≪Original > Studies on the Preparation of Organic Compounds Labelled by ³⁸Cl. (I)

Inorganic Yields of ³⁸Cl in Szilard Chalmer Reactions of Aromatic Chloro Derivatives

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Abstract

In order to clarify an effective procedure of labelling organic chloro compounds by 38Cl, phenyl chloro derivatives (7 kinds), chloro nitrobenzenes (6 kinds), chloro anisoles(2 kinds), chloro anilines(3 kinds), chloro toluenes(3 kinds), benzyl cholorides(4 kinds), and other comparing samples(3 kinds) were irradiated in the TRIGA Mark-II research reactor and the inorganic 38Cl yields were compared with the irradiation times after extracting the inorganic portion with an aqueous solution of alkali. It was found that the relative change between the inorganic 38Cl yield and the irradiadiation time depends a great deal on the state of the sample, and a solid sample gave a lower and steady inorganic yield. The inorganic ³⁸Cl yield was decreased in the order of phenyl chloro derivatives <chloro toluene
benzyl chloride<cyclohexyl chloride<chloro nitrobenzene<chloro aniline. Among compounds of a simlar chemical structure, ortho and para isomers were shown to give a relatively small diffrence in their inorganic yields. A linear relationship was observed between the inorganic 38Cl yield of homo functional compounds and the number of chlorine atoms on the benzene ring. Generally, poly chloro substituted derivatives could give a higher yield than those of less chloro substituted. The results were discussed and the feasibility of these results for labelling purpose was criticized.

요 약

방사성 ³⁸Cl로 표지된 유기 하로겐 화합물의 제조방법을 가려내기 위하여서 Phenyl halides(7종), Chloro nitrobenzene(6종), Chloro nitroanisole(2종), Chloro aniline (3종), Chloro anisidine(3종), Chloro toluene(3종), Benzyl chloride 유도체(4종) 및 기타 대조 화합물 3종 등을 원자로 내에서 조사하고 생성물을 알카리 수용액으로 주출하고 각각의 무기 ³⁸Cl 수율을 조사시간 별로 검토 하였다. 그 결과 화합물의 구조 보다도 표적물의 상태에 따라서 무기물 수율에 변화가 있고 고체상의 표적물의 경우에는 수율이 일반적으로 얕고 변화가 고르다는 것을 확인 하였다. 화학구조에 따르

는 무기 **Cl의 생성수율의 감소는 현재까지의 결과로 보아서 Phenyl chlorides
Chloro toluenes
Benzyl chlorides
Cyclohexyl Chlorides
Chloro nitro benzene
Chloro aniline의 순서이였다. 같은 확학구조의 화합물에 있어서 o-, m-, 그리고
p-, isomer 사이에는 수율변화가 크지 않았다. 같은 관능기를 갖인 화합물에 있어서
는 benzene 고리의 염소원자수와 무기 **Cl 수율간에 Linear relationship 가 있었으며
염소원자수가 많을수록 그 수율이 얕었다. 실험 결과를 논의 하였으며 표지 반응에
대한 본 실험 결과의 응용성가능 여부를 고찰 하였다.

1. Introduction

During the course of a research on the preparative work of organic chloro aromatics in this laboratory, various compounds labelled by ³⁸Cl were required for tracer works. It was, therefore, attempted to conduct a study on the recoil labelling of aromatic chloro compounds on basis of literatures reported.

According to the literatures on the labelling of organic compounds by a recoil reaction of ³⁸Cl, aliphatic chlorine compounds were irradiated in the reactor to give less than 30% yield of the original compound labelled after being separated by a complicate procedure of a gas chromatography1). In case of aromatic compounds there appeared several papers on the radio-chlorination of various mono substituted benzens²⁾ by a recoiled ³⁸Cl, a study on the reaction of a recoiled 38Cl of a mixture of monochloro benzene and various alcohols3), and the reaction of a recoiled 38Cl with BHC derivatives4). Generally, the mechanism of the recolied reaction appeared to be a very complicate mixture of an exchange reaction between Cl and 38Cl2, 3), ionic chlorination or radical chlorination reaction2), and collision between the atoms and molecules at the near surface of the solvent cage2, 3), but no detailed feature of the reaction could be given except some possibilities of the formation of a π complex intermediate between the recolied chlorine atom and the benzene ring of the compound³⁾. It was, however, stated that the radiation stability of a benzene ring towards a

neutron beam may be able to cut down the number of the components of the irradiated target, and consequently the labelling yield of the original sample may relatively be higher than those of other type of compounds²⁾. The influence of the steric structure of a solid state of a sample on the inorganic yield of an irradiated target could be of significant in case of BHC isomers⁴⁾.

