



LSC 2017

Advances in Liquid Scintillation Spectrometry

# Standardization of Na-22 by CNET method

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# Outline

- 1** Sample preparation
- 2** Process of standardization
- 3** Results
- 4** Discussion 1: Influence of tSIE
- 5** Discussion 2: Influence of kB
- 6** Conclusions

# Preparation of LS Samples

## ● Sodium -22

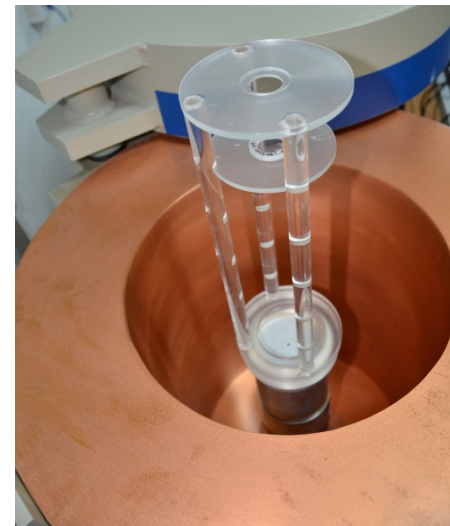
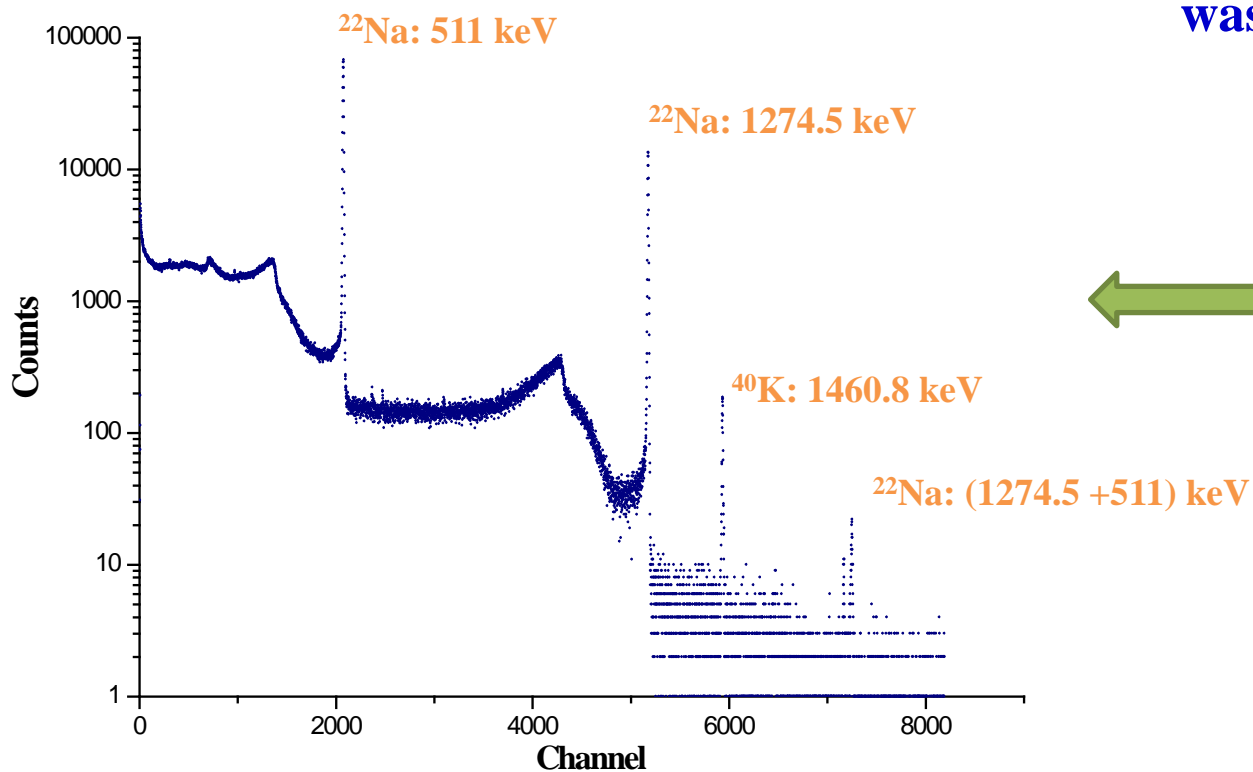
- ❖  **$^{22}\text{Na}$  master solution:** Activity concentration of about 11.6 MBq/g
- ❖ **Carrier:** about 25 $\mu\text{g/g}$  NaCl in HCl (0.1 mol/L)
- ❖ **Quenching agent:** Nitromethane ( $\text{CH}_3\text{NO}_2$ )
- ❖ **Scintillator:** Ultima Gold LLT

## ● Tritium (tracer)

- ❖ A set of  $^3\text{H}$  samples were prepared in the same way

# Impurity check of $^{22}\text{Na}$ solution

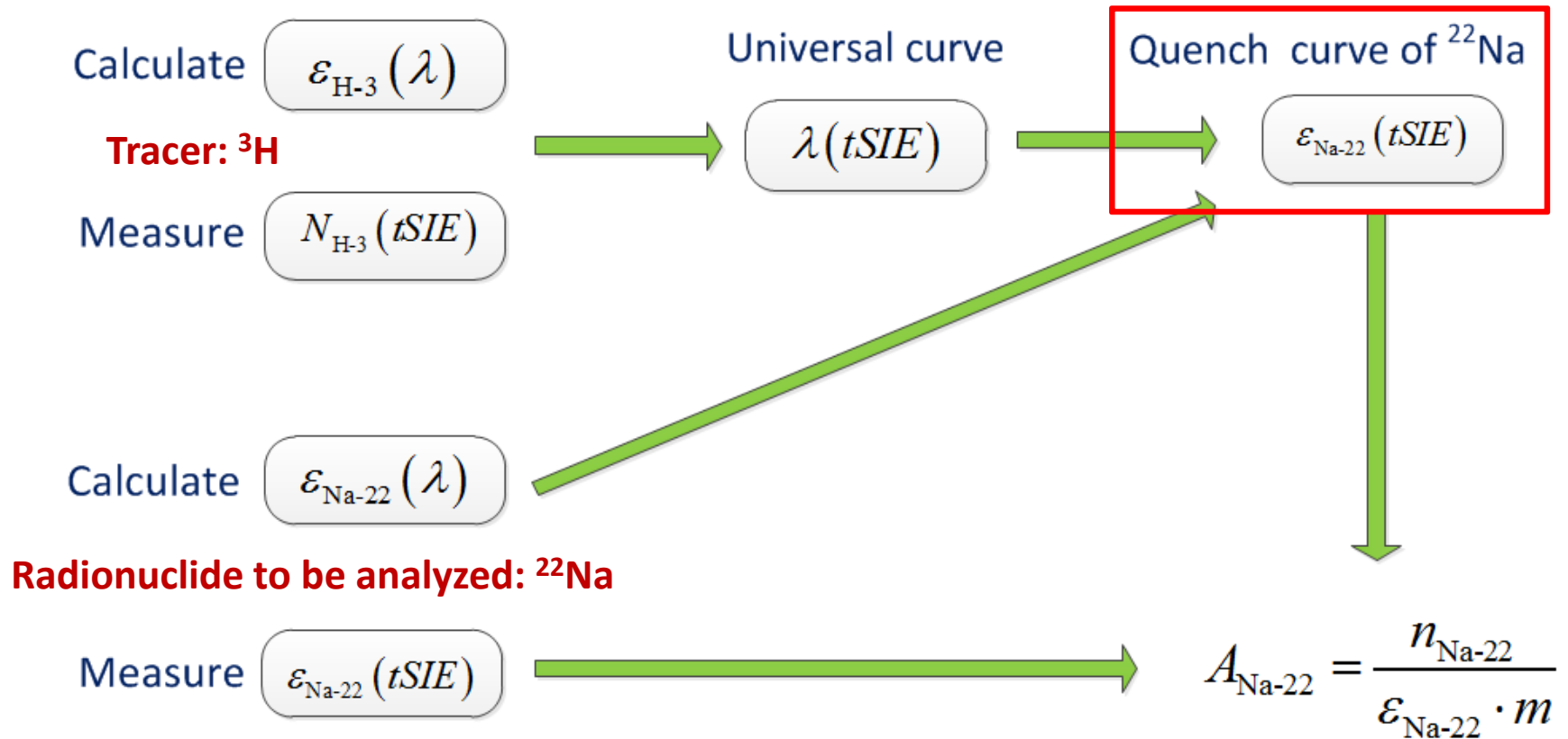
No  $\gamma$ -emitting impurity was detected.



