

Preparation and Properties of Some Bis(triphenylphosphine)-iminium Salts, $[(\text{Ph}_3\text{P})_2\text{N}]\text{X}$

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A number of bis(triphenylphosphine)iminium salts, $[(\text{Ph}_3\text{P})_2\text{N}]\text{X}$, abbreviated $[\text{PNP}]\text{X}$, of small inorganic ions have been prepared. All the salts, except the fluoride, can be prepared from the easily available $[\text{PNP}]^+\text{-chloride}$ by precipitation with excess alkali metal salts in water.

The salts are simple to purify and are non-hygroscopic. Their solubilities in a number of dipolar aprotic solvents suggest that the $[\text{PNP}]^+$ -cation is a valuable alternative to the usually employed onium cations for synthetic and kinetic studies in this class of solvents.

In recent years various tetraorganoammonium, phosphonium and arsonium cations in addition to numerous crown ether complexes of the alkali metal cations have been used extensively as counter ions.¹⁻⁵ Salts of these ions may be both very soluble and well dissociated in a number of aprotic solvents^{6,7} allowing reactions to be performed homogeneously and even to be studied kinetically.⁸ The weakly polarizing effect to these cations allows the isolation of a number of salts of anions which otherwise would not be possible; for a review, cf. Ref. 2.

Unfortunately, these cations have their disadvantages; difficulties in preparing various salts in their pure states,⁹ solubility and hygroscopicity of their salts,^{9,10} the ability of some of these cations to catalyze oxidation reactions,^{11,12} and, in the case of Ph_4P^+ , Ph_4As^+ and especially crown ethers, their exorbitant cost.

As part of a program to study the solution behaviour and structural properties of certain inorganic anions we have synthesized a number of salts of the bis(triphenylphosphine)iminium

cation, $[(\text{Ph}_3\text{P}_2\text{N})^+]$,¹³ abbreviated $[\text{PNP}]^+$.^{*} We offer our results as proof of the utility and facility of $[\text{PNP}]^+$ as a valuable counter ion.

Preparation of $[\text{PNP}]^+$ -salts. $[\text{PNP}]^+\text{-chloride}$, which serves as the starting material for the preparation of the various $[\text{PNP}]^+$ -salts, can now be readily obtained in high yield, > 90 %.¹⁵ It is the experience of these authors that it is considerably simpler to prepare this compound than $\text{Ph}_4\text{P}\text{Cl}$ and $\text{Ph}_4\text{As}\text{Cl}$. Although $[\text{PNP}]\text{Cl}$ is prepared from the rather costly triphenylphosphine and the maximum total conversion into $[\text{PNP}]\text{Cl}$ is only ~60 % (3 mol of Ph_3P yield 1 mol of $[\text{PNP}]\text{Cl}$ ¹⁵), this compound should compare favourably with $\text{Ph}_4\text{P}\text{Cl}$ and $\text{Ph}_4\text{As}\text{Cl}$ with regard to cost.

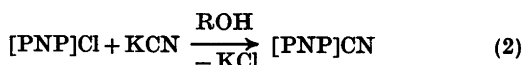
Part of the facility by which $[\text{PNP}]^+$ -salts can be employed comes from the fact that a salt of a desired anion, X^- , can be rapidly obtained in high yield, > 80 %, by precipitation with excess alkali metal salts, MX , in warm water according to eqn. 1:

* The bis(triphenylphosphine)iminium ion is the usual name for this ion, but since this ion is fundamentally not an iminium ion, various names have been suggested: The bistrisphenylphosphinenitride ion,¹⁴ the μ -nitrido-bis(triphenylphosphorus) (1+) ion,¹⁵ the bis(triphenylphosphoranylidene)ammonium ion,¹⁶ the bis(triphenylphosphoranylidene)iminium ion,¹⁷ and the hexaphenyldiphosphazanium ion.¹⁸ *Chemical Abstracts* recommends "triphenyl-(*P,P,P*-triphenylphosphine imidato-*N*)phosphorus (1+)", a name which is not particularly handy. To avoid confusion we will only use $[\text{PNP}]^+$. Some authors have used the abbreviation PPN^+ , but since this formulation may indicate that the positive charge resides on the nitrogen atom, which is not the case,¹⁹ this abbreviation should be avoided.

