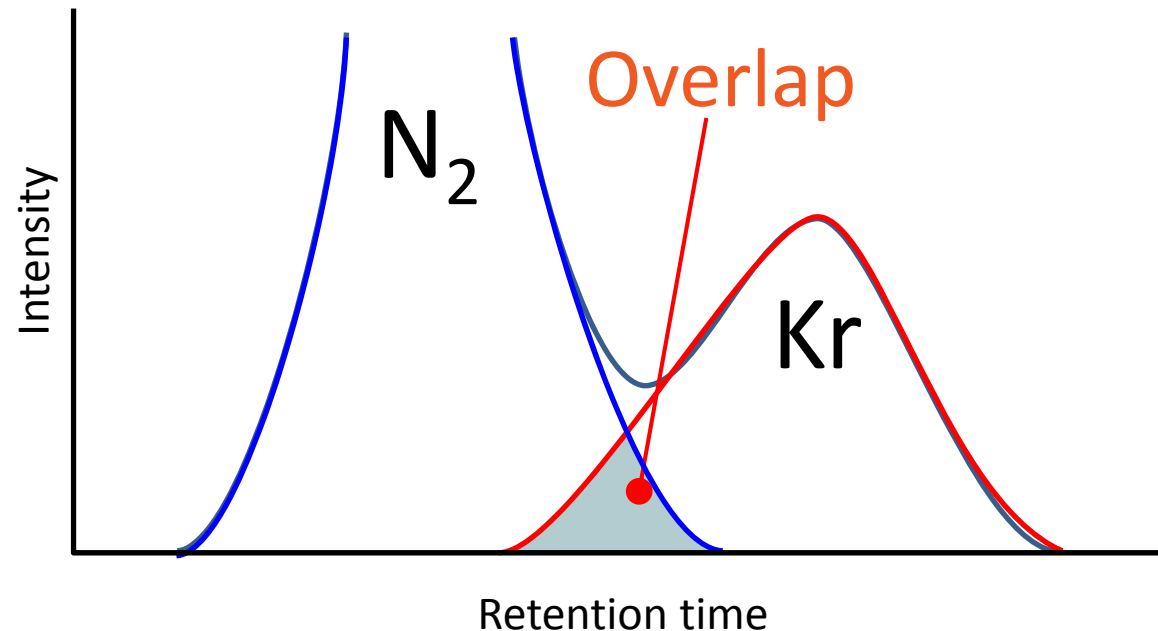
3. Software approach

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4. Summary

3.1 Peak analysis

For the Peak overlap of Kr and N₂

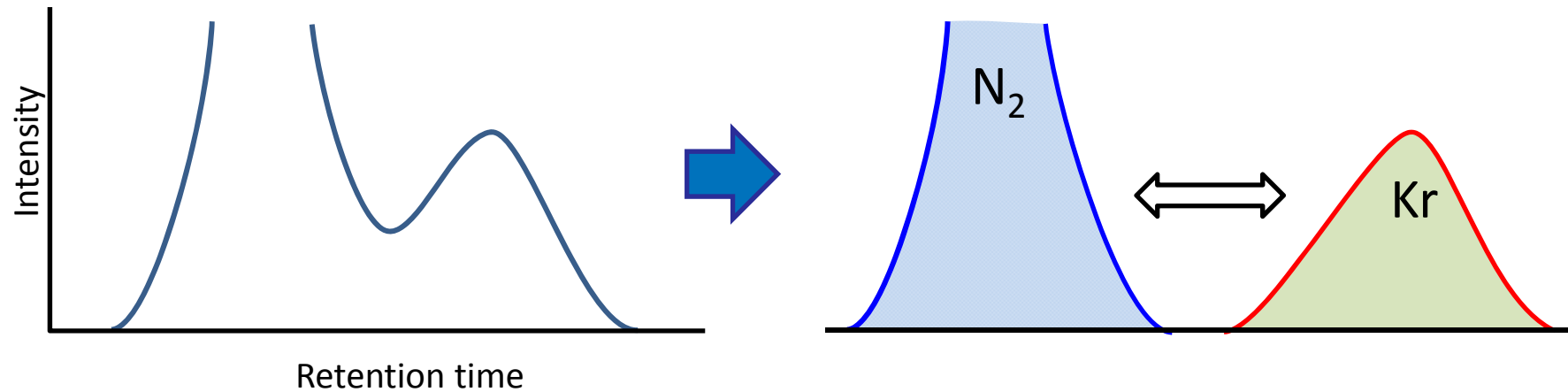


This overlap was caused by the **closed retention times** of the Kr and N₂.