Differences in the phase of the target materials also cause different reactions of the recoiled species, and hence different yields of the recoil product may be obtained with solid, liquid, and frozen target. Inorganic target materials such as ployphosphates⁵⁾ and others, when the crystalline form was used as target materials, could result a number of recoiled reactions in a solid matrix, and the differences in vield due te the chemical structure could disappear when the materials were irradiated in aqueous solutions6). Numbers of papers have been published along these lines. Organic target materials such as benzene derivative would, on the contrary, be expected to result less complicate recoil products, when they are irradiated in a soild state or frozen state, since the reactive proximity of liquid cages, the molecular movement, and fragment of molecules caused by an irradiation would remarkably be inactivated towards a recoiled atom in these phases. Researches along these lines have been reported in many cases of organic radiation reactions⁶⁾, and a hot atom chemistry, even though the detailed features of the reaction mechanism have not yet been

defined.

The attempted study was, therefore, concentrated to investigate the mode of the variation of the inorganic 38Cl yield of an irradiated target v.s. the irradiation time adopting as many numbers of aromatic chloro derivatives of solid state as possible. By checking the inorganic yield, the labelling condition of the highest yield may indirectly be estimated, since the yield of extractable chlorine in aqueous solution is more specific for a target compound than the radio chemical yield of the activated organic phase, in which many 38Cllabelled organic species may be produced. Yields dependence on the irradiation time, chemical structure, and other essential parameters may also be explored to clarify the general mode of the recoiled 38Cl atom in the target material, though the primary intersts are concentrated to check the feasibility of labelling the desired compound by the recoil process of a 38Cl atom. In the present paper the general features of preliminary aspects of the study were described and potent conditions of labelling aromatic chlorine compounds were considered. On basis of the present results a detailed study on the separation of the organic phase to isolate the labelled compounds desired will further be attempted and be published in the subsequent papers.

2. Experimental

(1) Materials

The aromatic chloro derivaties of reagent grade used in this experimental were obtained from a commercial source(Tokyo Kasei) and other derivatives were prepared in this laboratory by the author's procedure⁷. All samples were checked their melting points, G.C. data, and spectrophotometric identifications.

(2) Neutron Irradiation

The sample which amount around 0.2-0.4g was sealed in a polyethylene tube(I.D. 1cm,

height 2cm) and the tube was irradiated at the pneumatic tube facilities of the TRIGA MARK-II research reactor(250K. W., neutron flux: $3.0\times10^{12} n/cm^2/sec.$, Cd ratio 2.0(Au)). The irradiated tube was annealed at the room temperature for 30 minutes in a fume hood and the content of the tube was processed as follows.

(3) Extraction of the Inorganic Portion and the Derermination of Inorganic Yield

The irradiated tube was opened in a fume hood and the content of the tube was poured in a beaker which contains about 20ml of benzene. The benzene solution was shaked with a equal volume of 2N-KOH solution in a separate funnel and the organic layer was separated. The process of shaking was repeated using another portion of 2N-KOH solution in an another separatory funnel and the organic portion was again separated. The shaking was thus repeated four times. In each shaking process the inorganic portion was kept in a mess cylinder and 1 ml of the portion was taken in a screw capped test tube. The last portion of the organic layer separated from the shaking process was washed once with an equal volume of distilled water and the organic layer was separated. The inorganic layer was kept in a mess cylinder and 1 ml of the layer was taken in a screw capped test tube. The organic layer was measured in a mess cylinder and 1 ml. of the layer was taken in a test tube. Total five tubes of the inorganic portion and one tube of the organic portion were thus obtained and they are counted in a r-well type scintillation counter(Fujitsu. Model TM-II) at an equal geometric position*. The determination of the inorganic 38Cl yield was conducted on basis of

^{*} The standard deviation of the background of the counter was calculated. The amount of the background was 510 counts/min. and the standard deviation was 22.6 counts/min.

the following calculation.