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## The procedure of the CIEMAT/NIST Method



## 2.1 Measurement details

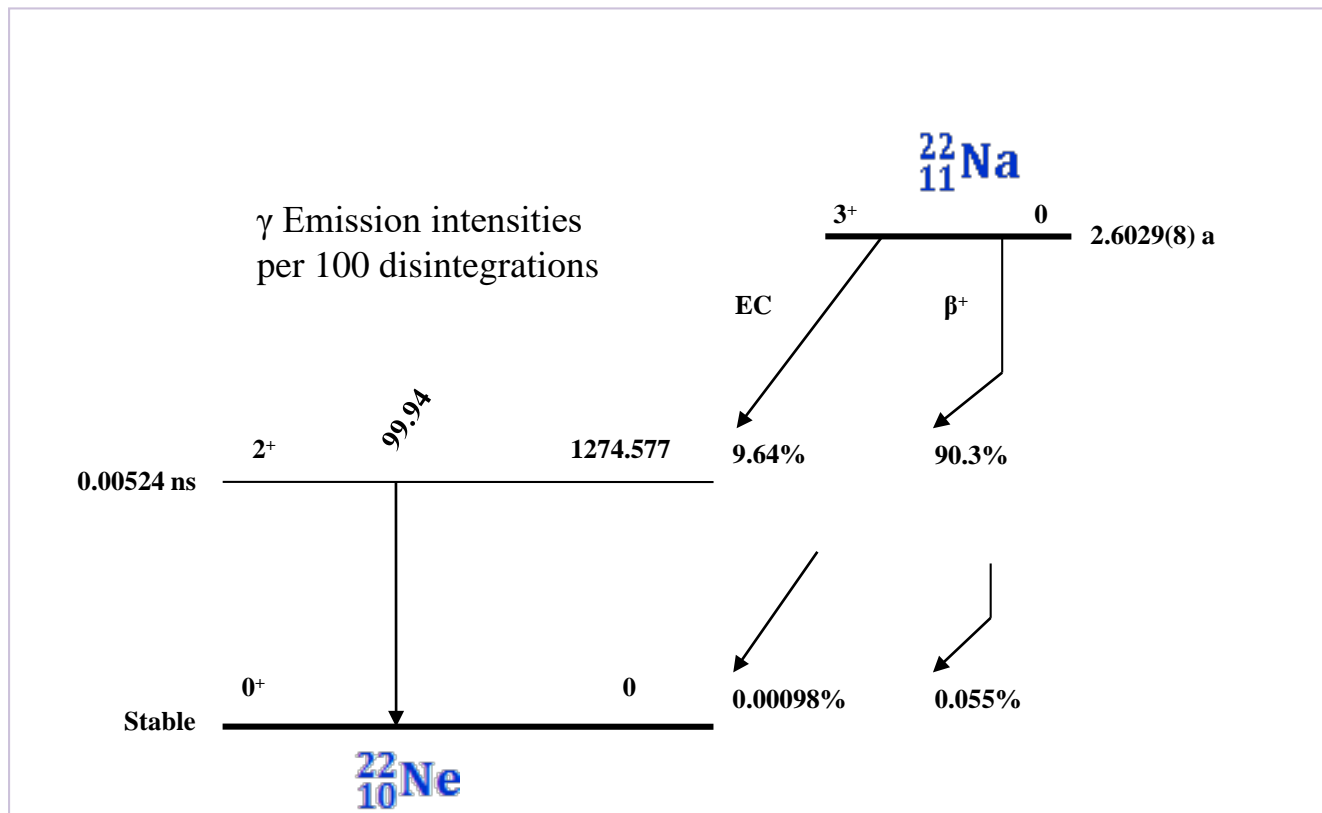
LS spectrometers: **PerkinElmer Tri-Carb 3100TR**

- Quench indicating parameter : tSIE
- 5 cycles of 20 minutes per source
- At least  $10^6$  counts for each source



## 2.2 Efficiency calculation

### ❖ Decay scheme of Na-22





## 2.2 Efficiency calculation

# Decay data of Na-22

### ❖ Nuclear data of Na-22 (Cited from DDEP)

#### 2 Nuclear Data

$T_{1/2}(^{22}\text{Na})$  : 2,6029 (8) a  
 $Q^+(^{22}\text{Na})$  : 2843,02 (21) keV

#### 2.1 Electron Capture Transitions

	Energy keV	Probability × 100	Nature	lg <i>ft</i>	$P_K$	$P_L$
$\epsilon_{0,1}$	1568,44 (21)	9,64 (9)	Allowed	7,41	0,923 (4)	0,077 (4)
$\epsilon_{0,0}$	2843,02 (21)	0,00098 (25)	Unique 2nd Forbidden	14,91		

#### 2.2 $\beta^+$ Transitions

	Energy keV	Probability × 100	Nature	lg <i>ft</i>
$\beta_{0,1}^+$	546,44 (21)	90,30 (9)	Allowed	7,4
$\beta_{0,0}^+$	1821,02 (21)	0,055 (14)	Unique 2nd Forbidden	14,9

#### 2.3 Gamma Transitions and Internal Conversion Coefficients

	Energy keV	$P_{\gamma+ce}$ × 100	Multipolarity	$\alpha_K$ ( $10^{-6}$ )	$\alpha_T$ ( $10^{-6}$ )	$\alpha_\pi$ ( $10^{-5}$ )
$\gamma_{1,0}(\text{Ne})$	1274,577 (7)	99,94 (13)	E2	6,36 (9)	6,71 (9)	2,34 (3)

## 2.2 Efficiency calculation

### Decay data of Na-22

- ❖ Atomic data of Na-22 for efficiency calculation using the KLM rearrangement model

Name of parameter	Symbol	Value	Reference
Fluorescence yield	$\omega_K$	0.0152	Bambynek-1972
	$\omega_L$	0.0001	
Relative probabilities of the K Auger electrons (sum=1)	$P_{KLL}$	1	E. Schönfeld and H. Janßen-1996
	$P_{KLM}$	0	
	$P_{KMM}$	0	
Relative probabilities of the L Auger electrons (sum=1)	$P_{LMM}$	0	
Relative probabilities of the K X-rays (sum=1)	$P_{KL}$	1	Firestone (1998)
	$P_{KM}$	0	
Relative probabilities of the L X-rays (sum=1)	$P_{LM}$	0	Firestone (1998)
Average energies of the Auger electrons	$E_{KLL}$	0.8268	Larkins (1977)
	$E_{KLM}$	0.8484	
	$E_{KMM}$	0.8701	
	$E_{LMM}$	0.0216	Larkins (1977)
Average energies of the X-rays	$E_{KL}$	0.8486	Firestone (1998)
	$E_{KM}$	0.8701	
	$E_{LM}$	0.0216	Firestone (1998)

## 2.2 Efficiency calculation

### Code and parameters

$^3\text{H}$

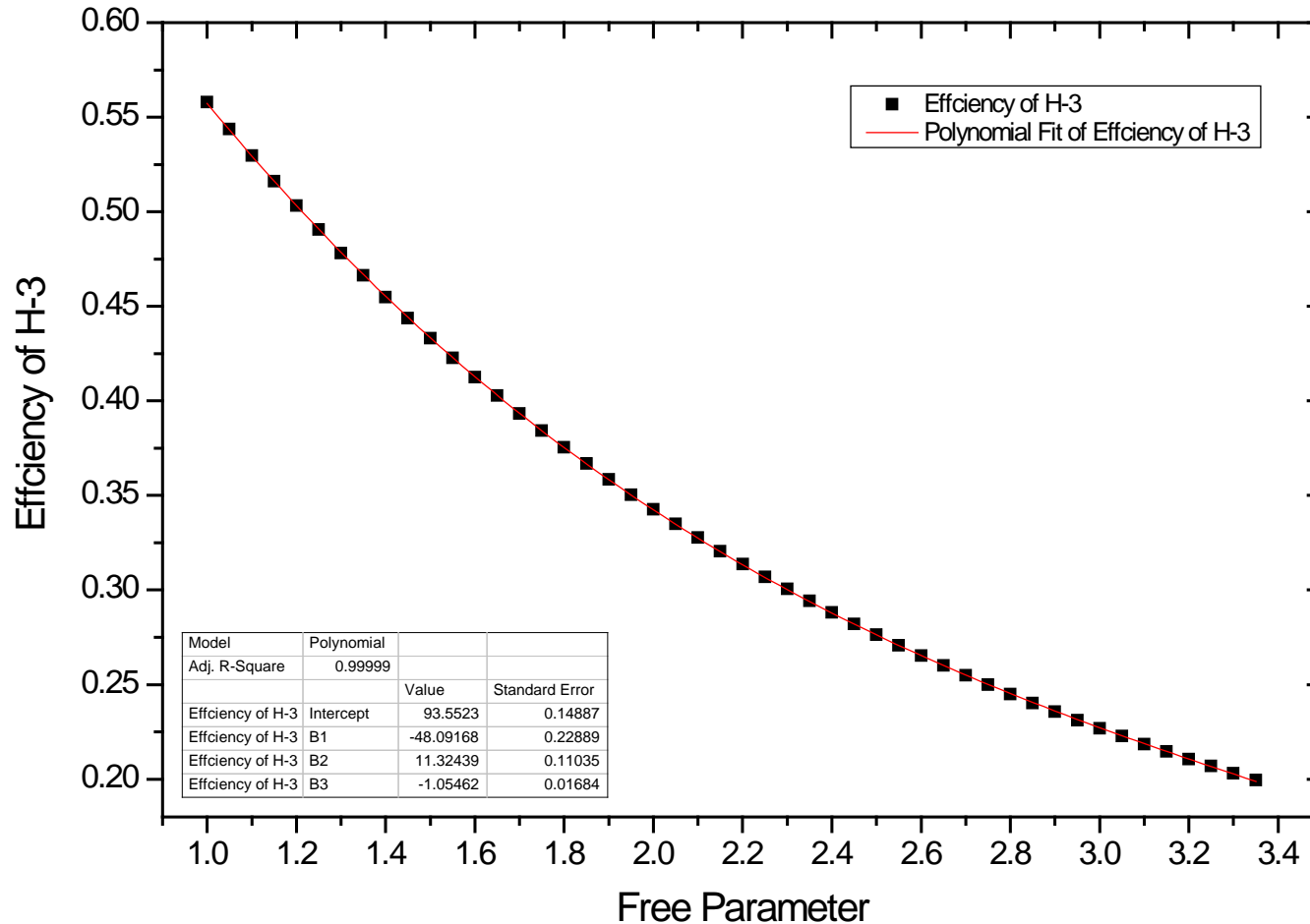
- Decay type : **Pure Beta decay**
- Code: **CN2003**

