原子核理論研究室

—— ある原子力事故の検証 ——

議論するテーマ

1. **Cs-137 の問題**
2. **原子核分裂と臨界**
3. **ＪＣＯ事故の再検証**

1　Cs-137 の問題

福島の原発事故で沢山の放射性物質が原子炉から放出された
特に Cs-137 はその寿命が約３０年のため人体への影響が問題
ここでは東大教授早野達の最近の論文について簡単に解説する

1.1　福島の原発事故

(1) 巨大地震後の津波による電気系統の崩壊

(2) 使用済み核燃料の温度上昇

(3) 放射線による水分子分解

(4) 水素分子の大量蓄積

(5) 水素分子の引火爆発

(6) 核物質の灰（放射性元素）の大量放出

1.2　早野達の解析

(1) 放射線汚染地区の住民（約３万人）の体内の Cs-137 分析

(2) 最初は多数の住民から有限量の Cs-137 を検出

(3) 検査前に白衣に着替えて測定：Cs-137 が検出されなくなった

(4) ４人の老人については白衣に着替えても Cs-137 が検出された
問診の結果、毎日裏山のキノコを食している
食生活を改善した結果 Cs-137 が検出されなくなった

1.3　問題点

Cs-137 は原子状態としては体内に蓄積される事はない

これが有機 Cs になる事はないのだろうか？

(無機水銀は無害でも有機水銀は極めて有害であった: 水俣病)

2　原子核分裂と臨界

${}^{235}_{92}U$ は熱中性子 (≤ 0.1 eV) による核分裂で
膨大なエネルギーを放出

$$n + {}^{235}_{92}U \rightarrow A_1 + A_2 + 2.5\ n + Q\ (\sim 200)\ \mathrm{MeV}$$

しかし、${}^{238}_{92}U$ は熱中性子を吸収しても核分裂しない

自然界に存在するウランは９９．３％が ${}^{238}_{92}U$

${}^{235}_{92}U$ を全体の (3∼5) ％に濃縮したものが核燃料

2.1　科学的疑問

(1) 何故、${}^{235}_{92}U$ は熱中性子を吸収して核分裂し
${}^{238}_{92}U$ は核分裂しないのか？

(2) ペアリング力の問題である事が解明された
（注：原子核中において、中性子同士はペアを作りたがる）

この理論の解説は３年生には難しすぎるので省略

2.2 臨界

(1) **連鎖反応**：核分裂反応 $n +^{235}_{92} U \rightarrow A_1 + A_2 + 2.5\ n$ で放出された平均 2.5 個の中性子が再び核分裂反応に関与する事

中性子の寿命は15分程：連鎖反応が持続する時 **臨界**という

(2) **問題点**：
核反応で放出される中性子のエネルギーは約 1 MeV
熱中性子による核反応断面積の 100 分の1 以下

2.3 原子炉

原子炉: 核分裂で生成された中性子を水で減速させる

(1) **中性子の減速**：
中性子が水分子中の陽子と衝突すると減速する

(2) **中性子の平均自由行程** ℓ_n：
水中で中性子が陽子と衝突するための平均距離 ($\ell_n \sim 1$ cm)
中性子が核分裂を起こす反応時間 τ： $\tau \leq 10^{-5}$ 秒

3　JCO の事故の再検証

東海村 JCO 臨界事故の概要：

(1) 1999 年 9 月 30 日　東海村の核燃料加工施設
株式会社 JCO が起こした原子力事故（臨界事故)
被曝による死亡者が出た

(2) 至近距離で中性子線を浴びた作業員 3 名中
2 名が死亡、1 名が重症

(3) ウラン化合物の粉末を溶解する工程：
(a) 裏マニュアルではステンレス製バケツを用いた
(b) 手順最終工程である製品の均質化作業で
背丈が低く内径の広い容器（沈殿槽）使用
これは冷却水のジャケット（2 cm 幅）に包まれている