Total count of the inorganic portions × 100

Inorganic Yield(%)= Total count of the inorganic portions+Total count of organic portion.

Example:

Sample: 1, 2, 4, -trichloro benzene

Irradiation Time: 1 minute. (30 minutes annea-

ling) Counting Time: 30 Seconds

Fractions	Volume ml.	Ratio activty Per ml.	Total Activity (Calc.)
Organic	24.5	58, 324	1, 428, 938
Inorganic 1	18.5	13, 769	254, 727
Inorganic 2	19. 5	959	18, 701
Inorganic 3	20.0	192	3, 840
Inorganic 4	20.0	152	3, 020
Inorganic 5	19. 5	115	2, 243
Total			1, 711, 469

Inorganic Yield(%) =
$$\frac{282,531}{1,711,469}$$

$$\times 100 = 16.5\%$$

The calculated inorganic yields of samples were plotted against the irradiation time as shown in Fig. 1-5 and summarized in Table 1-4.

Table 1. Inorganic 38Cl yield of neutron irradiated chloro toluenes

Type of compounds	State of sample	Irradia- tion time min.	Inorg. yield %	Remarks
Cl CH ₃	Liquid	10	24	
CI "	//	1	49	
CH ₃	"	10	33	}
, "	"	1	52	
CI—CH ₃	"	15	30	
<i>"</i>	"	10	29	
<i>"</i>	"	5	32	
//	"	2	28	
<i>"</i>	"	1	49	
,	"	1/2	49	

Table 2. Inorganic 38Cl yield of neutron irradiated halogen containning aniline derivaties

Type of compounds	State of sample	Irradia- tion time min.	Inorg. yield %	Remarks
CI NH ₂	Liquid	10	87	
"		1 .	87	
$C1 - NH_2$	Solid	10	78	
" .cı		1	75	
NH ₂	"	10	64	
Cl // OCH3		1	62	
NH_2	"	10	69	
CI //		1	73	
Cl OCH3	"	5	76	
W NH ₂		1	61	
CI CI CI OCH3	"	10	45	}
Cl NH ₂		1	58	

Table 3. Inorganic 38Cl yield o fneutron irradiated chlorine containning some derivatives

Type of compounds	State of sample	Irradia- tion time min.	Inorg. yield %	Remarks
(C) c-c1	Solid	10	16	
₩ OH	"	1	12	
$(CCI_3 - C - CH_3)$	//	1	65	
" CH3	"	10	54	

(4) Identification of 38Cl- in the Extracted Aqueous Phase

The aqueous phase was examined by means of a radio thinlayer chromatography(Plate: Silica gel coated; Merck F254 D, Developing Solvent⁸⁾: MeOH: n-propanol:H₂O: Ammonia: 10% sol. of CCl₃COOH (50:30:15:8:1.5 v/v)) The R_f value of the developed spot on the plate was compared with that of non radio active chloride. (Rf value was 0.53-57 in the both cases)

for 10 n	ninutes in	the resea	rch reactor				
Type of compound	State of sample	Inorg. yield %	Remarks	Type of compound	State of sample	Inorg. yield %	Remarks
<u>(S)</u> -c1	Liquid	41		O_2N $C1$	Liquid	65	
CI CI	"	11		Cl-NO ₂	Solid	48	
CI—CI	Solid	11		NO ₂	"	53	}
CI	Liquid	14			"	35	
CI CI	"	9		O ₂ N CI	//	60	
CI	Solid	7		CI-NO ₂	//	18	15 minutes irradiated
(_') -cı	"	10		CI-OCH ₃	//	58	
ci -()- cı	"	7		CI-CI-OCH3	"	56	
ClCH ₂ Cl	Liq	36		C1 COOH	Solid	64	
H ₃ C-CH ₂ C1	"	46		CI CI OH	"	44	
H ₃ C CH ₂ C1	"	40		CI CI Tetra chloro-			30 minutes
O ₂ N-(\)-CH ₂ Cl	Solid	5.4		fluorescein	"	72	irradiated.