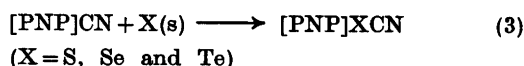


The cause for this simple anion exchange procedure is that most $[\text{PNP}]^+$ -salts, except the fluoride, have only a limited solubility in water at room temperature while $[\text{PNP}]\text{Cl}$ is exceptionally soluble in warm water, ~ 100 g/l at 80°C . Even at 25°C is the solubility of $[\text{PNP}]\text{Cl}$ considerable, 7.1 g/l, as judged from the UV spectrum of a saturated aqueous solution.

The cyanide, $[\text{PNP}]\text{CN}$, may also be prepared according to eqn. 1. However, in order to avoid co-precipitation of the fairly insoluble hydroxide, the cyanide can only be precipitated slowly from dilute solutions. This compound is therefore easier to prepare from the chloride and excess potassium cyanide in dry methanol or ethanol taking advantage of the high solubility of $[\text{PNP}]^+$ -salts in this class of solvents; eqn. 2:



From the cyanide, the thiocyanate, the selenocyanate and the tellurocyanate are easily prepared in acetone or acetonitrile according to eqn. 3:

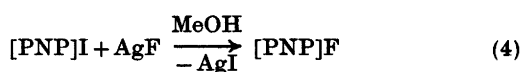


The thiocyanate and the selenocyanate may be prepared as well according to eqns. 1 and 2.

The procedure depicted by eqn. 2 is especially convenient for the synthesis of small quantities of the cyanide. Thus, from the costly potassium ^{13}C cyanide a high molecular weight

^{13}C cyanide is readily available. With the $[\text{PNP}]^+ - ^{13}\text{C}$ cyanide in hand, the corresponding thiocyanate, selenocyanate and tellurocyanate can be prepared as depicted by eqn. 3. In Table 1 are listed the ν_{XCN} in the 2000–2200 cm^{-1} region for the ^{13}C and the ^{13}C pseudo-halide ions.

The fluoride, $[\text{PNP}]\text{F}$, cannot be prepared from $[\text{PNP}]\text{Cl}$ and excess potassium fluoride, eqn. 1. Upon cooling of the reaction mixture only a highly viscous solution is obtained. However, from the iodide and silver fluoride in methanol, the nicely crystalline fluoride can be obtained in high yield, eqn. 4:



Properties of $[\text{PNP}]^+$ -salts. In Table 2 are listed the various $[\text{PNP}]^+$ -salts prepared in the present study. All the salts have rather sharp and well-defined melting points or decomposition temperatures. In recent years a number of $[\text{PNP}]^+$ -salts have been prepared, notably salts of heavy metal carbonyl ions,^{15,22–24} but also of anions of carboxylic acids²⁵ and various main group inorganic anions.^{26–28} Several X-ray structure determinations have been performed; for a survey of references, cf. Ref. 29. When considering the range of anions with which the $[\text{PNP}]^+$ -cation makes stable and well-crystalline salts, the versatile nature of this ion as a counter ion in synthesis seems well established.

The success of this ion as a counter ion for the preparation of stable salts seems to be due to two factors. The positive charge of the cation is well distributed in the triphenylphosphine moieties¹⁹ causing this cation to have a negligible polarizing effect. In this respect this cation should be comparable with other bulky onium cations, notably the tetraphenylphosphonium cation, Ph_4P^+ , and the tetraphenylarsonium cation, Ph_4As^+ . The major advantage of the $[\text{PNP}]^+$ -cation, however, comes from its apparently exceptional hydrophobic nature causing all known salts of this cation to be non-hygroscopic. No decomposition of potentially unstable anions will thus be initiated by the presence of polarizing water molecules. A number of Ph_4P^+ - and Ph_4As^+ -salts are well-known to readily pick up 1 to 3 mol of water.^{10,20}

Table 1. IR data of $\text{X}[^{12}\text{C}]\text{CN}^-$ and $\text{X}[^{13}\text{C}]\text{CN}^-$ in acetonitrile (X = S, se and Te).