(4) 濃縮度 (18.8 %の $^{235}_{92}U$) の硝酸ウラニル水溶液：
貯蔵した容器の周りの 2 cm 幅の冷却水が中性子反射材となり
溶液が臨界状態となって中性子線が大量に放射された

(5) 「約 16kg のウラン溶液を溶解槽に移している時に青い光が出た」

3.1　科学的疑問

(1) 最初の中性子はどこから来たのか？

(a) 最初の中性子源：

$^{238}_{92}U$ は自然崩壊して中性子を放出

1g の $^{238}_{92}U$ 毎秒約 0.01 個の中性子を放出

(b) 2.4 kg のウラン粉末： 毎秒 約２０個の中性子放出

(2) 即発中性子が核分裂を起こす反応時間 τ： $\tau \leq 10^{-5}$ 秒

(3) この臨界事故での総核分裂数は約 2.5×10^{18} 個程度と予測

(4) ウラン粉末を溶解する工程で水をどれだけ入れたのか？

(a) 2.4 kg のウラン粉末に硝酸を 1.7 ℓ, 純水を 1 ℓ 入れる
さらに水を追加して溶液全体を 6.5 ℓ とした

(b) この溶液を沈殿槽に移す工程を１バッチとする

(c) 7バッチ目の段階で（沈殿槽から？）青い光が見えた

(d) 青い光はチェレンコフ光：
高エネルギー電子が「物質中での光速」を超えた時
（例：水中 0.75 c 以上，電子のエネルギー 0.27 MeV）

3.2 臨界はどうして起こったのか？

１バッチの総量： 溶液全体: 6.5 ℓ
$^{235}_{92}U$ の総量: 378 g
水: 4.3 ℓ

(1) 核分裂直後の中性子のエネルギー $\boxed{E_n \sim 1 \text{ MeV}}$

(2) 中性子が次の核分裂を起こす平均自由行程 (m.f.p.)

$$\boxed{\ell_f \sim 67 \text{ m}}$$

従ってこの中性子による連鎖反応はここでは起こらない

(3) この溶液中で中性子が水分子の陽子と衝突する
平均自由行程 (m.f.p.)： $\boxed{\ell_n \sim 1 \text{ cm}}$
１回の衝突で半分のエネルギーを失う
25 cm 走ると熱中性子エネルギーになる

(4) 熱中性子が次の核分裂を起こす平均自由行程 (m.f.p.)

$$\boxed{\ell_f \sim 12 \text{ cm}}$$

直径 45 cm の沈殿槽に 45.5 ℓ のウラニル溶液
沈殿槽における全ウラニル溶液の高さ $h \simeq 28$ cm

連鎖反応が起こり臨界になる可能性はかなり高い！

Journal of Modern Physics, 2016, 7, 1500-1505
Published Online August 2016 in SciRes. http://www.scirp.org/journal/jmp
http://dx.doi.org/10.4236/jmp.2016.712136

Reexamination of Criticality Accident in JCO

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Received 22 June 2016; accepted 22 August 2016; published 25 August 2016

Abstract

Nuclear chain reactions are, by now, commonly used in the nuclear reactors, and thus it seems that there is no basic problem in fission processes from the scientific point of view. However, the criticality accident that occurred in JCO in 1999 suggests that one should carefully examine this accident from the nuclear physics point of view. Indeed the chain nuclear reactions should have taken place in the small area of space with 45 cm diameter disk times 30 cm height tank. In fact, when people carry the uranium nitrate solution into sedimentation tank, then this solution with uranium should get into the critical state at the 45ℓ of uranium nitrate solution. The root cause of the accident should not be very simple from the nuclear physics point, and it should be quite important to examine why the uranium nitrate solution with 45ℓ could have become critical.