Table 4. The comparison of the inorganic 38Cl yields of aromatic chloro derivatives* irradiated for 10 minutes in the research reactor

3. Results and Discussion

The inorganic ³⁸Cl yield obtained for each sample was plotted against the irradiation time in Fig. 1-5 and was partly listed in the Table 1-3 to check the general tendency of the variation of the yield. The variation of the inorganic yield v.s. the number of chlorine atoms on the benzene ring of the compound was shown in the Fig. 6-8 and the yield dependance on the positional isomers of a phenyl derivative was also shown in the Fig. 9-10.

Solid

Quantitative explanations on these observations will, of course, require an extensive study on the components of the organic phase of each sample irradiated and a complete set of compounds of required structures to elucidate them quantitatively. The author had, however, felt that the following discussions and evaluations on the results will be essential to develop an efficient way of labelling aromatic compounds by ³⁸Cl as attempted,

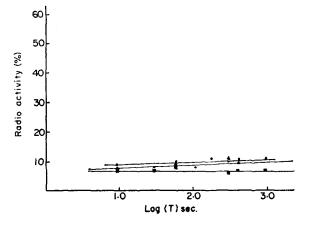


Fig. 1. Inorganic ³⁸Cl yields v.s. irradiation time of the neutron irradiated phenyl halides(solid).

Remarks: Points on the curres denote the followings:

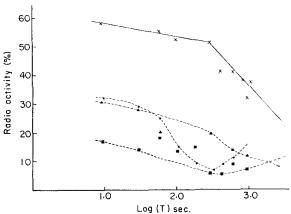
- · P-dichloro benzene,
- ▲ 1, 3, 5, -trichloro benzene, and
- 1,2,4,5,-tetrachloro benzene, respectively.

though they are of a qualitative approach.

(1) The State of a Sample

According to the tables and figures, liquid

^{*} The data on chloro anilines, chloro anisoles, chloro tolutenes, trichloro ter-buthyl alochol, and triphenyl chloro methane were listed in the Table 1-3.



Log (T) sec.

Fig. 2. Inorganic 38Cl yields v.s. irradiation time of the neutron irradiated phenyl halides(Liq.).

Remarks: points on the curves denote the followings;

- · Mono chlore benzene,
- × Cyclohexyl chloroide,
- ▲ m-dichloro benzene, and
- 1, 2, 4, -trichloro benzene, respectively,

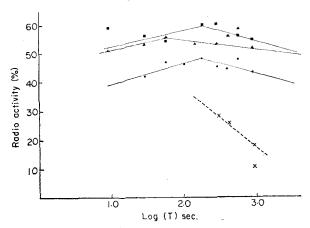


Fig. 3. Inorganic ³⁸Cl yield v.s. irradiadiation time of the neutron irradiated nitro chloro benzenes(Solid).

Remarks: points on the curves denote the followings.

- · 2,4-dichioro nitrobenzene
- × 2, 3, 4, 5, 6, -pentachloro nitro benzene,
- ▲ 2-nitro 4-chloro anisole, and
- 2-nitro 4,6,-dichloroanisol, respectively.

samples gave irregular and abrupt variations of their inorganic yields at the early stages of the irradiation, but were gradually becoming of regular as the irradiation time was

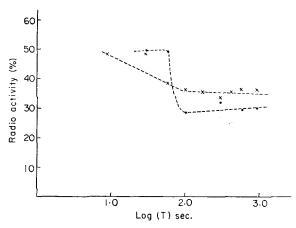


Fig. 4. Inorganic ³⁸Cl yields. v.s. irradiation time of the neutron irradiated chloro toluenes and benzyl halides. (Liq.)

Remarks: points on the curves denote the followings:

- · denote p-chloro toluene and
- × denote benzyl chloride, respectively.

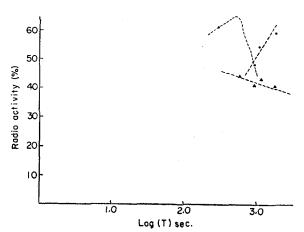


Fig. 5. Inorganic ²⁸Cl yields v.s. irradiation time of the neutron irradiated aromatic chloro phenol and acid.

