

# The Structural Modification of Al-Si Alloys

## Al-Si Alařımlarının Yapısal Modifikasyonu

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### ABSTRACT

Al-Si alloys ranging in composition from 14 to 20 % by wt. of Si, were solidified at different growt conditions. Observations made by optical microscopy indicate that eutectic and primary silicon crystals can show several growt morphologies resulting from various growt processes. It was observed that the morphological transition in silicon crystals due to strontium addition is gradual rather than abrupt.

### ÖZET

% 14 - 20 Si içeren Al-Si alařımları farklı şartlarda katılařtırılmıřlardır. Ütektik ve primer silisyum kristallerinin yapısal özellikleri optik mikroskopla incelenmiř ve farklı büyüme şartlarından kaynaklanan farklı kristal yapıları gözlenmiřtir. Stronsiyum ilavesi, silisyum kristallerinde tedrici (ani olmıyan) bir yapısal dönüşüme neden olmaktadır.

### 1. — INTRODUCTION

Alloys of aluminium with 5 to 25 % by wt. of silicon find extensive commercial aplication as structural materials. Their practical viability is due to a combination of their good casting characteristics, relatively low density, high resistance to corrosion, low thermal expansion, high thermal conductivity and good mechanical properties.

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While crystallisation of silicon phase play an important role in determining the microstructure, aluminium phase appears as an isotropic matrix in eutectic. Similarly, primary  $\alpha$ -aluminium dendrites were present in a simple form in hypo-eutectic alloys. Primary silicon crystals in hyper-eutectic alloys showed quite different growth forms. Solidification studies of Hellowell and co-workers (1, 2), in particular, have led to much better understanding of this system. Morphological observations indicated that eutectic silicon crystals have mainly flake-like nature in large range of growth conditions. This corresponds to the solidification conditions encountered in most casting processes. However, directional solidification experiments carried out at very low growth velocities beyond the foundry practice, showed quite different array of silicon morphology and eutectic microstructure. On the other hand, at large growth velocities such as in chill and die castings, finer eutectic microstructure accompanied, by rod-like silicon fibres were observed.

Na modification of Al-Si alloys has been a commercial practice for many years. Na or Sr additions must be made to castings that solidified at a slower rate in order to induce the flake-fibre transitions. The fibrous structure behaves more like a composite and improves the mechanical properties of the casting. The improvement of such mechanical properties as tensile strength, ductility and hardness is resulted by the changing of microstructure. The growth of primary silicon crystal has also been affected by third metal addition to hyper-eutectic alloy and thus refinement of these crystals were observed.

Many studies have been carried out in order to describe the mechanism of modification and many hypotheses have been formulated. However, from recent reviews (3,4,5) it is evident that the role of the modifying element is not yet clearly understood. The subject of this paper is to study the morphological transition from flake to fibrous form and to discuss the modification effects on primary growth. Therefore, the effect of strontium level on the primary and eutectic silicon morphology of aluminium-silicon alloys was investigated in order to gain more information to resolve some of the existence doubts.

## 2. — EXPERIMENTAL

Hyper-eutectic Al-Si alloys containing about 14 to 20 % by wt. of Si. and minor amount of Sr. has been prepared by melting under argon gas pressure, using 99,999 % purity aluminium and 99 % purity strontium. The phase diagram of the Al-Si system is illustrated in Fig. 1.