Keywords

Nuclear Fission, Criticality, Mean Free Path

1. Introduction

The criticality accident that occurred in JCO in 1999 must be most serious, and it should not be very easy to understand why the nuclear chain reactions could proceed in a small area of space for a finite period of time. In this sense, it should be quite important to carry out the careful examination of criticality accidents from the nuclear physics point of view. It should be, of course, difficult to claim that the JCO accident can be a target of the scientific study since one cannot make the experimental study of the JCO type accidents. However, we believe that the basic mechanism of the criticality accident should be clarified why it could naturally occur in the small area of space.

This criticality accident occurred when workers in JCO company were carrying the uranium nitrate solution (18.8% enriched uranium) into sedimentation tank [1]-[3]. Here, we should explain the working procedure

How to cite this paper: Oshima, S., Hisada, M., Saito, Y. and Fujita, T. (2016) Reexamination of Criticality Accident in JCO. *Journal of Modern Physics*, **7**, 1500-1505. http://dx.doi.org/10.4236/jmp.2016.712136

which is taken by the workers in JCO. First, they make the uranium nitrate solution which is composed of 2.4 kg U_3O_8 with the nitric acid of 1.7ℓ in the stainless vessel. In addition, they add water to the uranium nitrate solution until the total volume becomes 6.5ℓ. Then, they carry the 6.5ℓ solution into the sedimentation tank, and this working procedure is called one batch.

The criticality accident should have occurred in the middle of the seventh batch since the workers noticed blue lights that should be due to the Cherenkov radiation. In fact, two of the workers suffered from the neutron radiation.

A question should arise as to how the nuclear chain reactions could proceed within the small sedimentation tank (45 cm diameter, 60 cm high). There are, of course, some analysises of this criticality accident [4] [5]. However, these studies are mainly carried out for the computer simulation such that the total energy emitted via radiations can be reproduced in some way or the other. These investigations are, of course, very important in order to understand the accident cause. However, it is also important to carry out the study of the criticality accident from the nuclear physics point of view.

In this paper, we carry out careful calculations of the criticality accident in terms of the multiple scattering theory. Here, we want to understand why the nuclear chain reactions can proceed in the small area of space. In particular, we trace the nuclear fission reactions (nucleon-nucleon collision together with nuclear fission) each by each, and we clarify the microscopic processes why and how the criticality accident occurred. As a result, we should understand some specific reasons why the chain reactions can proceed, and this can be done by making use of the mean free path which is the result of the nuclear multiple scattering theory.

However, when we clarify how the criticality accident occurs, we face to the most difficult question as to why the criticality could stop. In this study, we find an answer for this question, though not necessarily sufficient. This mechanism of stopping criticality may be related to the quick settle of the uranium compound.

As a result of our calculation, we find a possible dangerous situation which was thought to be due to the 8th batch, if it were carried into the sedimentation tank. We see that the estimated energy release after the virtual 8th batch should become the same order of magnitude as the Chernobyl nuclear accident.

2. Nuclear Chain Reactions

Nuclear fission reaction by incident neutrons can be written as [6]

$$n + {}^{235}\text{U} \to A_1 + A_2 + (2 \sim 3)n \tag{1}$$

where A_1, A_2 are new nuclei which are produced in the reactions. In this reaction, there are two important points. The first one is concerned with two or three neutrons which are produced in the reactions. The second point is that the probability of this nuclear reactions is strongly based on the incident neutron energy, and the biggest cross section is for the incident neutron with almost zero energy (thermal energy).

The chain reactions indicate that the produced neutrons should be absorbed by another ^{235}U such that the nuclear fission can proceed further on. In addition, if the chain reactions continue to proceed without the aid of other external neutron sources, then this situation is called a criticality stage. In reactors, this criticality must be kept by controlling the number of neutrons involved in the chain reactions.

In normal reactors, a few % enriched uranium should be commonly used, but in this JCO accident, 18.8% enriched uranium were used, and this high enrichment should be one of the strong reasons why the nuclear reactions run wild.

3. Why Criticality?

Now, a question is as to why the criticality is realized in the small area of the sedimentation tank with 50ℓ of the uranium nitrate solution. That is, why nuclear chain reactions continue to occur in this small area. Here we clarify the basic mechanism of the criticality accident.

