Development of New Releasing Agents for Preparation of Thin Self-Supporting Target Films

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Several kinds of materials were examined for the usefulness as releasing agents in the preparation of various thin self-supporting target films for use in nuclear reaction experiments. NaCl, BaCl₂, KCl, CsI, Teepol, glucose, KIO₃, mica, nitrocellulose or Formvar was deposited onto glass plates as the release agent by vacuum evaporation or dipping method. The obtained target film was tested on impurities from the release agent by using nuclear reactions. The relative effectiveness of each release agent was also considered from ease in the stripping of target films.

1. Introduction

In recent studies of nuclear physics, the target materials for use in nuclear reaction experiments have been required as thin, pure and uniformed self-supporting foils less than a few mg/cm² in thickness. Especially, it is required to use very thin targets (less than several hundred µg/cm² in thickness) in the case of high resolution experiments by using high quality beams. Usually, such target foils are obtained by means of vacuum evaporation. It is hard practically to prepare self-supporting foils of such materials as alkali and alkali earth metals. These materials must be supported by other backing foils. The contamination in the target films from release agents will become serious in the measurement of small reaction cross sections, because only a slight amount of impurities with low atomic numbers gives rise to a large background owing to their low Coulomb Barriers. Various release agents for making self-supporting thin target films have been reported in some papers. NaCl, CsI, BaCl₂ and PCB-100 were investigated by D.N. Braski and glucose by T. Yamaya, et al.

In addition to above release agents, we report the results on KCl, KIO₃, mica and nitrocellulose, in this paper. Target films were analyzed for contaminants from the release agents by using nuclear reactions with the proton beam from INS FM-Cyclotron.

The most effective release agent was also looked for in the separation of the evaporated carbon layers from substrates (micro slide glasses). Release agents tested were as follows; NaCl, BaCl₂, CsI, KCl, KIO₃, Formvar, Teepol, mica and nitrocellulose. Thin layers of them were formed by vacuum evaporation or coating on a substrate. Carbon was chosen as the films to be released for the test of the relative effectiveness of each agent, since carbon films of several µg/cm² in thickness are rather strong and stable, and since they can be prepared easily in a laboratory and frequently used as backing foils for the target. The relative effectiveness of each release agent was determined by ease in the stripping process.

2. Experimental Procedures

2.1 Cleaning of substrates

The 25×76 mm slide glasses for use in optical microscope were used as the substrate. The slide glasses were cleaned ultrasonically in trichloroethylene for about 30 minutes. They were, then, cleaned by Teepol detergent, rinsed sufficiently in distilled water and finally wiped with gauze. The cleaned slide glasses were transferred into vacuum chamber as soon as possible to avoid possible contamination with dust particles, which resulted in pin
holes in the film.

2.2 Release agents

Substrates were coated with release agent by dipping the slide glasses in solutions of Teepol and glucose in distilled water, nitrocellulose in acetone, and Formvar in ethylene dichloride. The substrates were smeared as uniformly as possible after being dried in a room and polished carefully with a clean cloth until no visible traces of the release agents were observed.

The release agents of BaCl$_2$, NaCl, CsI, KCl and KIO$_3$ were formed by vacuum evaporation from a tantalum boat (80 μm in thickness, 10 mm in width and 50 mm in length) onto the cleaned slide glasses, which were placed at 25 cm from the boat. The thickness of the release agents was several ten μg/cm$^2$. Mica was also used as a release layer itself, in which the surface of mica was wiped wholly with gauze wet with pure acetone.

2.3 Preparation of carbon foil

Several methods$^{4-29}$ have been developed for the preparation of very thin carbon foils by vacuum evaporation. With these methods self-supporting carbon foils as thin as 20 μg/cm$^2$ can be obtained. The method used in the present experiment is a modification of the Bradly and Dearnaley method. We have thus easily prepared carbon foils of several tens μg/cm$^2$ in thickness.

A 10 in. vacuum evaporator was used for making thin carbon foils. Two carbon rods of spectroscopical grade with 6 mm in diameter were set in an evaporator vessel as arc electrodes. These carbon rods were supported with tantalum pipes which were also of high purity so as to reduce outgassing during the evaporation. One of the carbon rods was a column and the other was of a truncated conical end to make a point contact with each other. Springs were used to maintain a light contact of the rods during the evaporation.