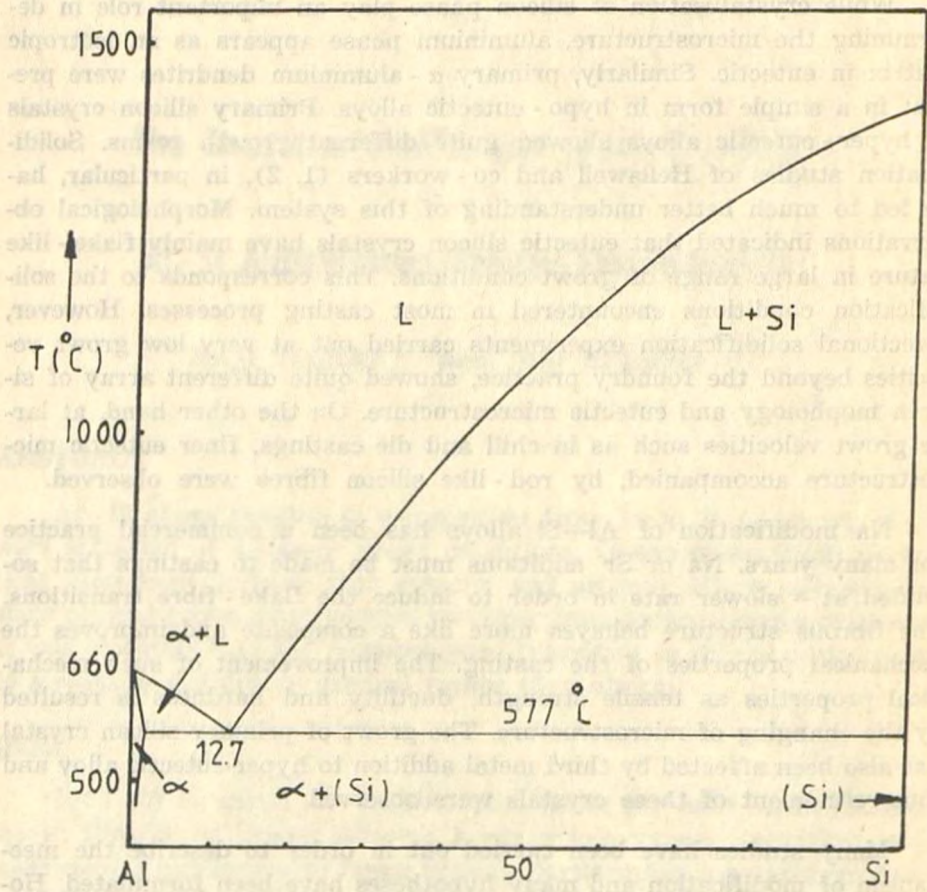


Figure. 1. — The aluminium - silicon phase diagram.

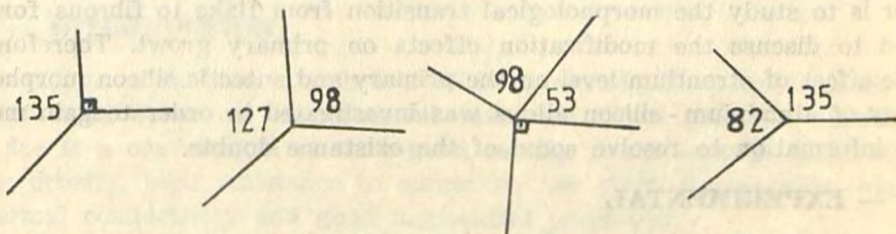


Figure. 2. — Angular relationship in plate-like silicon particles on transverse section.

Accurately weighed amounts of aluminium and silicon were melted in a high purity graphite crucible, which had been preheated to drive off volatile impurities. Melting was achieved by means of a high frequency induction heating unit and the operation was performed under flowing argon to prevent oxidation. Addition of strontium has been made to the melted Al-Si alloy after homogenisation.

Some of the melted alloys were filled in alumina tubes (I.D.=2 mm) and remelted at 850 C° in a directional solidification apparatus and then solidified under various growth velocities (6). Some of the alloys were remelted also at 750 - 850 C° in horizontal furnace and then solidified at different cooling rates (6).