3.1. Neutron Source

The nuclear chain reactions should require thermal neutrons to start for the initial fission reactions. Since neutrons should decay within 15 minutes, they do not exist as a natural source. Neutrons should be produced in some way or the other. Here in this accident, the neutron source should be the decay of ^{238}U spontaneous fissions. The life time of ^{238}U is about 4.5 billion years and, in addition, the rate of the spontaneous fission to the total width is around 5.45×10^{-7}. Therefore, 1 g of ^{238}U make the spontaneous fission of 0.01 times per second. Since one batch contains 1.6 kg of ^{238}U, we should find about 20 neutrons per second in the one batch solution.

3.2. Mean Free Path of *n*-^{235}U Fission (Fast Neutrons)

The probability of nuclear fission of ^{235}U induced by neutrons should be evaluated in terms of mean free path of λ inside the uranium nitrate solution. This mean free path of nuclear reactions can be obtained from the multiple scattering theory as

$$\lambda = \frac{1}{\rho\sigma_f} \tag{2}$$

This derivation of the mean free path (2) is based on the Glauber theory [7], and this theoretical frame work is well examined in atomic and nuclear reactions [8] [9]. Here, ρ denotes the number density of ^{235}U in solution and σ_f corresponds to the nuclear fission cross section of ^{235}U induced by neutrons. In fact, the number density of ^{235}U in one batch solution is $\rho \simeq 1.5\times10^{20}\ \text{cm}^{-3}$ which is a constant. On the other hand, the nuclear fission cross section σ_f of ^{235}U induced by neutrons crucially depends on the incident energy of neutrons. The incident energy dependence of the observed cross sections σ_f can be written as [10]

$$\sigma_f \simeq \begin{cases} 585\ \text{b} : E_n \simeq 0.025\ \text{eV} \\ 1\ \text{b} : E_n \simeq 1\ \text{MeV} \end{cases} \tag{3}$$

where $1\ \text{b} = 10^{-24}\ \text{cm}^2$.

Mean Free Path of Prompt neutrons in Nuclear Fission

In fission process, the average energy of prompt neutrons is around 1 MeV, and therefore the average mean free path of the prompt neutrons after fissions becomes

$$\lambda_f = \frac{1}{\rho\sigma_f} \simeq 67\ \text{m}. \tag{4}$$

This is quite long in comparison with the scale of the tank, and therefore this prompt neutrons by themselves cannot induce subsequent fissions in corresponding solution in the tank. In this respect, we ask a question as to why the criticality should take place within the small sedimentation tank.

4. Collision between Neutrons and Water Molecule

In reality, the prompt neutrons may collide with protons in water molecule, and they should lose their energy by nucleon-nucleon collisions. Since the nuclear fission cross sections become largest for the thermal neutrons, the fission processes should start in case the prompt neutrons lose most of their energy inside the uranium nitrate solution.

4.1. Energy Loss after the Collision of Prompt Neutrons with Protons in Water

When the prompt neutron scatters with protons in water, this neutron should lose a half of its energy. This can be easily understood in the following way. First, we denote the incident momentum and energy of the neutron by $\boldsymbol{p}, E_n$ with $E_n = \frac{\boldsymbol{p}^2}{2M}$, and the final momentum and energy by $\boldsymbol{k}, E_n'$ with $E_n' = \frac{\boldsymbol{k}^2}{2M}$. In this case, we find an equation from the conservation law of momentum and energy as

$$\frac{\boldsymbol{p}^2}{2M} = \frac{\boldsymbol{k}^2}{2M} + \frac{(\boldsymbol{p}-\boldsymbol{k})^2}{2M} \tag{5}$$

which can be solved and its solution becomes

$$k = p\cos\theta. \tag{6}$$

Since the observed scattering cross section does not depend on the scattering angles, we can make an average over the angles, and we obtain the average energy after the scattering

$$E_n' = \frac{1}{\pi}\int_0^{\pi} \frac{k^2}{2M}\mathrm{d}\theta = \frac{1}{\pi}\int_0^{\pi} \frac{p^2}{2M}\cos^2\theta \mathrm{d}\theta = \frac{1}{2}E_n. \tag{7}$$

This means that a neutron should lose a half of its energy in each scattering process.