$^{22}\text{Na}$

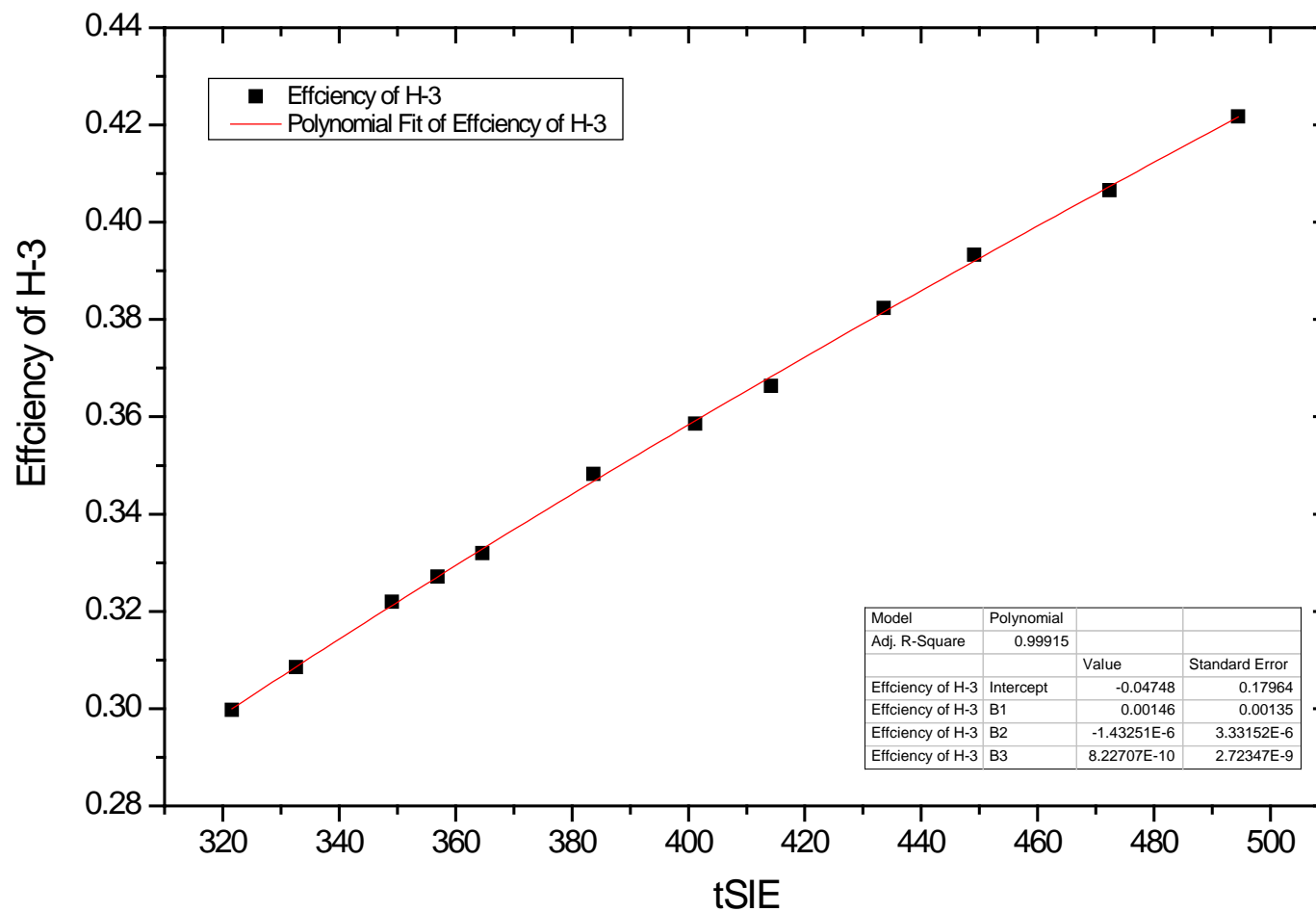
- Decay type :  **$\beta^+$ /EC decay**
  - Code: **CN2003**
  - Electron-capture transition atomic rearrangement model: **KLM**
- **Computation of ionization quenching function**
    - (1) The ionization quenching parameter:  $KB=0.075$
    - (2) The stopping power values(default)
    - (3) Scintillator composition, density (modify to Ultima Gold LLT)

# The procedure to obtain the counting efficiency for Na-22

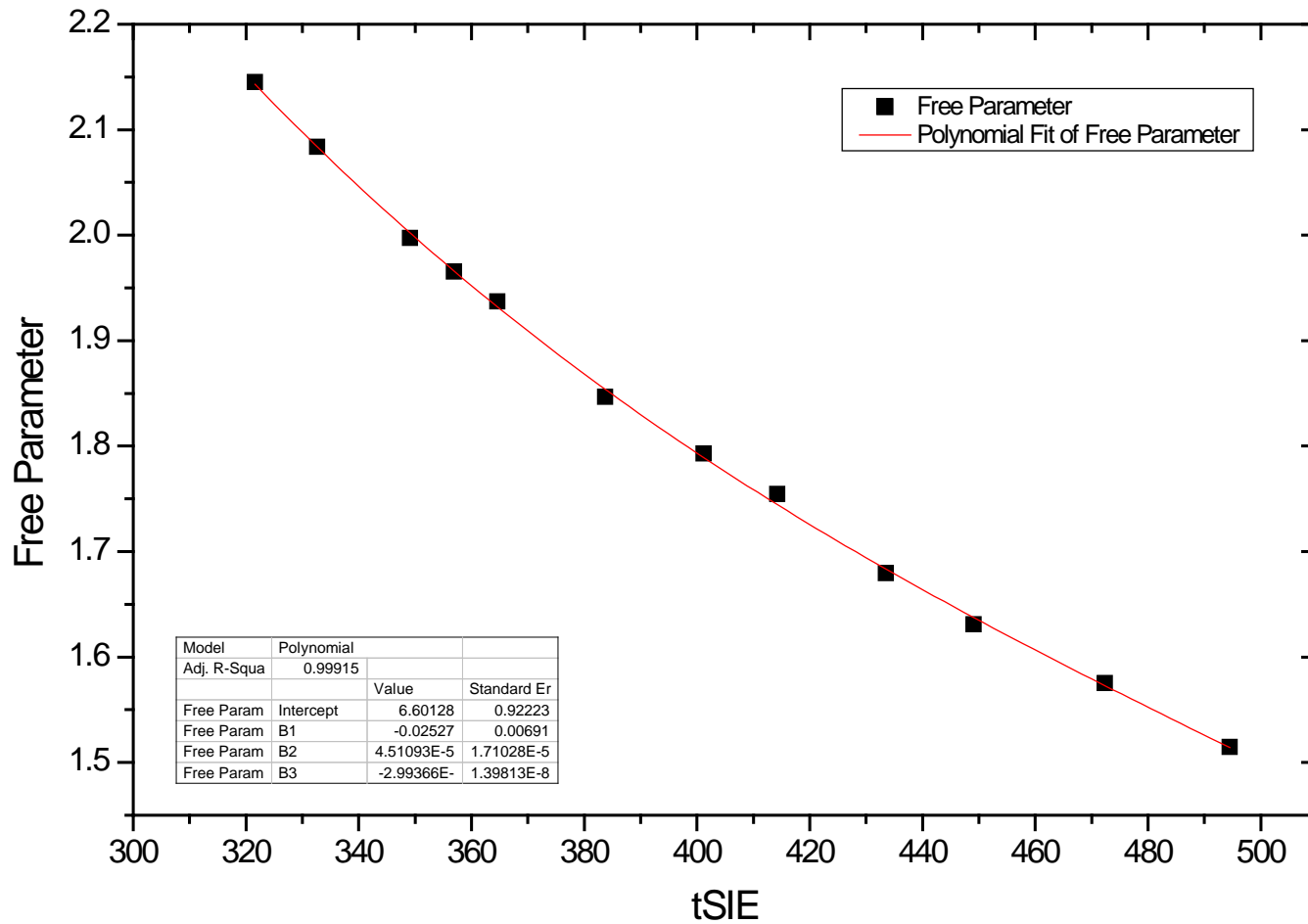
## (a) The theoretical counting efficiency curve of $^3\text{H}$