Carbon was evaporated onto the slide glasses which had been already thinly covered by coating or evaporation with release agents. The arc-current of 100 to 150 A at 5 V was supplied for about 5 min, in 10$^{-4}$ Torr vacuum. The operation was repeated several times in the case to get thicker carbon films. Carbon layer with grey-brown color of semitransparency was thus deposited on the glass plate. Then, the carbon was stripped off by sliding the glass plates very slowly into warm distilled water (∼35°C) with the angle of about 30° to the water surface as shown in Fig. 1. After the release agent was dissolved, the foil floated on the water and was picked up carefully by a target holder.

![Fig. 1 Schematic diagram for the separation of carbon film from the substrate.](image)

2.4 Measurement of impurities

The measurement has been performed to observe the residual release agents contained in the carbon foils. The energy spectra have been obtained by irradiating these carbon foils with 52-MeV protons from INS FM-Cyclotron. The amount of the residual impurities were estimated from the scattering kinematics and yield. The particles were detected by a broad range magnetic spectrograph with 200-folds proportional counter array. We have also checked the elements contained in the release agents by bombarding very pure and thin aluminum foils coated with the agents.

Fig. 2 shows a typical energy spectrum of the reaction particles from a carbon foil when NaCl was used as a release agent at $\theta_{lab} = 165°$ bombarded by 52 MeV protons. The foil thickness was about 25 μg/cm$^2$. The amounts of silicon and oxygen shown in Fig. 2 were estimated to be about 3 μg/cm$^2$ and 1 μg/cm$^2$. 

(4)
respectively. These impurities probably came from the pumping oils of the vacuum systems and the distilled water used for the carbon film separation. No impurities except silicon and oxygen are observed in Fig. 2.

Fig. 3 shows a typical energy spectrum of the reaction particles from a carbon foil made by using a Teepol as release agent. The thickness of the carbon film was about 40 µg/cm². Many peaks from the impurities other than carbon are observed. These impurities were identified as the elements contained in Teepol, which was confirmed by measuring the energy spectra of the thin aluminum foils dipped in Teepol solution. These impurities could not so easily removed even by washing the foils with hot distilled water. Only Si of about 5 µg/cm² in thickness was observed in the carbon foil made by the thermal cracking method. The observed contaminants in carbon foil are shown in Table 1.

The experimental conditions and results of

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**Table 1 Experimental result on impurities**

<table>
<thead>
<tr>
<th>Release agents</th>
<th>Observed elements</th>
<th>Amount of elements (µg/cm²)</th>
<th>Release agents</th>
<th>Observed elements</th>
<th>Amount of elements (µg/cm²)</th>
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</thead>
<tbody>
<tr>
<td>NaCl</td>
<td>O, Si</td>
<td>~4, ~1</td>
<td>CsI</td>
<td>O, Si, Na</td>
<td>~9, ~5, ~7</td>
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<tr>
<td>BaCl₂</td>
<td>O, Si, Cl</td>
<td>~12, ~4, ~5</td>
<td>Teepol</td>
<td>O, Si, N, Na, Al</td>
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<tr>
<td>KCl</td>
<td>O, Si, Cl</td>
<td>~10, ~4, ~7</td>
<td>Glucose</td>
<td>O, Si, N</td>
<td>~10, ~7, ~8</td>
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<tr>
<td>KIO₃</td>
<td>O, Si, N</td>
<td>~17, ~5, ~10</td>
<td>Mica</td>
<td>O, Si, Al, Cl</td>
<td>~14, ~8, ~5, ~8</td>
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Table 2 Experimental conditions and results

<table>
<thead>
<tr>
<th>Release agent</th>
<th>Description</th>
<th>Method of application</th>
<th>Substrate temperature (°C)</th>
<th>Release film thickness (μg/cm²)</th>
<th>Release* agent ability</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl</td>
<td>Crystal grain</td>
<td>Vacuum evaporation</td>
<td>20, 30</td>
<td>30, 100</td>
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<tr>
<td></td>
<td>Reagent grade</td>
<td></td>
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</tr>
<tr>
<td>BaCl₂</td>
<td>Reagent grade</td>
<td>Vacuum evaporation</td>
<td>20, 30</td>
<td>30, 100</td>
<td>B</td>
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<tr>
<td>Formvar</td>
<td>Dissolved in</td>
<td>Dipping</td>
<td>20</td>
<td>Dipping -50% (wt) solution</td>
<td>D</td>
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<tr>
<td></td>
<td>ethylene dichloride</td>
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<td></td>
</tr>
<tr>
<td>Teepol</td>
<td>Dissolved in</td>
<td>Dipping</td>
<td>20, 30</td>
<td>100, 300</td>
<td>C</td>
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<td></td>
<td>distilled water</td>
<td></td>
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<td></td>
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<td>350</td>
<td>C</td>
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<tr>
<td>Mica</td>
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<td>CsI</td>
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<td></td>
<td>Reagent grade</td>
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<tr>
<td>KCl</td>
<td>Reagent grade</td>
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<td>20, 250</td>
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<tr>
<td>KIO₃</td>
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<td>30</td>
<td>C</td>
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<td>Nitrocellulose</td>
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<td></td>
<td>acetone</td>
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</table>