### 3. — MECHANICAL PROPERTIES

An indication of improvement in mechanical properties brought about by modification in Al-Si alloys is presented below (7) :

Alloy	Condition	Tensile strength (kg/mm <sup>2</sup> )	Elongation (%)	Hardnes (HB)
Normal	sand cast	12,7	2	50
Modified	»	19,7	1.3	58
Normal	chill cast	19,7	3.6	63
Modified	»	22,5	8.0	72

Steen and Hellawell (2), also carried out some tensile tests on unmodified as well as guench modified alloys and reported U.T.S, values 10 - 20 % higher than those given by Thall and Chalmers (7). Considerable variation was found in % elongation as well and these differences were attributed to the use of pure materials.

## 4. — STRUCTURAL MODIFICATION

### 4.1. — Eutectic Modification

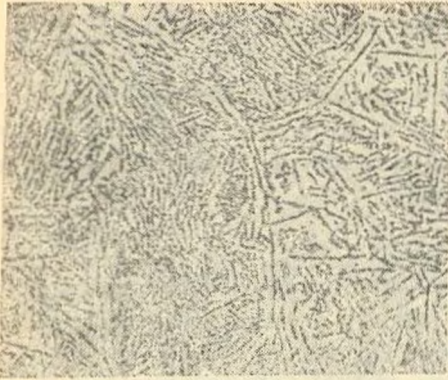
Al-Si alloys like Fe-C form anomalous structures and ordinary castings display flake-like silicon or graphite morphology respectively. Fig. 3(a) shows such an unmodified ordinary eutectic microstructure in Al-Si alloys. In this micrograph grain boundary of different eutectic particles are not discernable and there is no preferred orientation relationship between two eutectic phases. Two kind of silicon morphology

can be seen in this micrograph and in alloys solidified in sand casting. They can be discernable on transverse section as intersected plate-like separation and interconnected flake-like separation. Plate-like separation of angular silicon crystals showed long-range regularity with various number of side plates which are mutually inclined at a variety of angles (such as  $35^\circ$ ,  $53^\circ$ ,  $82^\circ$ ,  $90^\circ$ ,  $98^\circ$ , ...) on transverse section. (Figure 2). Figure 3(a) shows three side plate particles and irregular flake-like silicons in spaces in between the angular plates.

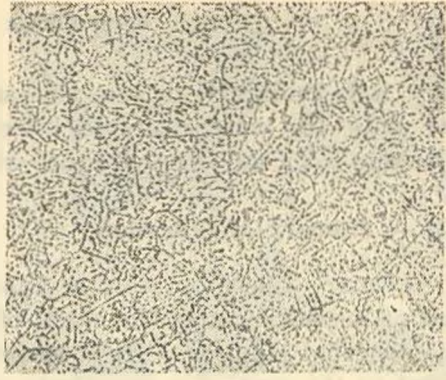
When small quantities of strontium were added to an Al-Si alloy, the microstructure did not change directly to a fibrous form. (Figure 3(d)). Figure 3(b) shows the eutectic microstructure observed in Al-17.1 % Si-0.0016 % Sr alloy. It was noted that, the microstructure consists of angular silicon crystals, occurring mainly in two perpendicular directions and generally originating from a central plate. Small dotted separations also appeared in the spaces in between the angular silicon plates. While in figure 3(a) less amount of angular silicon crystals with more flake-like inter angular separations were present, in figure 3(b) contain more angular crystals with less amount of inter angular separations. It was clearly defined that these dotted inter-angular separation is in fact intersecting of rods on transverse section (6). At higher strontium levels almost all these angular silicon plates had been changed interrelated rods. (Fig. 3(c)). Most of the rods appear in place of the side plates as shown in figure 3(b). Figure 3(d) shows the transverse section of Al-15 % Si-0,080 % Sr alloy. On this micrograph due to rather high strontium addition, there is quite regular dotted array and it is difficult to visualize interrelation between each dots.