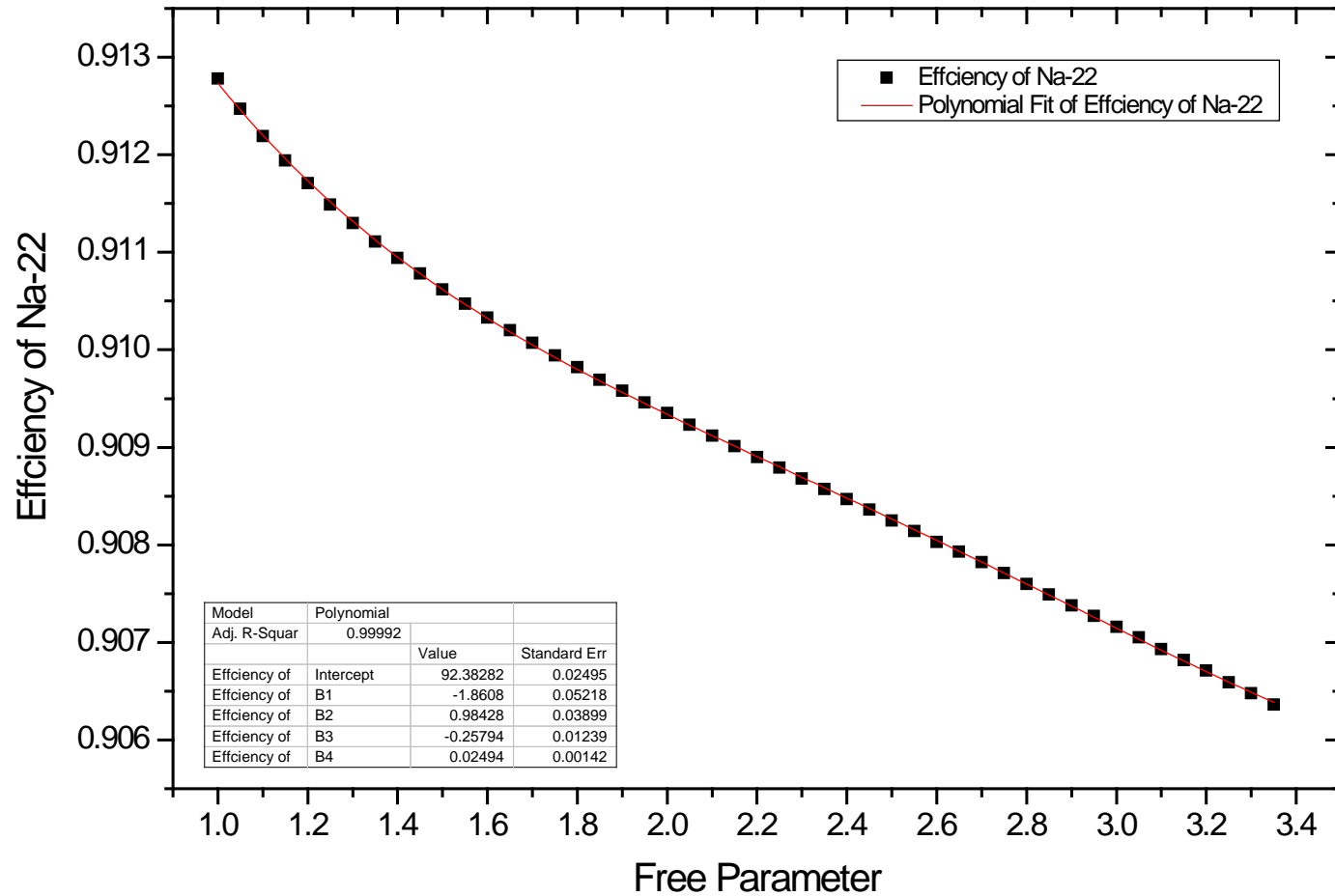
## (b) Experimental quench-correction curve of $^3\text{H}$



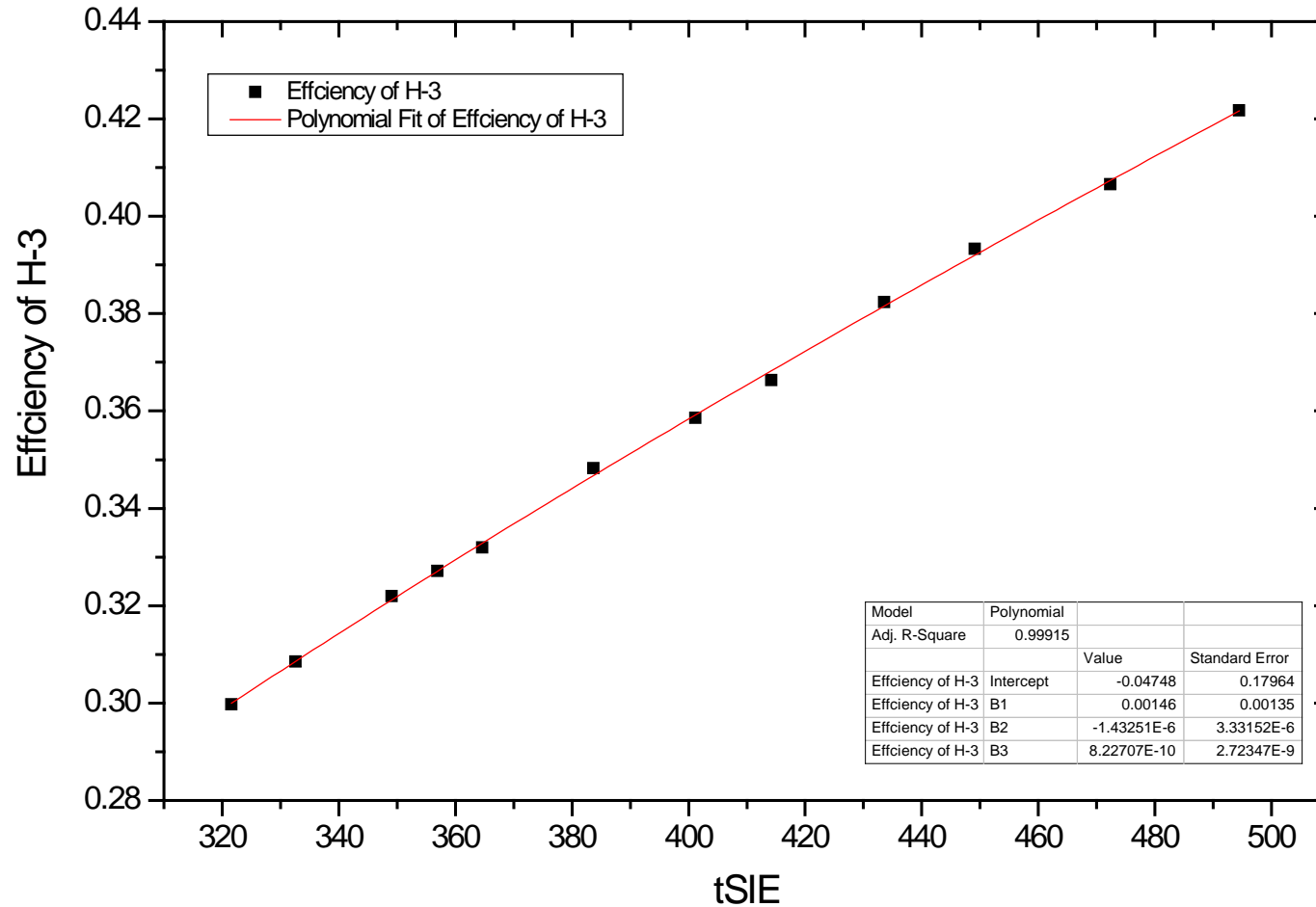
## (c) “Universal” curve



## (d) The theoretical counting efficiency curve of $^{22}\text{Na}$



## (e) Computed quench correction curve of $^{22}\text{Na}$





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Discussion 1: Influence of tSIE and kB