- Peak overlap of Kr and N₂ affects measurement accuracy of Kr.

3.1 Peak analysis

Separate overlapped peak

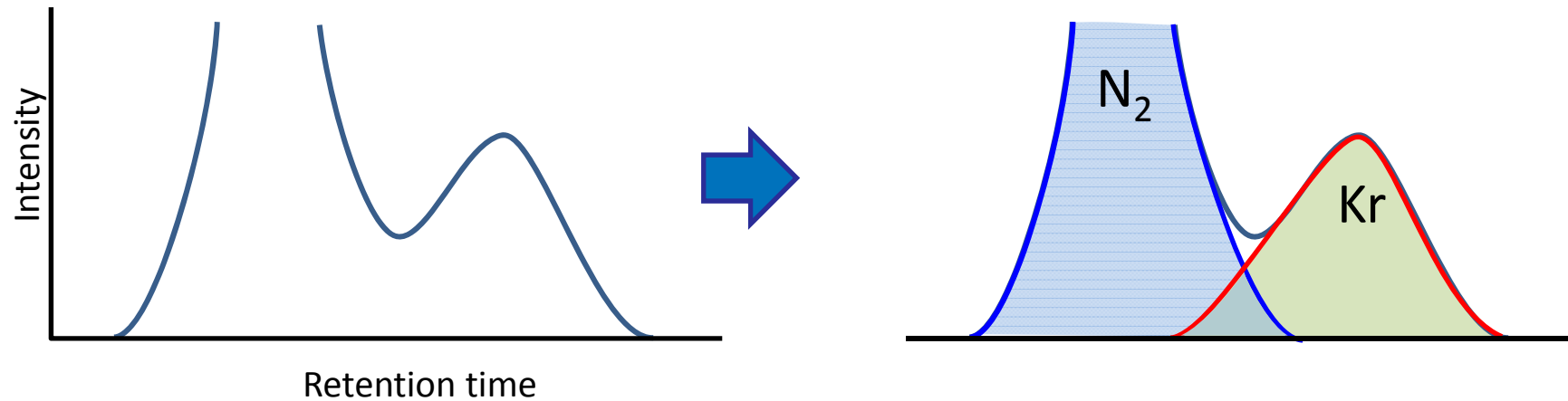


Optimization of measurement condition

- Longer measurement time
 - Large amount of gas
- Prevent these problems

3.1 Peak analysis

Peak analysis



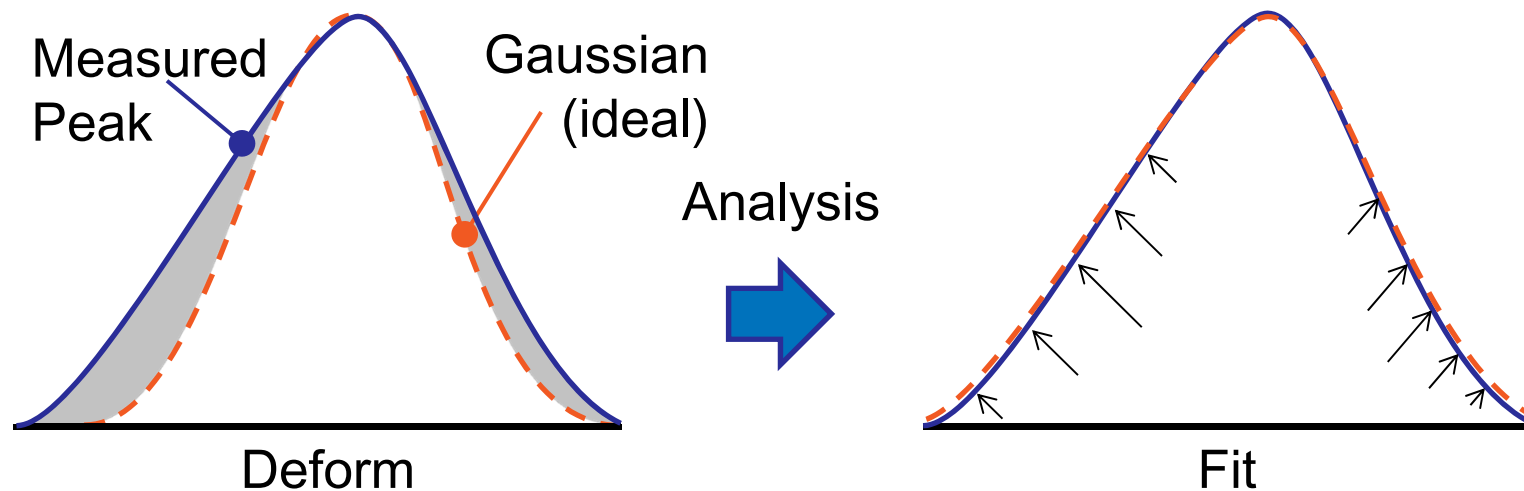
Deconvolute the overlapped peak

- Saving measurement time
 - Applicable for limited amount of gas
- Peak analysis was applied to overlapped peaks.

3.2 Evaluations

Peak function

- Peak shape is the Gaussian distribution in the ideal condition.
- **Deformed** from the ideal shape as leading and tailing.



Four different types of Gaussian Functions were compared to figure out the best fit function.

3.2 Evaluations

Peak function used for the evaluation.

Exponentially Modified Gaussian Function^{[3],[5]}

Empirically Transformed Gaussian Function^{[4],[5]}

Generalized Exponentially Modified Gaussian Function^{[3],[5]}