4.2. The Mean Free Path of Neutrons Inside Water

Now we calculate the mean free path of neutrons after the scattering with protons in one batch solution. The number density of protons in one batch solution is $\rho_p \simeq 4.9\times10^{22}\ \mathrm{cm}^{-3}$. The neutron-proton cross section at low energy is observed as $\sigma_{np} \simeq 20\ \mathrm{b}$ [11], and thus the mean free path of neutron in one batch solution becomes

$$\lambda_p = \frac{1}{\rho_p \sigma_{np}} \simeq 1\ \mathrm{cm}. \tag{8}$$

Therefore, a prompt neutron with 1 MeV energy should have its energy after it travels around 25 cm,

$$E_n' = 1\ \mathrm{MeV}\times\left(\frac{1}{2}\right)^{25} \simeq 0.03\ \mathrm{eV}. \tag{9}$$

This neutron does not have to travel linearly, but in any case, it should become a thermal neutron.

4.3. Mean Free Path of Thermal Neutron in the *n*-^{235}U Fission Process

We can easily calculate the mean free path of the thermal neutron before the nuclear fission in one batch solution. Since $\sigma_f = 585\ \mathrm{b}$, we find

$$\lambda_f = \frac{1}{\rho\sigma_f} \simeq 11\ \mathrm{cm} \tag{10}$$

From these considerations, we see that prompt neutrons with 1 MeV should travel around 25 cm, and then they become thermal neutrons. Further, after they travel 11 cm, they can induce nuclear fissions. Thus, if one carries 50ℓ of the uranium nitrate solution into the sedimentation tank with 45 cm diameter and 25 cm height, then nuclear chain reactions may well start quickly and proceed further on.

4.4. Reaction Time of Neutrons

Now we see that when prompt neutrons travel 36 cm, then they can induce nuclear fissions. Therefore, we should estimate the duration time that is necessary to travel this 36 cm. Since the nuclear reaction time must be smaller than 10^{-15} second, we can ignore this time duration. Since the prompt neutron with 1 MeV should spend $\tau_0 \simeq 7.6\times10^{-10}$ second to proceed 1 cm, its energy becomes a half of the previous energy after 1 cm walk. Therefore, the time to proceed the next 1 cm becomes larger by a factor of $\sqrt{2}$. In this way, if the prompt neutron proceed 25 cm, then the total time to spend must be

$$T_0 = \left(1+\sqrt{2}+\cdots+2^{25/2}\right)\tau_0 \simeq 15\ \mu\mathrm{s}. \tag{11}$$

After that, this neutron becomes thermal, and it should proceed 11 cm before the nuclear fission. Since the thermal neutron may have the energy of 0.03 MeV, it should take $\tau_{th} \simeq 46\ \mu\mathrm{s}$. Thus, the total time that is necessary for the prompt neutron to induce a fission reaction should be $T_{tot} \simeq 61\ \mu\mathrm{s}$.

5. Total Energy of Fission with Criticality

Here, we should estimate the total amount of energy which is released from this accident. This evaluation must be very difficult, but we want to calculate it in an approximate way and obtain an order of magnitude of the total energy.

First, the number of neutrons which is required for the criticality reactions should be taken as $n_r = 1.001$, which is assumed to be consistent with the total energy released as calculated from the computer simulation. In nuclear reactors, one should make use of all the possible techniques to keep the number as $n_r = 1$.