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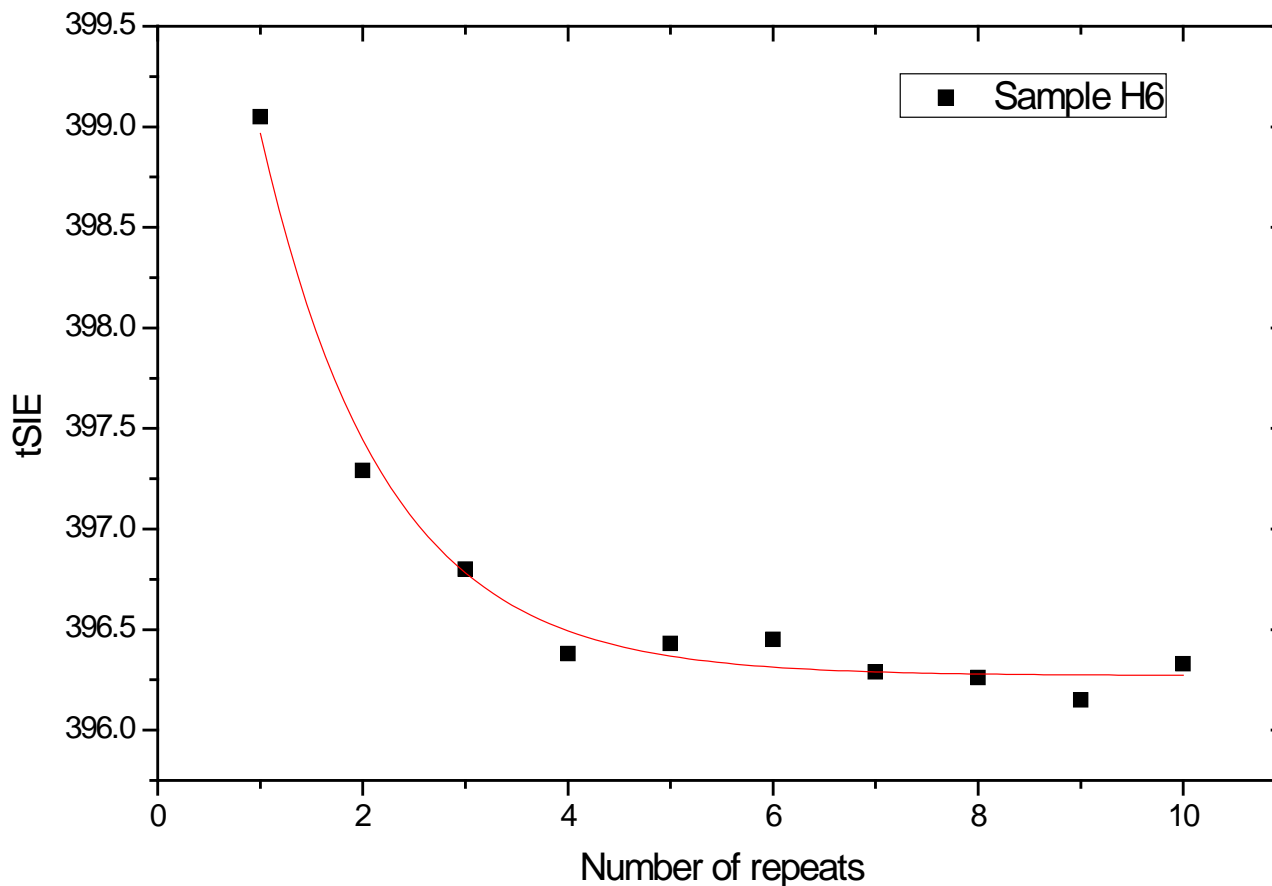
Discussion 2: Influence of kB values

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Conclusions

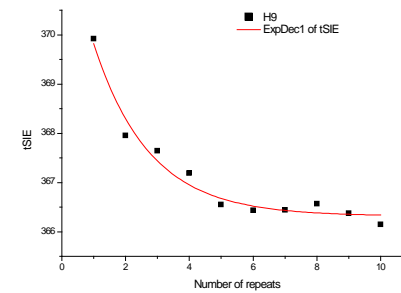
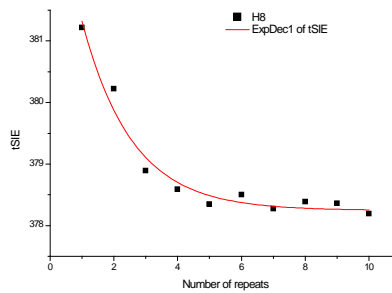
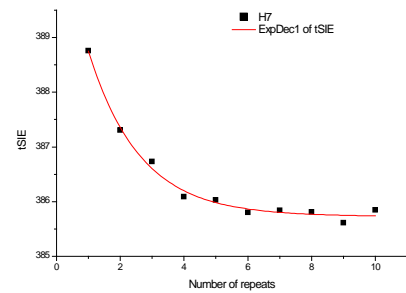
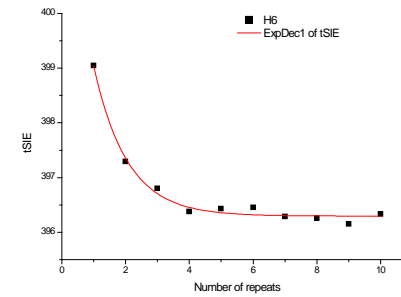
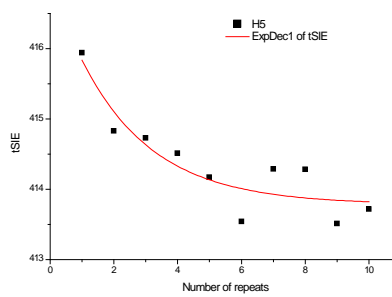
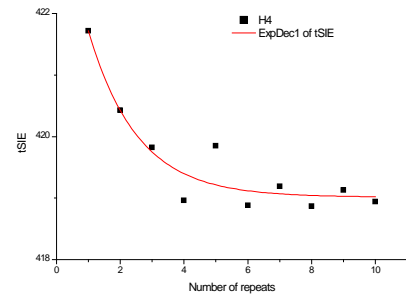
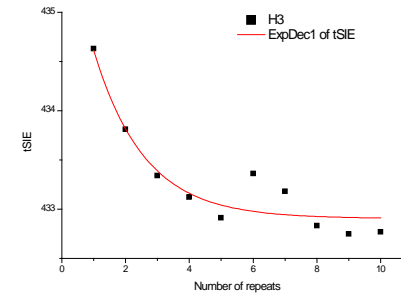
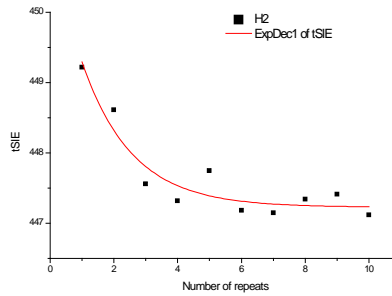
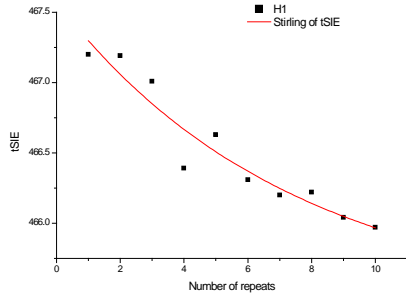
## 3.1 The stability of the counter