Polynomial Modified Gaussian Function^{[3],[5]}

- Each Sum of Squared Residuals was obtained by analysis of measurement peaks and estimated peaks.
- The best fits are obtained by minimizing the SSR.

[3] P.Nikitas J Chromatogr. A 912 (2001) 13

[4] J. Li. Anal. Chem. 69 (1997) 4452

[5] J. Li. J Chromatogr. A 952 (2002) 63

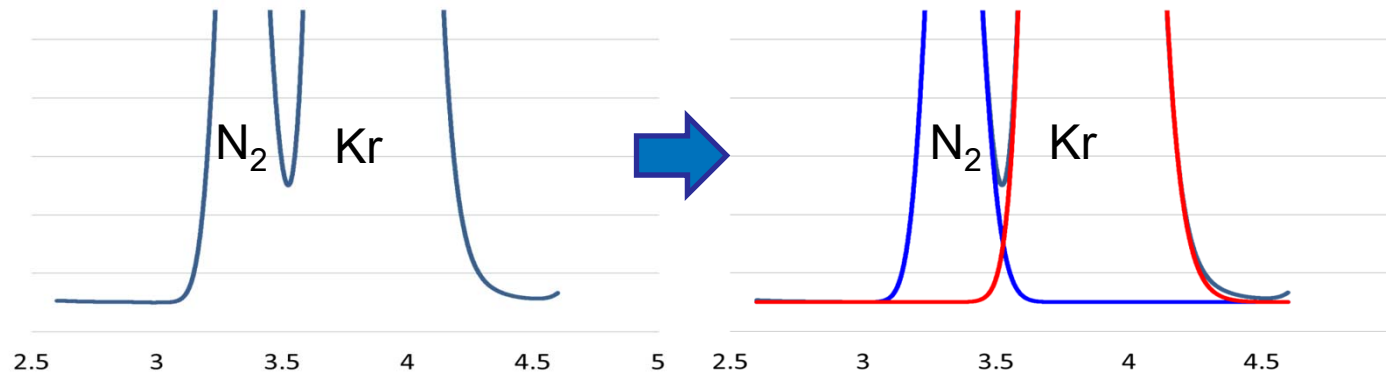
3.3 Results

Results of evaluation

- **ETG function** gave the **minimum SSR** values.

Function	EMG	ETG	GEMG	PMG2
SSR	4.7E+10	3.74E+9	4.37E+10	1.46E+12

Example of deconvolute Kr and N₂.



- Overlapped peaks of Kr and N₂ are deconvoluted **with ETG Function.**

4. Summary

Gas analyzer has been improved in two approaches, **hardware** and **software**.

Hardware approach

- PDD reached 50 times higher minimum detectability of Kr than TCD.

Software approach

- Overlapped peaks of Kr and N₂ are deconvoluted with ETG function.

Increasing of measurement accuracy is expected for the gas analysis of failed fuel on the RIA examination.