	ν_{max} cm^{-1} ^a	$\Delta\nu_{\frac{1}{2}}$ cm^{-1} ^b	$A \times 10^{-4}$ $\text{M}^{-1} \text{cm}^{-2}$ ^b
$\text{S}[^{12}\text{C}]\text{CN}^-$	2058	12	4.1
$\text{S}[^{13}\text{C}]\text{CN}^-$	2011		
$\text{Se}[^{12}\text{C}]\text{CN}^-$	2067	10	2.2
$\text{Se}[^{13}\text{C}]\text{CN}^-$	2021		
$\text{Te}[^{12}\text{C}]\text{CN}^-$	2081	12	1.8
$\text{Te}[^{13}\text{C}]\text{CN}^-$	2034		

^a $\pm 1 \text{ cm}^{-1}$, ^b Ref. 20.

Table 2. Melting points and analytical data for some [PNP]⁺-salts.

	M.p. °C	Found (Calc.) %
Cl ⁻	273–274 (268–270 ¹⁵)	
Br ⁻	253–255 (256 ²¹)	
I ⁻	252–254 (254 ²¹)	
F ⁻	177–178	C: 77.88 (77.55), H: 5.62 (5.42)
SO ₄ ²⁻	258–261	N: 2.67 (2.51)
S ₂ O ₃ ²⁻	210–212 (dec.)	N: 2.55 (2.39), S: 2.67 (2.73)
MnO ₄ ⁻	~155 (dec. violently)	N: 2.11 (2.36), S: 5.37 (5.39)
CrO ₄ ²⁻	234–235 (dec.)	N: 2.29 (2.35)
ClO ₄ ⁻	259–261 (264–266 ¹⁴)	
NO ₃ ⁻	225–226	N: 4.37 (4.79)
NO ₂ ⁻	233–234 (232 ²¹)	
N ₃ ^{-a}	214–215 (214–216 ¹⁴)	
Ac ⁻	114–115	C: 77.32 (76.37), N: 6.03 (5.57)
CN ⁻	212–214	N: 2.13 (2.34)
OCN ^{-a}	203–204	C: 78.12 (78.70), H: 5.49 (5.36)
SCN ^{-a}	187–188	N: 4.96 (4.47)
SeCN ^{-a}	182–185 (dec.)	N: 4.37 (4.79)
TeCN ^{-a}	190–193 (dec.)	C: 64.72 (64.20), H: 4.18 (4.37)
		N: 3.55 (4.05)

^a Determined by liquid IR in acetonitrile in the 2000–2200 cm⁻¹ range to be >98 % pure.³⁰

Several of the salts listed in Table 2 may be purified by crystallization from water. Although [PNP]⁺-salts generally do not crystallize with crystal water, (one known example, with the squarate²⁵), it is our experience that the removal of surface water in high vacuum at elevated temperature is quite time-consuming. Furthermore, some of the [PNP]⁺-salts, the bromide, the selenocyanate and especially the iodide, are not sufficiently soluble in water to apply this solvent as a convenient crystallization agent. (1 l of boiling water dissolves less than 1 g [PNP]I and the solubility product of this salt at 25.0 °C is only 3.0×10^{-9} M², 0.035 g/l). It is therefore recommended that an organic solvent be applied as crystallization agent whereby traces of co-precipitated alkali metal salts are removed as well. Acetone seems to be the best choice, even though only the iodide and the tellurocyanate of the compounds listed in Table 2 can be recovered from this solvent alone without excessive losses. No evidence for solvated acetone in any of the synthesized [PNP]⁺-salts could be found; [PNP]⁺-salts are known to crystallize as solvates from organic solvents like chloro-