The consistent facts are that there is an altering of the silicon microstructure in the presence of strontium and the altering of microstructure is associated with branching of silicon particles. Strontium additions change the plate-like arrangement of angular silicon to rod-like arrangement by branching frequently within the plate plane. On the other hand, flake-like growth in unmodified alloy was poisoned by strontium addition and replaced by rod-like arrangement branched in several directions.

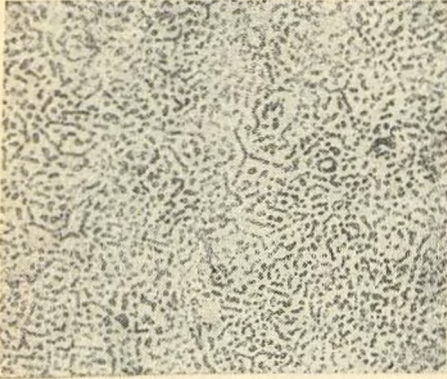
When aluminum-silicon alloys are solidified at relatively large growth velocities such as in chill and die castings, finer eutectic microstructures accompanied with rod-like silicon fibres were observed. Figure 3(e) shows two kind of microstructure in directionally solidified



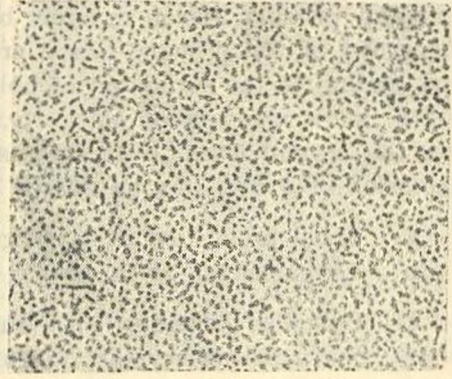
(a)



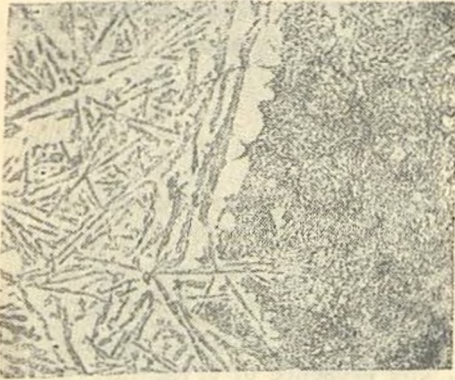
(b)



(c)



(d)



(e)

Figure 3. — Optical micrographes of directionally solidified Al-Si alloys. a) Transverse section of Al-15,2 % Si alloy, growth velocity (R): 470  $\mu\text{m}/\text{sec}$ . x772. b) Transverse section of Al-17,1 % Si-0,0016 % Sr alloy, R: 237  $\mu\text{m}/\text{sec}$ . x895. c) Transverse section of Al-14,35 % Si-0,0047 % Sr alloy, R: 79  $\mu\text{m}/\text{sec}$ . x826. d) Transverse section of Al-15 % Si-0,080 % Sr alloy, R: 95  $\mu\text{m}/\text{sec}$ . x901. e) Longitudinal section of Al-14 % Si alloy, R: 5  $\mu\text{m}/\text{sec}$ . x130. (Right side is quenched reg.)

alloy. Left hand side corresponds sand casting and shows coarse flake and plate - like microstructure while right hand side of picture shows relatively finer rod - like structure which corresponds to the chill castings.

#### 4.2. — Primary Modification

Primary silicon crystals in hyper - eutectic alloys show quite different of microstructure. The shape of primary crystals are mainly coarse interconnected plate or polyhedral form (Fig. 4(a)). When strontium is added to the melt, it is selectively adsorbed on easy growth sites of primary silicon crystals. They block the advance of the preferred growth sites and primary silicon crystals become more equiaxed. (Fig. 4(b)). The growth of the primary silicon crystal was stopped locally by strontium leading to the formation of holes. Primary silicon crystals in figure 4(c) show many holes. Similarly, increasing adsorption of strontium over the crystal leads to an increasing blocking of growth. The overall succession of growth may be related to adsorbed impurity as follows :

- a) Local prevention of layer growth leads to formation of holes
- b) Extension of holes results in branched crystals and dendrites
- c) Complete coverage with strontium leads to dispersion of primary silicon crystals (Figure 4(d)).

