

# The Superconductor System of 2H-Tantalum Disulfide Intercalated with Polyglycine

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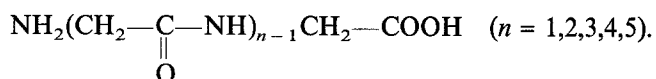
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**Abstract.** It was hypothesized that the formation of polypeptide bonds inside a lamellar superconducting compound such as 2H-TaS<sub>2</sub> could enhance the superconducting transition temperature. The preparation of tantalum disulfide intercalated with polyglycine is described. The intercalated systems were characterized by powder X-ray diffraction and thermogravimetric analysis. Superconducting transition temperatures were also measured. The results show that the system 2H-TaS<sub>2</sub>/polyglycine does not have a more favourable  $T_c$  value than 2H-TaS<sub>2</sub>/glycine.

**Key words:** Intercalation, 2H-TaS<sub>2</sub>, superconductor, polyglycine.

## 1. Introduction

Many superconducting intercalation compounds have been prepared during the last 15 years. In the present research, lamellar tantalum disulfide in its polytropic form 2H was selected for intercalation with polyglycine. Previous work reported by Chapela [1] shows that peptide intercalation is possible. The only sequence of polypeptides readily available is given by the general formula



Glycine ( $n = 1$ ) up to pentaglycine ( $n = 5$ ) are represented. The superconducting transition temperatures of the  $n$ -glycine polypeptides vary in the following order: glycine 3.4, diglycine 2.6 and triglycine 4.2 K. The issue then arises as to what happens when  $n$  becomes larger into the polyglycine range? The research reported here is intended to answer this question.

2H-tantalum disulfide is a superconductor at 0.8 K and has previously been intercalated with several organic compounds [2]. Polyglycine was obtained by Oró [3,4] starting from glycine, glycinamide or glycinamide hydrochloride (see Figure 1). Polyglycine does not have the required properties to allow direct intercalation, because it is almost insoluble and has no melting point. This makes intercalation by traditional methods impossible [5]. The formation of the system was achieved by first intercalating the monomer and then inducing the polymerization *in situ*.

## 2. Experimental

Polyglycine was first prepared starting from glycine, glycinamide and glycinamide hydrochloride, in order to determine the optimal conditions. Polymerization was confirmed by

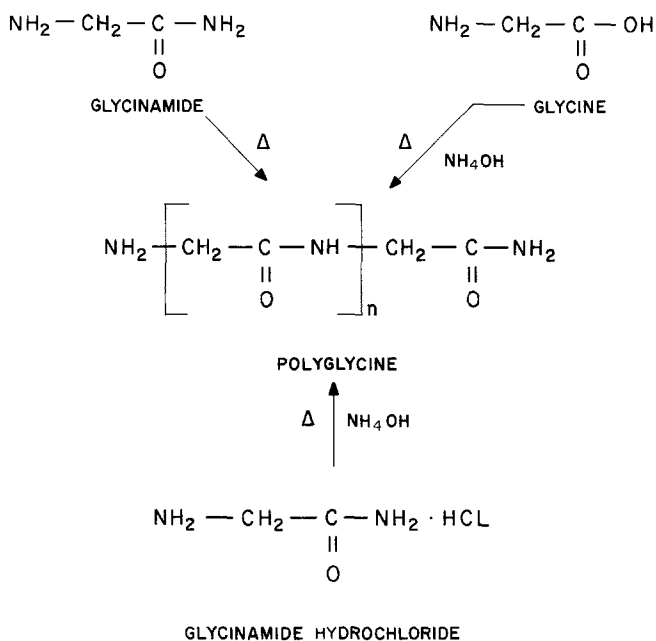


Fig. 1. Polyglycine obtention reactions.

infrared spectroscopy. The intercalation reactions of the compounds were performed by refluxing 2H-TaS<sub>2</sub> powder in solutions of differing concentration during one week. Water was used as solvent for the glycine intercalation at an intercalation temperature of 80°C [1]. Glycinamide was first prepared starting from glycine ethyl ester hydrochloride and neutralized with triethylamine [6]. Glycinamide hydrochloride was intercalated with anhydrous methanolic ammonia solution at 40°C. Polymerization of the intercalated systems was induced *in situ* by heat treatment in the presence or absence of ammonia, as necessary.