In addition, we assume that the number of nuclear fissions should be $N = 40000$. This number is chosen so that the total nuclear energy release should be consistent with the computer simulation which can reproduce all the observed radiation energies. In this case, the total reaction time of fission becomes $T_f \simeq 2.4$ s, and the total number of fissions becomes

$$N_{tot} = 1.001^{40000} \simeq 2.3 \times 10^{17}. \quad (12)$$

Further, we evaluate the neutron number at the beginning, and this neutron should come from the spontaneous fission of ^{238}U. The number of neutrons in one batch solution must be around 20, and we take a half of this number. The energy release from the nuclear fission must be around 200 MeV in each reaction, and therefore the total energy becomes

$$E_{tot} \simeq 4.6 \times 10^{26} \text{ eV} \quad (13)$$

which is just similar to the result of the computer simulation.

6. Why Does the Criticality Stop?

It is true that the criticality accident produced a huge amount of energy by the nuclear chain reactions, and the accident is indeed quite serious. In this sense, we here clarify as to how the chain reactions started and continued by reaching the critical stage. However, we face to the more serious problem at this point. That is, *why the criticality accident could stop*? We should understand any reason why the criticality could stop, namely there were only one burst and not any more burst, but why?

6.1. Nuclear Fission in the Seventh Batch

Here, we try to answer for this question, though it should be extremely difficult. In order to find a possible mechanism for the stopping of the criticality, we assume that the uranium compound should settle faster than any other compounds in the solution. Further, we assume that uranium should be settled within 20% height from the bottom of the sedimentation tank.

In this case, after the sixth batch, the uranium should be settled up to the 4.9 cm from the bottom. Thus, water should be found for 19.7 cm long in the sedimentation tank. By taking into account this fact, we can calculate the total energy release by nuclear fission as

$$E_{tot} \simeq 4.6 \times 10^{26} \text{ eV} \simeq 7.4 \times 10^{7} \text{ J}. \quad (14)$$

The duration time of this nuclear reactions can be estimated and should be around $T_f \simeq 2.4$ s, which should correspond to the time that the uranium compound is coming down to the bottom.

6.2. Nuclear Fission in the Sixth Batch

The same calculation can be carried out for the sixth batch case. In this case, we see that the total energy must be 1000 times smaller than that of the seventh batch case. This is not very large, but at the sixth batch, the nuclear chain reactions already started, and indeed there were a small burst.

From this calculation, we now understand the reason why the criticality stopped. In case the uranium were settled at the bottom of the tank, then the nuclear chain reaction cannot proceed further since the prompt neutrons cannot lose their energy because of the lack of water.

6.3. Nuclear Fission in the Eighth Batch

From now on, we only present a possible scenario of nuclear accident, if the 8th batch were carried into the tank. In this case, the number of uranium involved in the nuclear fission must be proportional to the height of water, and thus it should be $\frac{22.9}{19.7}$ more than the seventh batch. Thus, the number becomes

$$N = 40000 \times \frac{22.9}{19.7} \simeq 46500. \quad (15)$$

This means that the number of nuclear fissions should be also increased and the total number becomes

$$N_{tot} = 1.001^{46500} \simeq 1.5 \times 10^{20}. \quad (16)$$

Therefore, the total energy becomes

$$E_{tot} \simeq 3 \times 10^{29} \text{ eV} \simeq 4.8 \times 10^{10} \text{ J}. \quad (17)$$

This energy 4.8×10^{10} J corresponds to 11 ton of TNT powder which is quite a serious explosion. The accident of Chernobyl nuclear power plant is believed to correspond to around 100 ton of TNT powder, and therefore, if the 8th batch were thrown away, then the accident would have been more than serious.

7. Summary

We have discussed the basic mechanism of the JCO accident in terms of the nuclear multiple scattering theory. In this paper, we have clarified how the nuclear chain reactions could proceed in the small area of the sedimentation tank. The JCO accident should be studied from the point of view science, even though there must be no serious technical problems in nuclear reactors. In this respect, one may say that the JCO accident is rather similar to scientific phenomena, and it is essentially different from problems found in the nuclear power plants.

Acknowledgements

We are grateful to N. Yoshinaga for interesting discussions and useful comments.

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