* The Remarks of A, B, C, D and E indicate as following:
A: Excellent (very fast, smooth separation: excellent target films)
B: Good (fast separation: good target films)
C: Fine (slow separation: some chance of target film breakage)
D: Poor (very slow separation: high probability of target film breakage)
E: Failure (target films would not separate from substrate)

The stripping test are summarized in Table 2.

3. Discussion

NaCl and CsI were found to be better than Teepol for use in floating off the thin layers of target materials which were formed by vacuum evaporation. The main requirement of the release agent is to separate target film from a substrate easily. The target films should be floated off as quickly as possible after being removed from the vacuum chamber.

In target preparation, Teepol has been most commonly and frequently used as release agent. Teepol, however, has two demerits. Primarily, Teepol is apt to be deteriorated by the heat radiation of boat or crucible because the melting point is low (probably 100~200°C). Secondly, it is very difficult to be smeared uniformly. For these reasons Teepol should be used only for the evaporation at low temperature. We found that glucose have also the same properties as Teepol.

We have obtained thin, pure, uniform and as large as 15×15 mm in area self-supporting films of Al, Au, Ag, enriched isotopes of ¹⁰,¹¹B, ⁵⁸,⁶⁰,⁶²Ni, ¹¹⁶,¹¹⁸Sn and Y, etc. using NaCl and CsI as release agents. NaCl and CsI can be recommended as excellent release agents for target preparation for use in nuclear reaction experiments. NaCl and CsI are very soluble in water compared with Teepol and glucose. The main advantage of NaCl as release agent is that it can be evaporated onto slide glasses very uniformly without jump out of the boat in initial heating. The melting points of NaCl...
and CsI are about 800°C and 700°C, respectively, which make it ideal for preparation of films of high melting point materials. The thickness of NaCl and CsI layers is controlled very easy as compared with Teepol, glucose, Formvar and nitrocellulose. The most suitable thickness of NaCl and CsI layers was found to be in the range of 20-30 μg/cm².

The carbon films made with release agents of the nitrocellulose and Formvar could be separated. The carbon films from the KIO₃ were separated very slowly and sometimes broken off.

In the case of mica, a rapid separation was possible for films of large areas. However, a film thicker than several hundred μg/cm² was not successfully separated.

Separations using BaCl₂ and KCl as release agents were relatively good. However, sometimes the separation was difficult and needed a long time.

The thicknesses of release agents deposited were found to have an important influence on the strength of the carbon films, the speed of floating off and the surface roughness of the obtained films. For the alkali halide release the separation should be carried out as soon as possible, since the release agent layers deliquesce in air.

As a summary, it was found to be effective to use the NaCl or CsI release agents rather than KIO₃, KCl and BaCl₂.

The main advantages of NaCl and CsI release agents are as follows:
1) Self-supporting films are easily obtained for many kinds of target materials.
2) Large range of thickness from several ten μg/cm² to about several mg/cm² could be made by using these agents with an area up to 25 mm×25 mm.
3) The targets thus prepared have good uniformity in thickness with less than 5% deviation, and are mechanically very strong.
4) The targets contain negligible amount of impurities.

References
2) T. Yamaya, T. Tohei and S. Morita: *ibid.*, 49, 173-75 (1967)
5) P. Maier-Komor: *ibid.*, 102, 485-86 (1972)

要 旨

うすいセルフサポーティングのターゲット膜作成のための新しい剝離剤の開発

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原子核物理実験に使用するセルフサポーティングのターゲット膜を作成するときに用いる数種類の剝離剤の有効の度合を調べた。塩化ナトリウム、塩化バリウム、塩化カリウム、ヨウ化セシウム、ティボール・ブドウ糖、ヨウ素酸カリウム、雲母、ニトロセルロースおよびフォルンパールの剝離剤をスライドガラスに真空蒸着あるいはディピング法を用いて付着させた。

ターゲット膜作成過程で剝離剤からそのターゲット中に混入すると思われる不純物の元素とその量を原子核反応の弾性散乱法を用いて測定した。またおおむね剝離剤の有効の度合についてはターゲット膜の剝離の容易さによって決めた。