Remarks: Points on the curves denote the followings:

- · denotes p-chloro benzoic acid and
- ▲ denotes pentachloro phenol, respectively.

extended. (see Fig. 2.4.) In cases of solid samples they could, surprisingly, give a near linear relation-ship between the inorganic yield and the irradiation time, the gradiant of which was relatively of samall(see Fig. 1.3.) In cases of aromatic halides with functional

substituents on the benzene ring such as nitro, amino, methyl, methoxy, carboxylic, and hydroxy groupes, this tendency of the reaction became less definite, but the linear relationship could be maintained at a certain interval of the irradiation time, which was sufficient enough to predict a linear tendency of the reaction mode. (see Fig. 3.5.) These results may be due to the simplicity of the recoil reaction in a solid matrix of an organic compound as reported for other systems in papers^{6,9)}.

Probably, the liquid sample of an organic compound gave a more complicacy of the recoil reaction, which may be due to the high reactivity of a recoiled atom near at the liquid moiety of the organic portion of the sample as explained by Willard *et al.* 10) (The moiety of a solvent cage.)

In the point of a labelling procedure a solid sample would be better with respect to obtainning a steady and higher yield of the labelled product at the early stage of the irradiation than those of a liguid sample, whereas a liquid sample would be able to afford a relatively steady yield at the prolonged irradiation. (More than 10 minutes) Even a simplicity of the contents of the organic phase of a irradiated sample of the solid state may be expected, since the linear relationship between the inorganic yield and the irradiation time is quite definite for a soid sample.

(2) Aromaticity of a Sample

As shown in the Fig. 2, cyclohexyl chloride showed a higher inorganic ³⁸Cl yield than those of the corresponding aromatic chlorides such as monochloro benzene, dichloro benzene, and trichloro benzene at the entire ranges of the irradiation time studied. The recoiled ³⁸Cl atom in an aromatic chloride had, probably, less opportunity to react with reactive hydrogen atom or molecular fragments to produce inorganic chlorides since a benzene ring is

known to be stable towards an ionizing radiation²⁾(table 3.) Interestingly, chloro nitro benzenes both of a liquid and solid state gave a high inorganic yield close to that of cyclohexyl chloride. Since the electron density of the benzene ring of these systems is remarkably decreased as compared with that of chloro benzene derivatives, the recoiled 38Cl atom in these systems would not be reacted with the π electron to form a sufficient quantity of a π complex to promote Cl- ³⁸Cl exchange reaction in the organic phase, if Kontis³⁾ were proper to predict a π complex intermediate for Cl- 38Cl reaction in a recoil reaction. Thus, an inactivated benzene ring may decrease the exchange reaction between Cl-38Cl in the organic phase of a recoil reaction and hence increase the inorganic 38Cl yield. 'This may be an useful information to review the feasibility of labelling an aromatic chloro derivatives with respect to the aromaticity of a sample, that is the density of electrons on the ring.

Foregoing discussions on the radiation stability of a benzene ring should be reevaluated in these aspects, since the chlorine atoms on a benzene ring are known to increase the electron density considerably by a resonance contribution of the chlorine atom to the structure of the benzene ring.

(3) Influence of a Hydrogen Atom in a Molecule

Generally, poly substituted chloro derivatives with or without other functional groups gave a low yield of inorganic ³⁸Cl as shown in the case of trichloro benzene, pentachloro nitro benzene, and tetrachloro anisidine. (see Fig. 1-5) This is, however, not true for the case of pentachloro phenol which has a labile hydrogen atom on the phenylic group. Furthermore, chloro aniline derivatives gave a very high inorganic yields, though the electron density of the benzene ring is remarkably

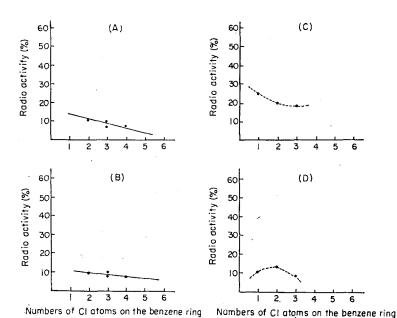


Fig. 6. Inorganic ³⁸Cl yields v.s. number of chlorine atoms on the benzene ring of the neutron irradiated phenyl halides.*

* Remarks: Curves A and B represent the solid sample irradiated for 10 minutes and 1 minute, respectively. Curves C and D represent the liquid sample irradiated for 1 minute and 10 minutes, respectively.