benzene,³⁰ acetonitrile,³⁰ benzonitrile,³¹ and dichloromethane.¹⁵

The [PNP]⁺-salts are generally very soluble in the usual dipolar aprotic solvents; acetone, acetonitrile, dimethyl sulfoxide, dimethylformamide, benzyl cyanide, nitromethane and nitrobenzene. Actually, their solubilities in most dipolar aprotic solvents prevent these solvents alone to be used as crystallization agents. Due to their considerable solubility in this class of solvents [PNP]⁺-salts may therefore serve as convenient sources of anions in substitution reactions (the nucleophilic anions), in salt-effect studies (the non-nucleophilic anions), in elimination reactions³² (the fluoride) and in oxydation reactions³³ (the chromate and the permanganate). The [PNP]⁺-salts are even very soluble in 1,1,2,2-tetrachloroethane and in dichloromethane. However, care should be exercised when employing dichloromethane since the most nucleophilic anions, notably the selenocyanate ion and the azide ion, react fairly rapidly with this solvent.³⁴ For this reason, [PNP]TeCN decomposes rapidly when dissolved in dichloromethane.

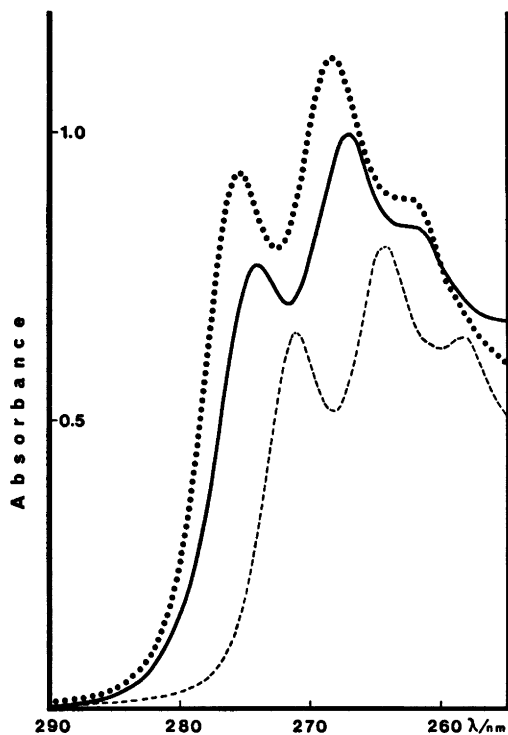


Fig. 1. UV spectra of 2.5×10^{-4} M solutions of [PNP]Cl (—), Ph_4AsCl (---) and $\text{Ph}_4\text{P}^+\text{Cl}$ (---) in acetonitrile in the 255–290 nm region. Path-length, 1 cm.

Stability of the [PNP]⁺-cation. The [PNP]⁺-cation seems to be completely unaffected by molecular oxygen and does not seem to be attacked by strong nucleophiles, *e.g.* the cyanide ion, in dry dipolar aprotic solvents. With ionic cyanide in moist acetone, however, a very slow decomposition takes place at reflux temperature. Presumably, the cation is very slowly attacked by the hydroxide ions present and triphenylphosphine oxide, Ph_3PO , is formed (see Experimental). It has previously been shown that

the [PNP]⁺-cation is attacked only very slowly by a warm concentrated solution of sodium hydroxide.²¹

Toward mineral acids the [PNP]⁺-cation is reported to be completely stable.²¹ No decomposition of [PNP]⁺-salts seems to take place during drying at elevated temperatures. The stability of the [PNP]⁺-cation may thus equal that of the Ph_4As^+ -cation. Since the Ph_4P^+ -cation is known to be decomposed rapidly by hydroxide ions,²⁵ Ph_4P^+ -salts of anions of weak acids, *e.g.* Ph_4PCN , are difficult to synthesize and can only be used in scrupulously dried aprotic solvents.