Powder X-ray diffractometry was used to show that the intercalation had occurred and the expanded van der Waals gap (defined as the difference between the separation of the TaS<sub>2</sub> layers in the intercalated complex and the original TaS<sub>2</sub> separation, and denoted  $\Delta$ ) was calculated.

Thermogravimetric analysis (TGA) was used to determine the weight loss of intercalated samples in the temperature range of 25 to 400°C. The complex composition (mole ratio between TaS<sub>2</sub> and the guest molecule) can also be evaluated and is referred to by the general formula TaS<sub>2</sub>(Intercalate)<sub>x</sub> where *x* is the mole ratio of the intercalated molecule (guest) to TaS<sub>2</sub> unit (host). At the same time, thermogravimetric analysis establishes the thermal stability of the complex.

The superconducting transition temperature of all the samples was measured using the change of magnetic susceptibility when the intercalated material becomes superconducting. The range explored in the present work was between 1.5 and 15 K. The superconducting transition temperature (*T<sub>c</sub>*) reported was measured when the magnetic permeability begins to change. The transition width ( $\delta$ ) was the temperature interval over which the total powder sample becomes superconducting.

Preliminary experiments with glycineamide and the glycineamide intercalation compound, as well as the glycineamide hydrochloride intercalation compound, carried out with a differential scanning calorimeter show that polymerization may have been induced inside the layers of the 2H-TaS<sub>2</sub>.

### 3. Results and Discussion

Table I summarizes the results of the 2H-TaS<sub>2</sub> intercalated compounds with glycine, glycineamide, and glycineamide hydrochloride. Powder X-ray diffraction patterns showed that intercalation had occurred, and they were also used to deduce the Ta-Ta interatomic distance along with the axis corresponding with the 00 $l$  reflections, and to calculate the expanded van der Waals gap ( $\Delta$ ). The results of the thermogravimetric analyses are given in percentage of weight lost by the samples, and in the change of  $x$ . The  $T_c$  values show that the highest temperature is obtained with the glycineamide hydrochloride intercalate (4.1 K) and also corresponds to the maximum concentration of guest (0.83 moles).

Table I. Results of the intercalation of glycine, glycineamide, and glycineamide hydrochloride in 2H-TaS<sub>2</sub>

Intercalate	X-ray		TGA		$T_c$	
	Ta-Ta (Å)	$\Delta$ (Å)	%	$x$	K	$\delta K$
glycine	9.64	3.60	3.5	0.12	2.4	0.62
glycineamide	9.29	3.25	13.1	0.50	3.3	0.80
glycineamide HCl	9.22	3.18	20.0	0.83	4.1	0.77

The data obtained with the aid of the differential scanning calorimeter showed the same transitions for the intercalation compounds with glycineamide and glycineamide hydrochloride. At the same time the observation that glycineamide hydrochloride demonstrates divergent behavior on annealing leads us to suspect that the HCl did not intercalate and was eliminated during reflux of the methanol/ammonia solution.

Table II shows the results with the *in situ* polymerized samples. Polymerization of glycine was induced with ammonia at 80°C and also at 140°C. Glycineamide hydrochloride was assumed to be identical to the glycineamide, which is why it is not reported independently. The glycineamide polymerization has to be performed at 100°C, since it does not occur at lower temperatures. The molar ratio ( $x$ ) is not reported here because the polymer molecular weight distribution is not known. As has been previously reported [1], an expanded van der Waals

Table II. Results of polyglycine intercalation in 2H-TaS<sub>2</sub> obtained from heat treatment of glycine and glycineamide

Reactive intercalated	Polymerization temperature (°C)	X-ray		TGA	$T_c$	
		Ta-Ta (Å)	$\Delta$ (Å)	%	K	$\delta K$
glycine	80	9.18	3.14	9.0	3.2	0.90
glycine	140	9.31	3.27	10.5	2.6	0.73
glycineamide	100	9.31	3.27	13.0	2.2	0.27

gap of 3.67 Å exists for glycine, while for pentaglycine a lower value of 3.4 Å was found. In all the 2H-TaS<sub>2</sub> intercalated with polyglycine the observed value was lower than 3.4 Å. The  $T_c$  values reported here for polyglycine intercalated into 2H-TaS<sub>2</sub> varied from 2.2 to 3.2 K and all the transition temperatures were lower than that obtained for glycine. Therefore one may conclude that  $T_c$  does not increase as the number of peptide bonds increases in the same molecule.

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