### a. The stability of tSIE in the measurement



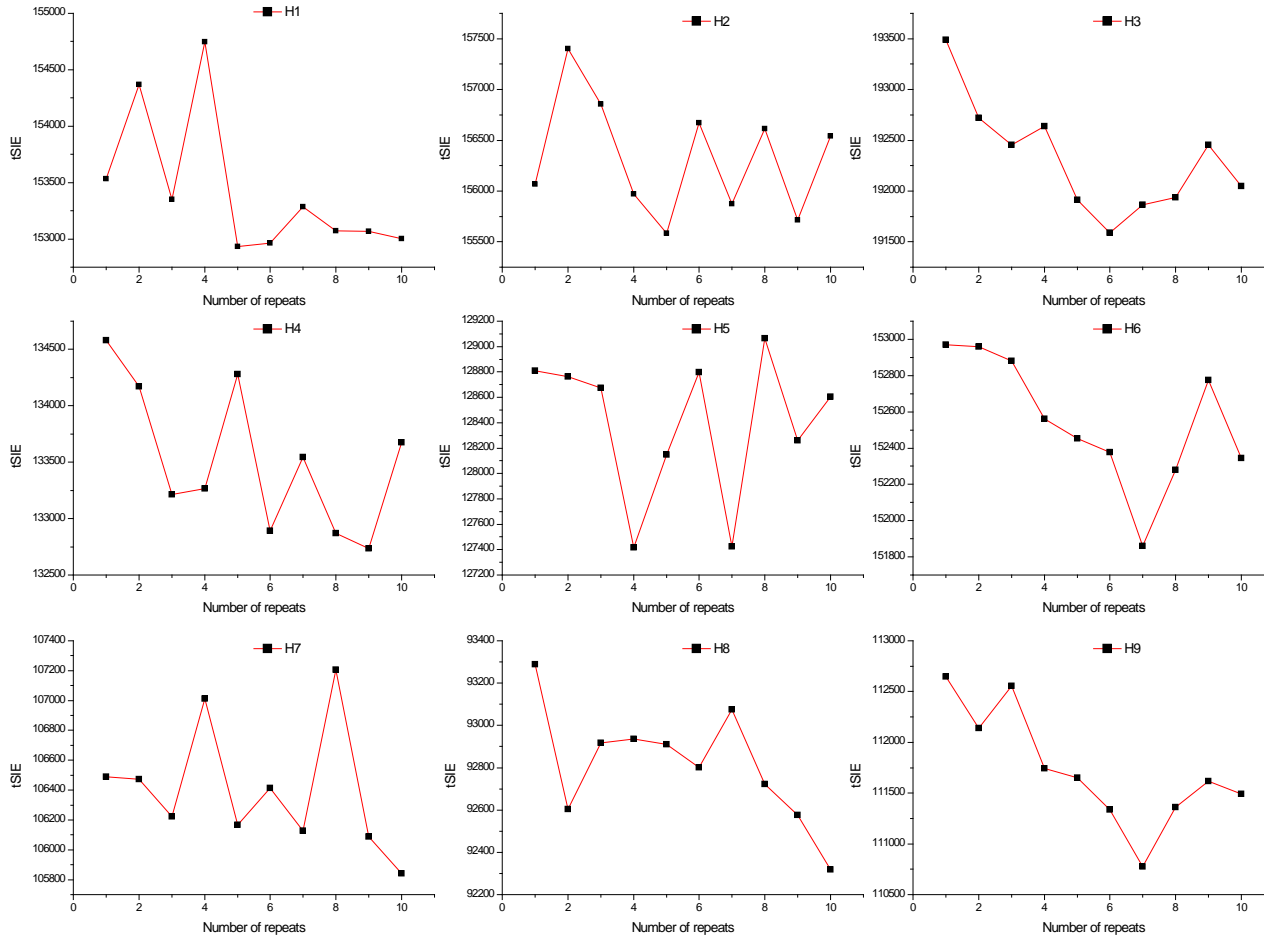
# 3.1 The stability of the counter

## a. The stability of tSIE in the measurement



# 3.1 The stability of the counter

## b. The stability of counting rates in the measurement

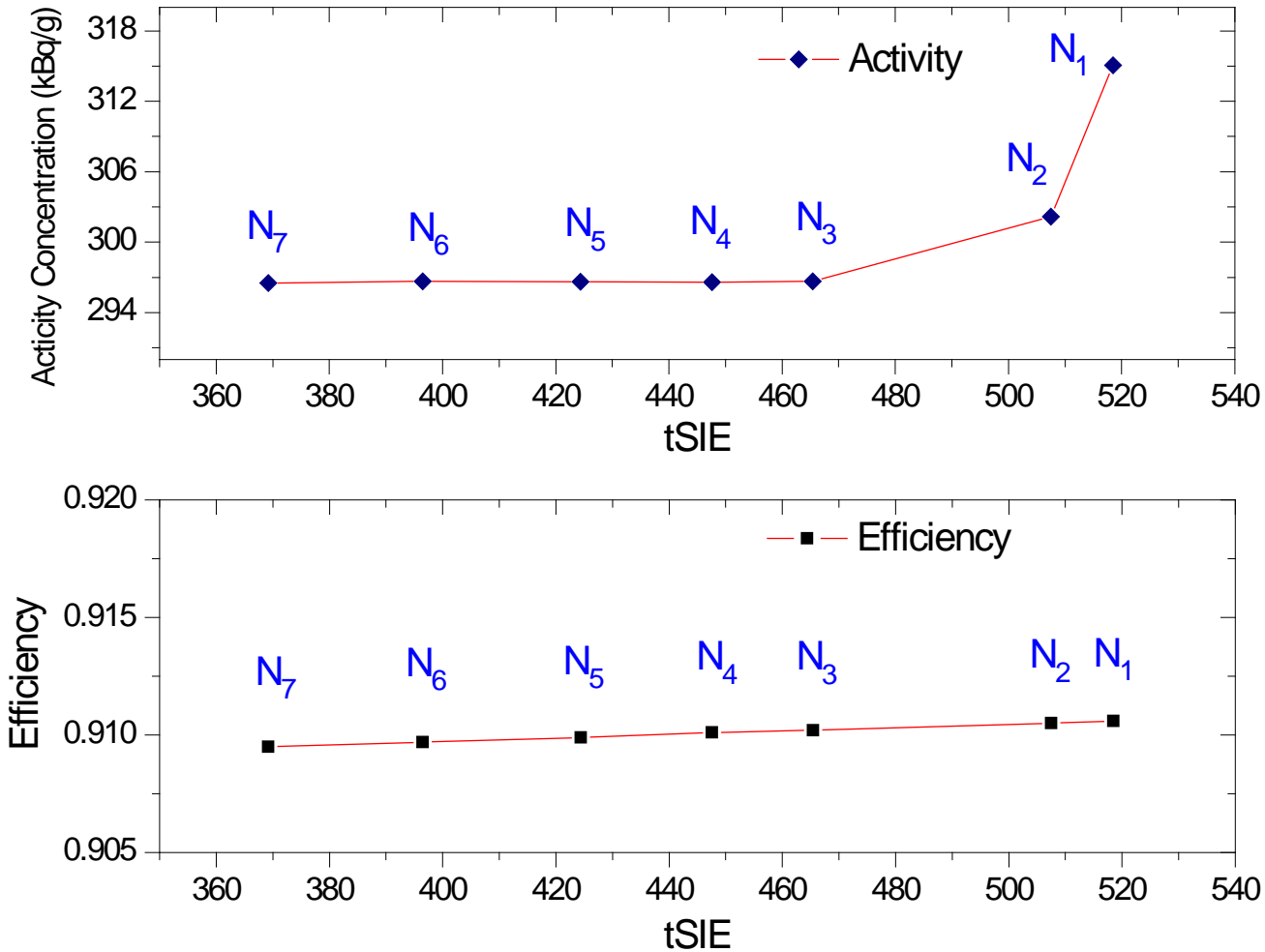


## 3.2 Results and uncertainty

### The activity concentration of $^{22}\text{Na}$

Sample No.	tSIE	Efficiency	Counting rate( $\text{min}^{-1}$ )	Mass(mg)	A( $\text{kBq}\cdot\text{g}^{-1}$ )
Blank	491.8	---	258	---	---
N1	518.5	0.9106	550407	31.96	315.07
N2	507.5	0.9105	601448	36.42	302.17
N3	465.4	0.9102	386208	23.82	296.68
N4	447.6	0.9101	360278	22.23	296.58
N5	424.4	0.9099	509733	31.46	296.62
N6	396.5	0.9097	373332	23.04	296.66
N7	369.2	0.9095	341983	21.12	296.51
Relatively standard deviation of N <sub>3</sub> to N <sub>7</sub>					0.02%

## 3.2 Results and uncertainty



## 3.2 Results and uncertainty

<b>Uncertainty components*, in % of the activity concentration, due to:</b>		
<b>Factor</b>	<b>U(a)/a in%</b>	<b>Remarks</b>
Counting statistics	0.05	Standard deviation of mean of 5 samples, including the source dispersion.
Weighing	0.09	
Background	0.03	
Dead time	0.10	
Quenching	0.04	
Tracer(H-3)	<0.01	
Input parameters and statistical model	0.19	Standard deviation of efficiencies with changes of input parameters
Ionization quenching and kB model	0.03	Calculated from the maximum entropy principle, considering the changes of kB values from 0.006 to 0.014 cm·MeV <sup>-1</sup>
<b>Combined uncertainty</b>	<b>0.23</b>	quadratic sum of all uncertainty components)

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Conclusion



## ➤ The problem of quench indicating parameter tSIE

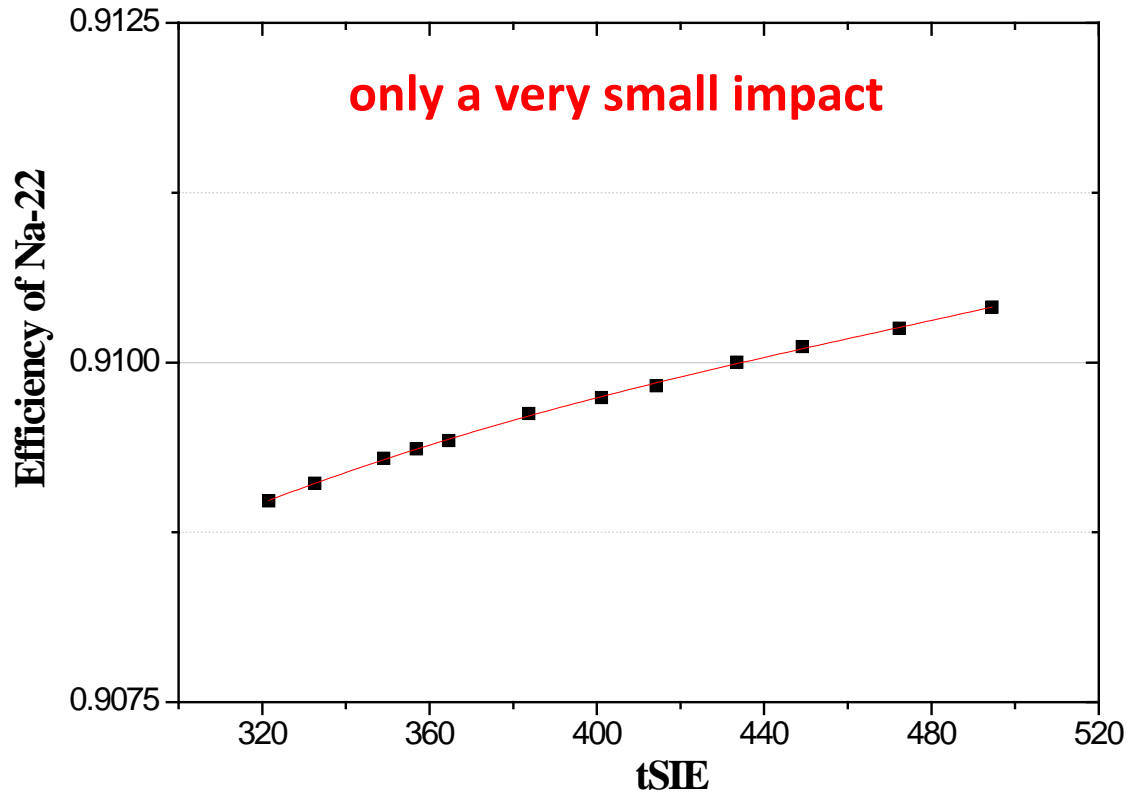
(1) The tSIE seems to be affected by the  $\gamma$  rays from Na-22 source.