Spectral characteristics of the [PNP]⁺-cation. In Fig. 1 is shown the UV spectra of 2.5×10^{-4} M solutions in acetonitrile of [PNP]Cl, Ph_4AsCl and $\text{Ph}_4\text{P}^+\text{Cl}$ in the 255–290 nm region. The three spectra are in principle quite similar exhibiting three main peaks. In Table 3 are listed the absorption maxima positions together with the molar absorptivities (ϵ). The Ph_4P^+ -cation is clearly the most absorbing one; the [PNP]⁺-cation being intermediate. Above 300 nm the absorption due to the [PNP]⁺-cation is negligible. Below 250 nm the [PNP]⁺-cation is by far the most absorbing one of the three ions. Measurements in water on [PNP]Cl showed a slight blue-shift, 0.3–0.4 nm, but with unaltered extinction coefficients.

The IR spectrum of the [PNP]⁺-cation shows strong absorptions at 803, 995, 1115, 1250(broad), 1440 and 1490 cm^{-1} in addition to the usual aromatic absorptions in the 650–750 and 3050 cm^{-1} regions. No attempt was made to assign the various peaks. The cation appears transparent in the 810–970 cm^{-1} region and in the 1650–3000 cm^{-1} region.

The ^1H NMR spectrum of the [PNP]⁺-cation consists of a complex multiplet in the benzene region while the ^{31}P NMR spectrum consists of a singlet at -22.3 ppm *vs.* phosphoric acid.¹⁵

Table 3. Absorption maxima positions (λ) and molar absorptivities, ϵ , of [PNP]Cl, Ph_4AsCl and $\text{Ph}_4\text{P}^+\text{Cl}$ in acetonitrile in the 255–290 nm region.

	λ_1	$\epsilon \times 10^{-3}$	λ_2	$\epsilon \times 10^{-3}$	λ_3	$\epsilon \times 10^{-3}$	$\lambda(\epsilon < 100)$
[PNP]Cl	262.3	3.35	267.4	4.01	274.3	3.10	287.5
Ph_4AsCl	258.4	2.44	264.6	3.06	271.4	2.48	285
$\text{Ph}_4\text{P}^+\text{Cl}$	263.0	3.21	268.6	4.38	275.7	3.60	291

EXPERIMENTAL

Acetonitrile was purified as previously reported.⁴ Acetone, *p.a.*, absolute methanol and ethanol were used without further purification, 1,1,2,2-Tetrachlorethane, benzene and diethyl ether were dried and purified according to standard procedures. All solvents were flushed with dry nitrogen prior to use.

The potassium and sodium salts were of analytical grade and used without further purification except potassium cyanide, potassium thiocyanate and potassium selenocyanate which were recrystallized and carefully dried prior to use. Potassium [¹³C]cyanide, 90.5 % enriched, was used as received from British Oxygen Co., Ltd.

[PNP]Cl was prepared as described by Ruff and Schlientz¹⁵ and finally crystallized twice from acetone/diethyl ether.

Preparation of [PNP]X. General procedure. (Except for X = F, CN and TeCN). [PNP]Cl, 1 to 2 g, was dissolved in a minimum amount of warm water, 60–80 °C, 25–40 ml, and the warm concentrated solution of KX or NaX, 5–10 g, was then added under constant stirring. (Boiling water frequently caused the product to separate as an oil). The reaction mixture was set aside at 0 °C for 1 h and filtered. The product was dissolved in acetonitrile (traces of water were removed azeotropically with benzene) and finally crystallized from acetone/diethyl ether (ethanol for the thiosulfate).

In the case of the acetate, the chromate, the sulfate and the nitrate, the precipitation procedure in water had to be repeated once to obtain chloride-free products. The chromate was both times precipitated with a sodium chromate solution which was 0.02 M in sodium hydroxide. In the final crystallization of [PNP]₂CrO₄ acetone containing 2 % triethylamine was used to avoid a trace of darkening of the crystals.