Untreated and strontium treated Al-20 % Si alloys were cooled down side by side quite rapidly. By comparison of figure 4(e) and (f) it can be seen that addition of strontium always makes primary crystals more round and equiaxed.

#### 5. — DISCUSSION

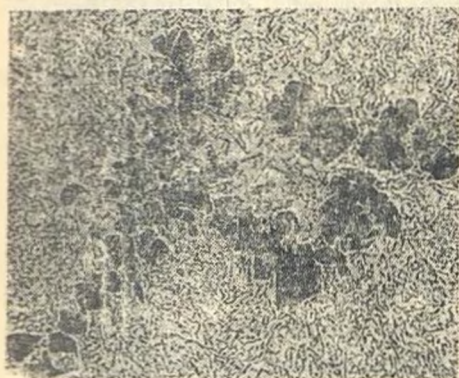
It is well established fact that flake - like silicon crystals in unmodified alloy grow by twin plane re - entrant edge (TPRE) mechanism (1). Multiple twin planes in these crystals supply easy growth sites and extended growth in certain directions can be observed. Twin planes in flake - like silicon crystals lie in (111) plane and they are formed either during nucleation or during growth. The presence of crystallographically different twins, such as (210) was also reported (1, 8) in silicon crystals. Plate - like angular silicon crystals may contain this sort of twinning as observed by Lemaignan and Malmejac (8). These authors suggested that angular silicon crystals grow by the aid of this twin planes. Ho-



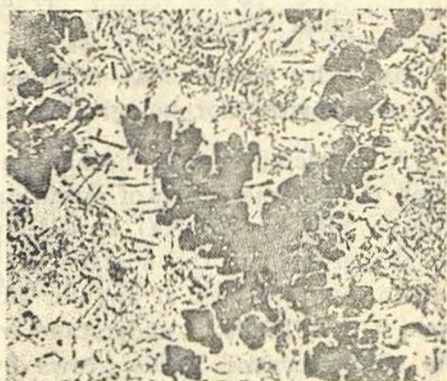
(a)



(b)



(c)



(d)



(e)



(f)

**Figure. 4.** — Primary silicon crystals observed in hyper - eutectic alloys. Alloys were cooled down from 750 - 850°C in graphite boats. a) Al-20 % Si alloy, x77. b) Al-20 % Si-0,01 % Sr alloy, x92. c) Al-20 % Si-0,02 % Sr alloy, x180. d) Al-20 % Si-0,03 % Sr alloy, x127. e) Al-20 % Si alloy, solidified under increased cooling rate, in quench, x108. f) Al-20 % Si-0,2 % Sr alloy, solidified under increased cooling rate, in quench, x108.



wever, the screw dislocations may be contributing growth sources for the growth of angular silicon crystals as suggested previously by Day and Hellawell (1). Quite recently (6, 9) it was clearly put forward that, the presence of twin planes in silicon crystal does not necessarily mean that growth mechanism is TPRES. Therefore, it was regarded that screw dislocations more likely contribute towards the growth of angular silicon crystals even though they contain (210) twins. Following points became unresolved if TPRES growth mechanism is accepted for angular silicon crystals :

- a) How angular silicon side plates grow? Each side plates of angular silicon crystals don't have twin planes (6, 11).
- b) Why Sr (or Na) addition did not affect the angular silicon growth morphologies?

A restricted growth mechanism has been proposed by Day and Hellawell (1) for the action of strontium (or sodium) in Al—Si alloys. They propose that Sr (or Na) is selectively adsorbed in the twin plane re-entrant groove on the surface of the growing silicon and this should lead to modifications. If angular silicon crystals grow by TPRES mechanism, it is difficult to understand why it is not modified whereas flake-like silicon crystal which grows by TPRES mechanism is modified by strontium addition (Figure 3(b)).