Sample No.	tSIE	$\text{CH}_3\text{NO}_2$ ( $\mu\text{l}$ )	Efficiency
Blank	491.8	0	---
N1	518.5	0	0.9106
N2	507.5	20	0.9105
N3	465.4	50	0.9102
N4	447.6	70	0.9101
N5	424.4	100	0.9099
N6	396.5	150	0.9097
N7	369.2	200	0.9095

## ➤ The influence of quench(tSIE) for Na-22 efficiencies

- ❑ The contribution of  $\gamma$ -rays to overall efficiency was taken into account in efficiency calculation.
- ❑ But the deviation of tSIE due to  $\gamma$ -rays was not considered.

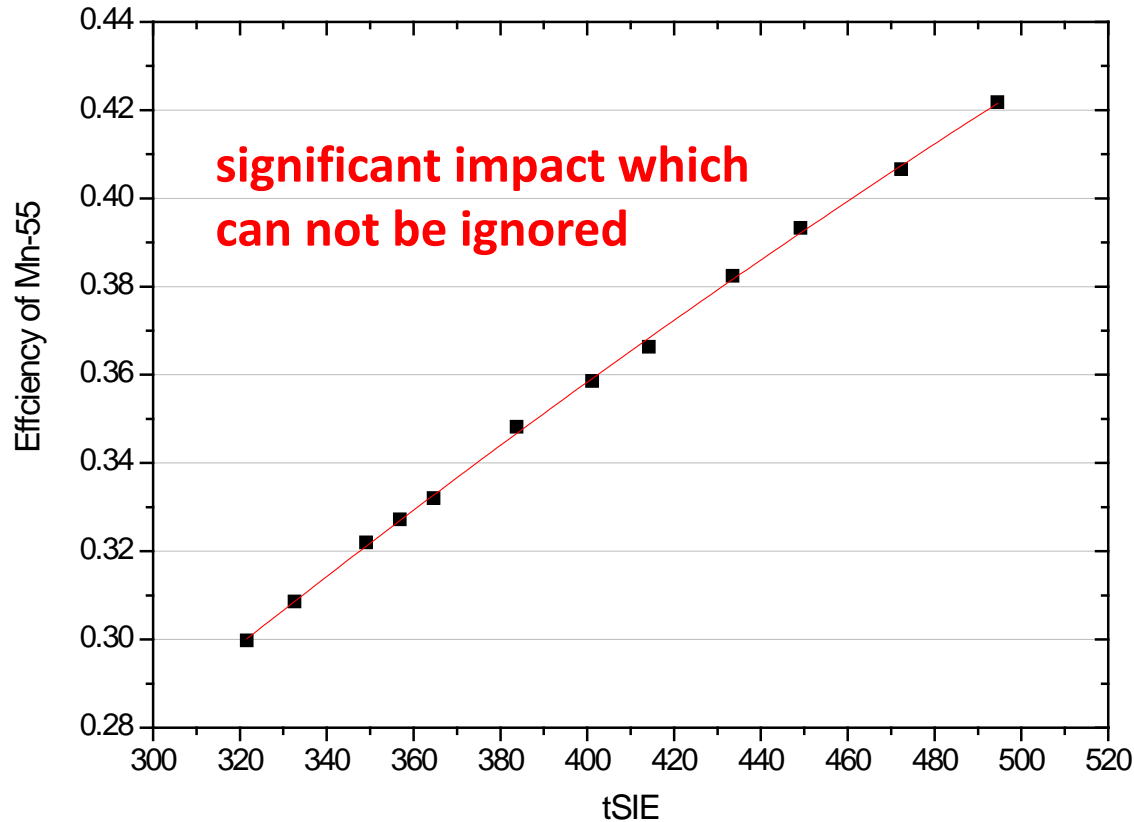
## ➤ The influence of tSIE for Na-22 efficiencies



The tSIE was varied over a range from **321-494**, which corresponds to  $^{22}\text{Na}$  efficiencies from **90.95%** to **91.06%**.

## ➤ The influence of tSIE for EC- $\gamma$ nuclides

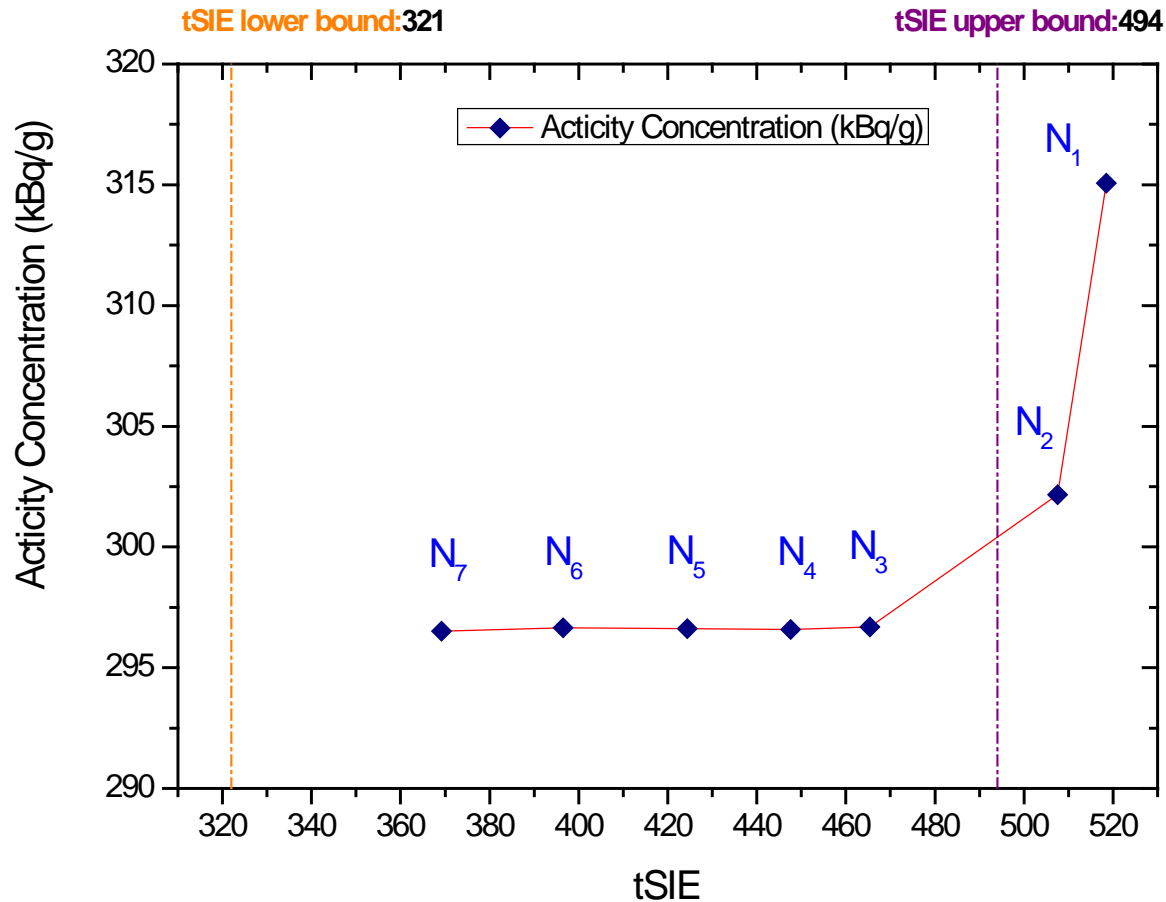
for example: Mn-54



The tSIE was varied over a range from **321-494**, which corresponds to  $^{22}\text{Na}$  efficiencies from **30.0%** to **42.2%**.

## ➤ The problem of tSIE

(2) The tSIE of  $N_1$  and  $N_2$  are over the upper bound of quench correction curve



What can we do?