[PNP]CN. In water, eqn. 1. To 1.4 g [PNP]Cl, dissolved in 100 ml water was added a solution of 5 g KCN in 25 ml water. The solution was stirred at room temperature for 10 min and set aside for 24 h at 0 °C. Yield of [PNP]CN, 1.15 g, 82 %. *In methanol, eqn. 2.* 2.3 g [PNP]Cl and 0.29 g KCN were dissolved in 25 ml warm methanol. After 2–3 h at 0 °C the precipitated potassium chloride was removed by filtration. To ensure a complete removal of ionic chloride this treatment was repeated with 0.1 g KCN. Yield from acetone/diethyl ether, 1.94 g, 85 %.

All operations with [PNP]CN in acetone or acetonitrile were carefully performed under nitrogen. If oxygen was allowed to enter, a yellow to orange solution was slowly obtained and the IR in the 2000–2200 cm⁻¹ region suggested that ionic cyanate among other products had been formed.

[PNP]TeCN. To 0.40 g [PNP]CN in 20 ml acetone was added 0.5 g black tellurium powder and the reaction mixture heated to 50 °C for

30 min. After filtration the tellurocyanate was precipitated with diethyl ether and finally crystallized from acetone. Yield 0.30 g, 65 %. The thiocyanate and the selenocyanate were prepared in a similar way in close to quantitative yield.

[PNP][¹³C]CN. To 0.85 g [PNP]Cl in 10 ml methanol was added 0.10 g potassium [¹³C]cyanide (5 % excess of cyanide). An additional amount of 0.01 g of the cyanide was added after the first crop of potassium chloride had been removed. Yield 0.72 g.

[PNP]F. 2.0 g [PNP]I in 25 ml methanol and an equivalent amount of AgF was stirred at 40 °C for 2 h. After filtration an additional amount of 0.1 g AgF was added. The reaction solution was repeatedly (six times) evaporated to dryness and redissolved in methanol and filtered until a clear colourless solution was obtained when the residue was dissolved in acetone or methanol. Yield from acetone/diethyl ether, 1.30 g, 60 %. An acidified solution of this salt did not give any precipitate with potassium chloride or silver nitrate.

Attempted decomposition experiments. (1) *[PNP]Cl and molecular oxygen in acetone.* 0.3 g [PNP]Cl in 25 ml acetone was kept at ~50 °C and flushed with oxygen for 2 h. No colouring of the solution was observed and the UV of the solution did not suggest that any decomposition had taken place. (2) *[PNP]CN in acetonitrile.* 0.3 g [PNP]CN in 25 ml acetonitrile, flushed with nitrogen, was kept at ~50 °C for 2 h. A faint yellow colour of the solution developed, presumably due to traces of oxygen having been present. No decomposition products could be detected. (3) *[PNP]CN in acetone/H₂O (acetone/D₂O).* 0.3 g [PNP]CN, dissolved in 10 ml acetone and 1 ml H₂O (D₂O), was refluxed for 3 h in a nitrogen stream. The yellowish solution was evaporated to dryness and treated with benzene. The benzene extract gave 0.015 g of a brownish, sticky residue. IR indicated that this residue was partly a [PNP]⁺-salt, but a broad peak in the 1200 cm⁻¹ region indicated that some triphenylphosphine oxide, maximum 0.010 g, had been formed. No peaks due to NH (ND) could be detected.

Instrumental. The IR measurements were performed with a Unicam SP 200 Infrared Spectrophotometer. The measurements on the pseudohalide ions were performed in acetonitrile employing liquid IR cells with a path length of 0.1 cm. The UV measurements were performed with a Varian-Techtron UV-VIS Spectrophotometer, Model 635, employing matched 1 cm quartz cells.

The melting points were determined by a Gallenkamp melting point apparatus. The melting points are corrected.

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