Addition of strontium firstly suppress the flake-like growth by poisoning easy growth sites. It can even clearly be seen on the micrograph with alternative plate-like silicon growth morphology replace the majority of flake-like silicon crystals. Secondary affect of strontium is the poisoning of these plate-like silicon crystals. Eventually, whole sequence induces fibrous silicon crystals (Fig. 3).

Alteration of microstructure is associated with a different growth mechanism. When strontium is added to the alloy TPRES growth mechanism can not operate at all. Alternative growth mechanism which initially promotes the formation of plate-like silicon and ultimately changing into rod-like silicon forms has been observed. Similar transition is observed at large cooling rate even strontium is not added. Under this condition aluminium shows, same poisoning affect as strontium does at slow cooling rate.

The results reported here show that the modification of

primary silicon crystals appear as a result of the strontium accumulation on the growing faces. Growing of the primary silicon crystals take place either by the aid of twinning or as a result of the initiation of each new growth layer by two dimensional growth mechanism (9). Strontium poisons the twin grooves and immobilize the growing crystals and increase the driving force for growth. Under this condition round or dendritic primary silicon crystals grow with several holes. Local variation in adsorbed strontium density will be sufficient to block the growing steps, and part of the surface will be retarded for growth. The remainder of the crystal will grow around these areas which will cease to develop because they will fail to compete for solute. The formation of holes in primary silicon crystals observed with strontium is apparently related to this phenomena. It was reported (10) that graphite growth in lanthanum treated iron and nickel alloys showed the same sequence. Lanthanum atoms are adsorbed on the growing faces of the crystal and block the advance of the growth steps. Limited adsorption blocks the growth locally leading to hole formation. Increasing adsorption on graphite leads to spherulitic growth.

## 6. — CONCLUSIONS

The following conclusions may be drawn from the present work :

- 1 — Eutectic microstructures of Al-Si alloys are modified by minor addition of strontium. The change of microstructure upon successive additions of strontium is the same as the change of growth condition. Therefore, it is suggested that angular silicon crystals are a profibrous form in modified alloy. It is noticed that quench modification may also show same sequence.
- 2 — Modification of primary silicon crystals can be explained in terms of a restricted growth mechanism. Adsorbed strontium atoms lead to non-uniform growth and thus primary silicon crystals show in succession, holes, by the extension of these holes lead to branched crystals and complete coverage leads to dispersion of primary silicon crystals.

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## APPENDIX

The dimensions of fibrous silicon particles in rapidly frozen or modified specimens lie the limit of resolution of the optical microscope. In order to obtain better resolution the scanning electron microscope (SEM) was employed. The SEM can produce micrograph images directly from solid specimens with a resolution and depth of focus considerable better than that of the optical microscope. Its large depth of focus allowed the examination of silicon morphology in three dimensions after deep etching. Transverse and longitudinal sections of unidirectionally solidified specimens were ground and polished as for the optical microscopy, and deeply etched in a solution of 1-2 % HCl. If microstructure was very fine, etching was started with 1 % HCl and the HCl concentration was increased daily up to 2 % HCl. The period of etching was generally 15 days. HCl removes the aluminium matrix from the surface regions and exposes the dark coloured silicon particles. Polished and etched sections dried and coated with a thin layer of gold to improve contrast and then mounted on stubs using a conducting glue (Silver dag). The stub was placed directly into the specimen stage of SEM for examination.

Two micrographes taken from the SEM are given below: While first micrograph shows silicon flakes, second one shows silicon fibres (Rod-like silicon morphologies). These corresponds to the optical micrographs of Figures 3(a) and (d) respectively.