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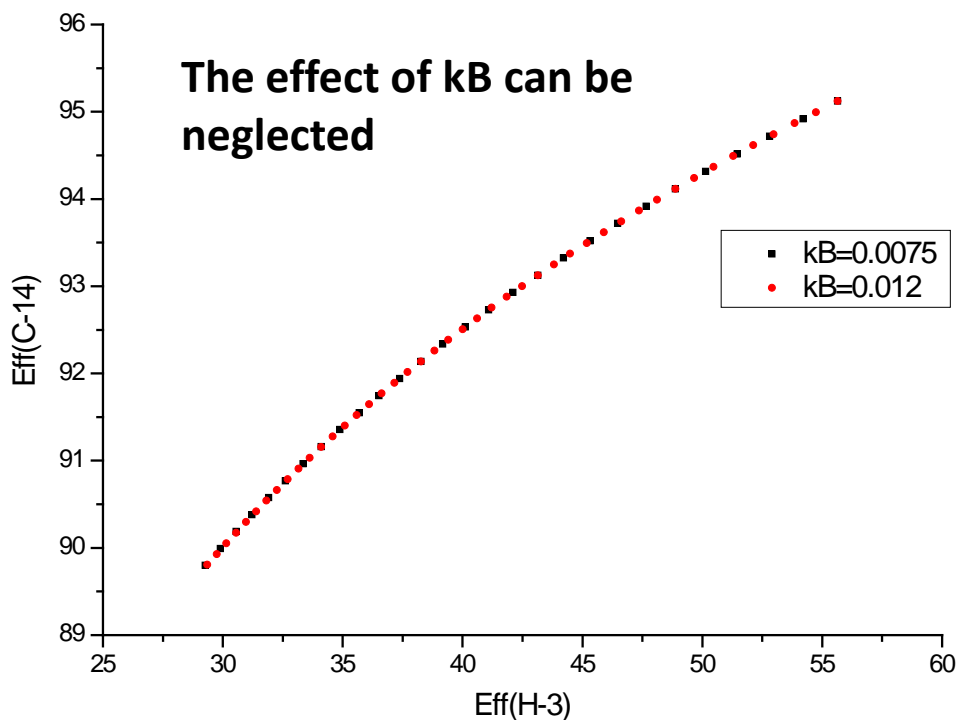
Conclusions

## ➤ The influence of kB values for CIEMAT/NIST method

- ❑ In some of the early CIEMAT/NIST method literatures, different values of Birks' parameter, kB, have been reported by various authors.
- ❑ But it doesn't matter for pure  $\beta$ -emitters, because the efficiencies were calculated twice in the procedure and enabled us to establish a link between the counting efficiencies of the two radionuclides (tracer and nuclide under study).
- ❑ Owing to cross-correlations, relative calculations are more likely to exhibit less uncertainty than absolute estimations due to the cancelation effects.

## ➤ The influence of kB values for CIEMAT/NIST method

- ❑ For CIEMAT/NIST method, although sometimes the kB value used in the calculation may be not the optimal one, but it has little effect on the result for pure  $\beta$ -emitters.

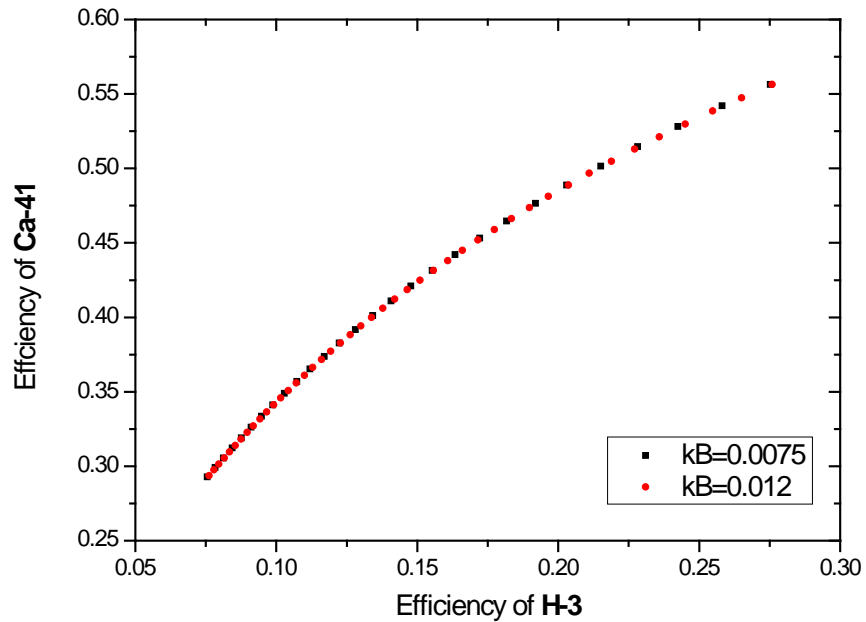




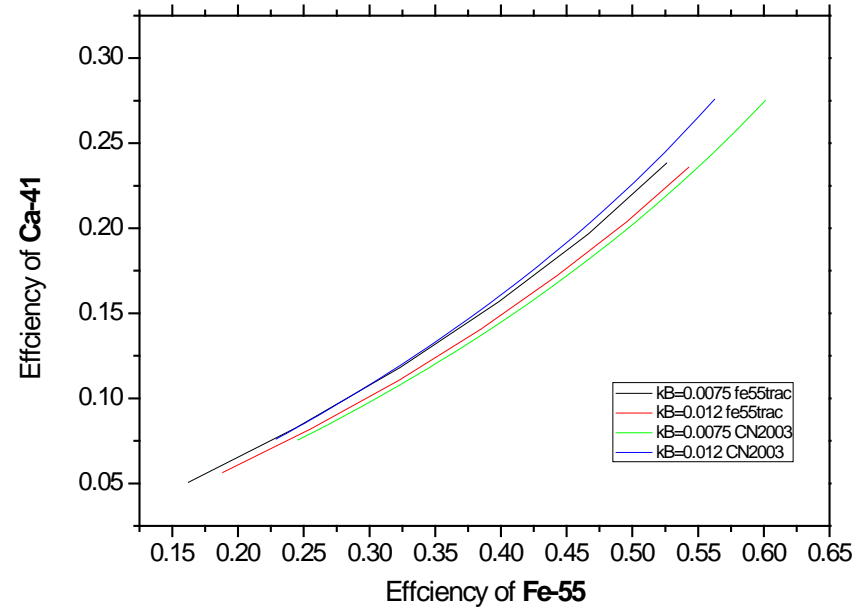
## ➤ The influence of kB values for CIEMAT/NIST method

❑ However, for EC decay nuclides:

for example: Ca-41

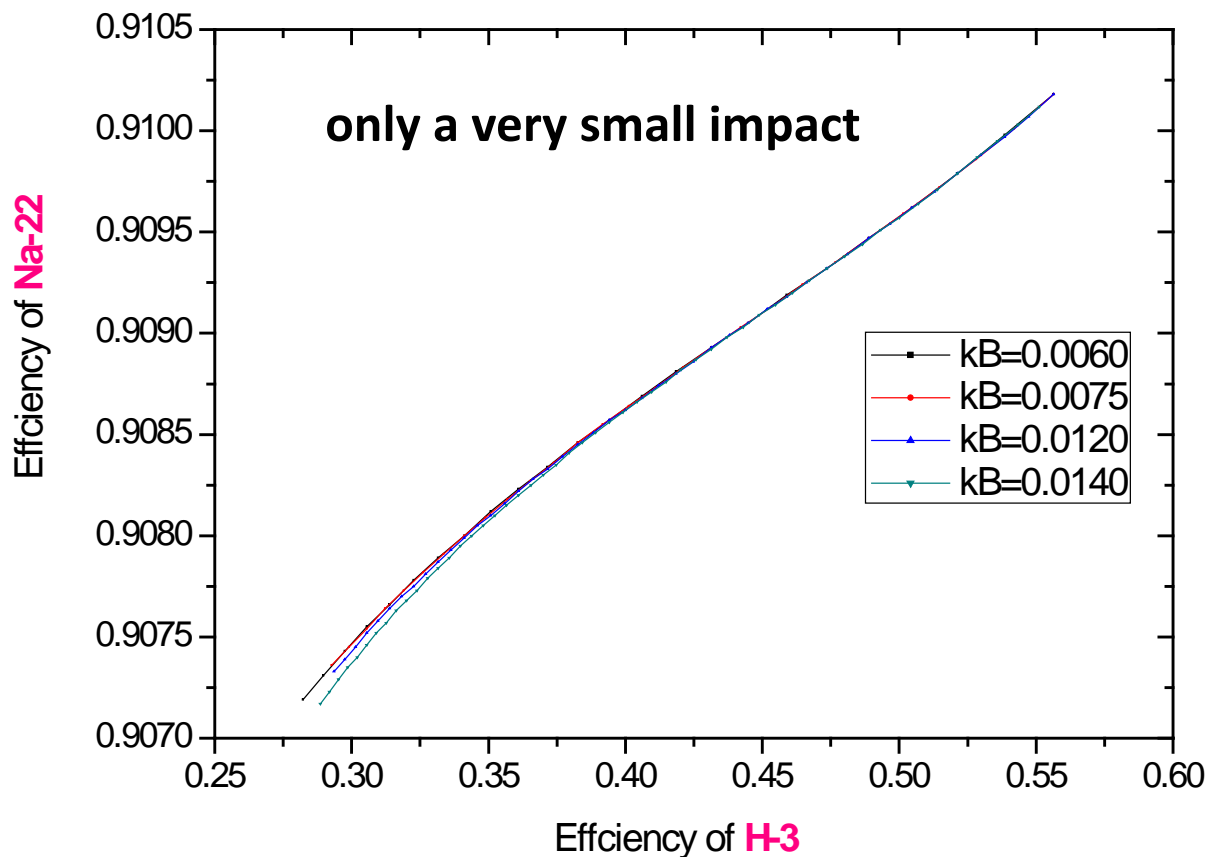


The effect of kB can be neglected



The effect of kB is obvious

## ➤ The influence of kB values for $^{22}\text{Na}$ efficiencies



There is a very small difference of using different kB values, within 0.03% in the whole interval..

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# Conclusions

## □ Influence of tSIE

- (1) If tSIE values are in the interval of quench correction curve, the influence of tSIE to  $^{22}\text{Na}$  can be neglected.
- (2) If tSIE values are out of range of quench correction curve, the data could not be used.

## □ Influence of kB values

- (1) The kB values are of only minor importance for  $^{22}\text{Na}$  efficiency calculation.



Thank you for your attention!