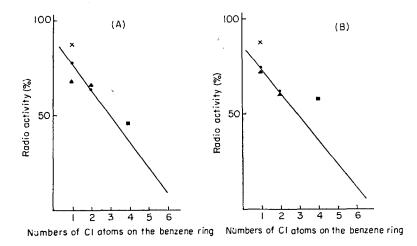


Fig. 7. Inorganic 38Cl yields v.s. the number of chlorine atoms on the benzene ring of the neutron irradiated chloro aniline derivatives.

Remerks: Curve A represent the sample irradiated for 10 minutes, whereas the curve B for 1 minute. Points on the figure denote as follows: x for the liquid state of chloro aniline, ▲ for chloro anisidne, and ■ for tetra chloro anisidne. As shown in the figures, anisidine deivatives show a slight deviation from the linearity, but the deviation becomes very minor when the sample is irradiated for 1 minute. These observations revealed that the yields depend on the chemical structure at a prolonged irradiation time.

increased by the amino group on the ring. These may be due to a reaction between the labile hydrogen or hydrogen atoms on the ring and recoiled ³⁸Cl atoms to form hydrogen chloride. Chloro toluence and benzyl chlorides gave also a lower inorganic yield than that of the corresponding chloro benzenes, though the benzene rings of these systems have a relatively high electron density.

This may be another information for labelling application.

(4) Radiation Reactitivity of the Functional Group on a Benzene Ring

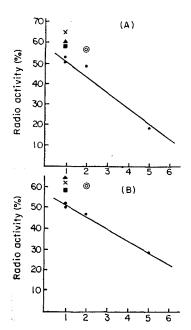
As shown in the Fig. 5, p-chloro benzoic acid gave a quite irregular and high inorganic yield at the later parts of the irradiation time studied. Since the decrease of electon density on the benzene ring is not so drastic enough to increase the inorganic yield as the case of nitro chloro derivatives, the increase of the inorganic yield may mainly be due to the abstract of hydrogen of the carboxylic group by the recoiled 38Cl atom and subsequent decarboxylation of the carboxylic ion to produce stable fragments of molecules in the system, which may produce inorganic chlorides with the recoiled Cl atom during the course of the irradiation. A decarboxylation of a carboxylic ion under an ionizing radiation is well known in many cases of radiation processes11).

Unfortunately, a study along this line could not be extended to other types of the functional group on the ring than the carboxylic group because of the availability of compounds. However, it may be said that a radiation sensitive functional group on a benzene ring increase the inorganic ³⁸Cl yield of the Szilard Chalmer reaction of the present type.

(5) Structural Isomerism

Isomers such as o-, m-, and p- chloro toluenes showed a very minor differences in their inorganic yields when they are irradiated

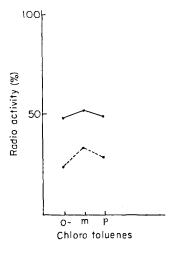
for 1 minute. (Fig. 9) These differences in the inorganic yield become little larger but still small enough to ignore any significant ortho effect. The same is true for the cases of 1,2,4,-, 1,2,3,-, and 1,3,5,- trichloro benzenes, where the large size of chlorine atoms on the ring is usually expected to exert a greater ortho effect than the case of chloro toluenes. Especially, trichloro benzenes of a solid state gave an extremely minor difference in the inorganic yield when the sample



Numbers of CI atoms on the benzene ring

Fig. 8. Inorganic ³⁸Cl_yields v. s. the number of chlorine atoms on the benzene ring of the neutron irradiated nitro chloro benzene derivatives.

Remarks: Curve A represent the sample irradiated for 10 minutes, whereas the curve B represents the sample irradiated for 1 minute. Points on the figures denote as follows: x and denote the corresponding liquid samples and and denote anisole derivatives. As shown on the curve anisole and liquid sample showed a significant deviation from the linearity and the deviation was not changed according to the irradiation times. Presumably, the electron density on the benzene ring infulenced on the yield.