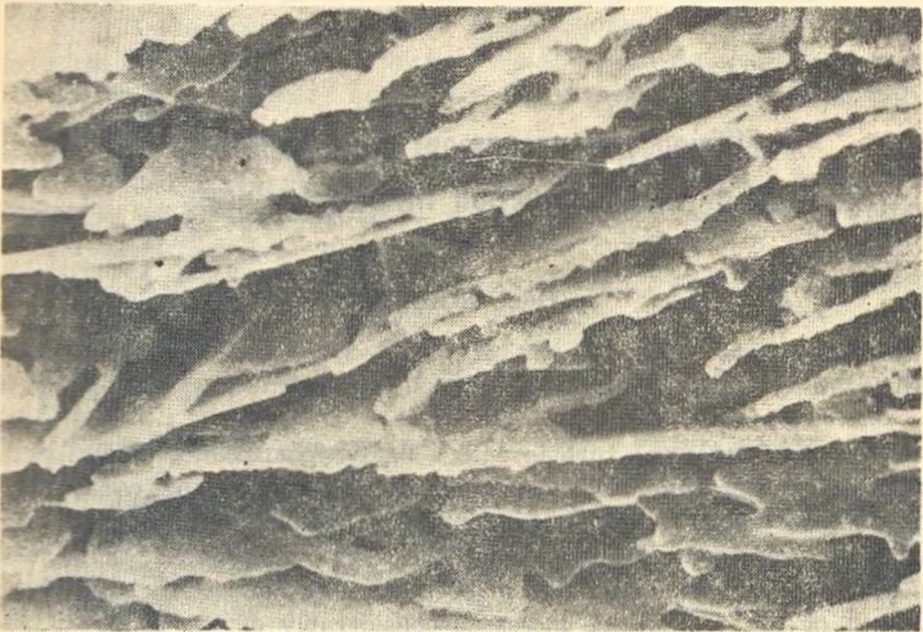


Figure. 1. — Scanning electron micrograph of normal Al-Si eutectic.

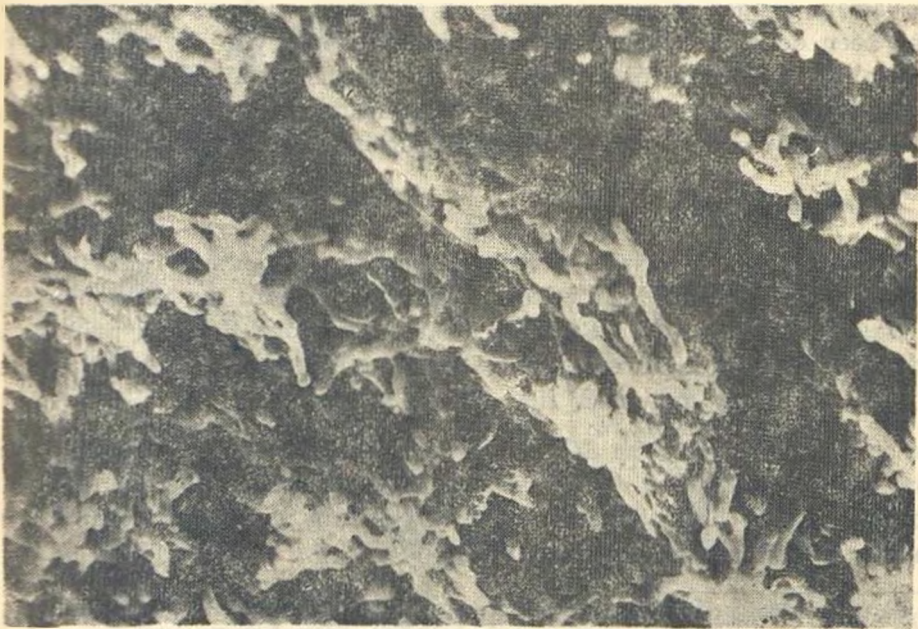


Figure. 2. — Scanning electron micrograph of modified Al-Si eutectic.