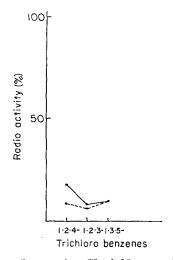


Fig. 9. Inorganic ³⁸Cl yields v.s. the position isomers of the neutron irradiated chloro toluenes.

Fig. 10. Inorganic ³⁸Cl yields v.s. the position isomers of the neutron irradiated trichloro benzenes.

*Remarks: The full line denotes the sample irradiated for 1 minute, whereas the dotted line represents the sample irradiated for 10 minutes. In case of trichloro benzenes, 1, 2, 4, -derivative is liquid and showed a severe change in the yield, whereas 1, 2, 3, - and 1, 3, 5, -derivatives of solid states did not much change in the yields.

was irradiated for 10 minutes. These observations revealed that a rigid structure of the molecule at the present system had no influence on the inorganic ³⁸Cl yield, whereas Shimojima⁴ had observed a significant increase of the inorganic yield for the case of β-BHC, the structure of which is said to be very rigid from a stereochemical point of view. Presumably, the coplanar structure of a benzene ring would exert a less steric effect towards a recoiled ³⁸Cl atom than the case of a cyclohexyl ring.

(6) Number of Cnlorine Atoms on the Benzene Ring

As shown in Fig. 6-8, the inorganic ³⁸Cl yields of the Szilard Chalmer reactions of phenyl chlorides, nitro chloro benzenes, and chloro anilines showed a linear relationship with the number of chlorine atoms on the benzene ring. This relationship was, especially, pronounced at a shorter irradiation time (less than 1 minute) and in a solid sample. As it may be understandable from the exsiting

concepts of conventional electronic, steric, and resornance effects of a chlorine atom on a benzene ring, the number of chlorine atoms on a benzene ring are able to represent the chemical moiety of the ring and hence it may be considered as a scale of the causes of discussions described hereto. Difficulties of elucidating the casuses specifically will be encountered to discriminate each cause individually in a Szilard Chalmer process like the present case, because each cause is mingled together showing a multiplicity in a system. However, the linear relationship shown in Fig. 6-8 may encourage that these causes may be summarized and a general procedure of labelling the present system with ³⁸Cl may be predicted pending that a detailed separation of the organic phase is achieved.

In conclusion the following considerations would be essential to label aromatic chloro derivatives with ³⁸Cl atom by a Szilard Chalmer process. A high electron density on the benzene ring, a solid state of the sample, a

prolonged irradaition time for a liquid sample, and the highest number of chlorine or substituents on the ring would favor a low yield of the inorganic ³⁸Cl, whereas a labile hydrogen in the substituent on the ring, a non aromaticity of the ring, and a radiation sensitive functional group substituted would increase the inorganic ³⁸Cl yield. A structural isomerism of the system would result a minor difference in the inorganic yield.

These conceptions may, of course, overlap in a system to make a specific prediction of the labelling yield difficult, but a general tendendy of a labelling procedure would well be of predictable if these conceptions are scrutinized as described hitherto.

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References

- A. Black and A. Morgan, Int. J. Appl. Rad. & Rad. Isotp. 21 5-10(1970)
- G. Stocklin and W. Tornau, Radiochim, Acta.
 9 95 (1968)
- 3) S.S. Kontis and D.S. Urch, Ibid. 15 21(1971)
- H. Shimojima and T. Ko. Lim, J. Nucl. Sci & Tech. (Tokyo) 8 (4) 179(1971)
- N. Matsuura, N. Shinohara, and, T. K. O., Lim, J. Inorg. Nucl. Chem. 31 1251(1969)
- J. A. Knight, Jr., Contract AT(40-1) 3519(c.f.
 Nucl. Sci. Abstr. # 11584(U.S. AEC. 1970))
 and Related Literatures
- 7) Y.S. Kim., Unpublished Results(1972)
- 8) Ishikawa, et al. "The thin layer chromatography" p. 224 (Nanko Do. Tokyo, Japan, 1968)
- J. Otterbachm, Nucl. Sci. Abstr. No. 47866
 (U.S. AEC. 1972) and references cited therein.
- 10) J.E. Willard, et al J. Phys. Chem. 62 1351 (1958) and subsequent papers.
- 11) J. A. Russell. Radiat. Res. 50 No.1 41-7(1972)