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Institut für Material- und Festkörperforschung
Projekt Schneller Brüter

## Mechanical Properties of Cladding Materials after Annealing with Carbide Fuels

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# Mechanical Properties of Cladding Materials after Annealing with Carbide Fuels[+)]

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[+)] presented at the Fuel and Materials Specialist Meeting of the Coordinating Group on Gas cooled fast Reactor Development, Würenlingen, October 24/25, 1973

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# Mechanical Properties of Cladding Materials after Annealing with Carbide Fuels

## Abstract

Carburization of the cladding material is the only critical chemical interaction between fuel and cladding that is to be expected in a carbide pin. Carburization greatly affects the mechanical properties of the cladding material.

The mechanical properties of the steels (elongation and tensile strength) are most affected when annealed with carbide fuels at 600°C. At 500°C, carburization is still low. At higher temperatures (above 700°C) a resolution of carbide precipitates takes place. Fuel composition and bonding medium have a marked influence on the mechanical properties.

Room temperature elongation values for steel as well as for vanadium base alloy specimens annealed at 600°C are very low. With sodium bonding they drop to practically zero. However, elongation values at higher temperatures are still satisfying. Hardness measurements, mechanical properties and carbon contents of the cladding materials after annealing can well be related.

# Mechanische Eigenschaften von Hüllmaterialien nach Glühung mit carbidischen Brennstoffen

## Kurzfassung

Die Aufkohlung des Hüllmaterials ist die einzige kritische chemische Wechselwirkung zwischen Brennstoff und Hülle, die in einem Karbidbrennstab zu erwarten ist. Sie beeinträchtigt in starkem Maße die mechanischen Eigenschaften der Hülle.

Die mechanischen Eigenschaften der Stähle (Dehnung und Zugfestigkeit) werden am stärksten beeinflußt, wenn die Glühung mit karbidischem Brennstoff bei 600°C durchgeführt wird. Bei 500°C ist die Aufkohlung noch gering. Bei höheren Temperaturen (über 700°C) findet eine Auflösung oder Vergröberung der Carbidausscheidungen statt. Die Brennstoffzusammensetzung und das Bindemittel machen sich deutlich bemerkbar.

Die Raumtemperatur-Dehnwerte der Stahl- wie auch der Vanadinlegierungsproben, die bei 600°C geglüht wurden, sind sehr niedrig. Bei Natriumbindung fallen sie auf praktisch null ab. Die Dehnwerte bei höheren Temperaturen sind jedoch noch zufriedenstellend. Härtemessungen, mechanische Eigenschaften und die Kohlenstoffgehalte der Hüllmaterialien können in Einklang gebracht werden.

## 1. Introduction

Chemical incompatibility between fuel and cladding is one of the problems connected with the use of uranium-plutonium oxide as a fast breeder nuclear fuel. Fission products play a major role in the cladding attack of an oxide fuel pin. In a carbide fuel pin the situation is somewhat different. Fission products do not take part to a noticeable extent in the reactions between fuel and cladding in a carbide pin. This could be clearly demonstrated in out-of-pile annealing tests. Fig. 1 shows the difference in cladding attack of a so-called simulated irradiated oxide fuel pin and a similarly treated carbide fuel pin. The simulation was performed by adding fission product elements and oxygen to the oxide fuel and likewise to the carbide fuel in amounts that correspond to a burn-up of 1o at.%. Heavy attack on the cladding material could be observed in the oxide specimen whereas the fuel/cladding chemical interactions in the carbide specimen have been limited to those occuring between the cladding material and an overstoichiometric carbide fuel without the addition of fission product elements. The results of the tests with simulated irradiated fuel,which were published in other reports [1-3] ,clearly demonstrated that carburization of the cladding material is the only critical chemical interaction between fuel and cladding to be expected in a carbide pin.

Carburization greatly affects the mechanical properties of the cladding materials. Measuring the change in mechanical properties is both a sensitive method to detect chemical interactions between fuel and cladding, and to demonstrate the effect of those chemical interactions on the operational behaviour of the cladding materials.

## 2. Experimental Procedure

In our experiments we used as cladding materials several types of austenitic steels such as the niobium stabilized 1.4988 type steel, which is favoured in Germany as cladding material for a liquid metal cooled fast breeder reactor, the AISI 316 and 3o4 type steels, and also vanadium base alloys. As nuclear fuels we used uranium carbides with various carbon contents and uranium carbonitride. Both gas bonding and liquid metal bonding was

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employed in the tests. Annealing temperatures ranged from 5oo to 8oo°C, annealing periods from 144 to 1ooo h. Strip specimens of the cladding materials were embedded in the fuel and enclosed in steel capsules. The steel capsules again were encapsulated in silica capsules which were heated in muffle ovens.

The preparational procedure was carried out under a pure argon atmosphere. After annealing,tensile testing was done at room temperature and at annealing temperature. Specimens that were annealed at 8oo°C were tested at room temperatures and at 7oo°C.

Compositions of the cladding materials and fuels used in these experiments are given in Tables 1 and 2.

## 3. Results

Most of the effect that carburization of the cladding materials has on the mechanical properties was reached already after a relative short period of time ($\sim$ 2oo-5oo h) as demonstrated by Fig. 2 and 3. Fig. 2 shows the fracture elongation at room temperature of 1.4988 type steel after annealing at 6oo, 7oo and 8oo°C in static argon, overstoichiometric UC and U(C,N) for periods up to 1ooo h. Fig. 3 shows the results for 316 type steel.

The greatest effect on the ductility is being realized by annealing at 6oo°C. At higher temperatures obviously a resolution of carbide precipitates in the steels takes place. At 5oo°C the reaction rates are probably too low to cause much carburization as can been seen from Figs. 4, 5 and 6. There is practically no decrease in ductility after annealing at 5oo°C for 1ooo h compared with the extrapolated values for annealing in argon and also for the as received condition. (All the values in the following figures are for annealing periods of 1ooo h unless stated otherwise). After annealing at 6oo°C the decrease in elongation is greater for the 316 steel than for the 1.4988 steel, even though similar values are obtained for both steels. Sodium bonding has an effect on cladding carburization only at higher temperatures.The mechanical properties at annealing temperatures are shown in

Fig. 7 and 8. Most apparent again is the decrease in ductility for 600°C annealing and test temperature for the 316 steel if compared with the 1.4988 steel. However, for both steels enough ductility is still reserved at the apparently most critical temperature condition at 600°C. Generally, the rupture and yield strength values have increased after annealing in contact with the fuel. One should take notice of the rather strong increases in the yield strength values which should have some influence on the creep properties. This effect is looked upon as rather beneficial for the operational behaviour of the cladding material.

The influence of the carbon content of the fuel on the change in mechanical properties is shown in Fig. 9 and 10 for a steel of type 1.4988 annealed at 600°C for 1000 h. There is a steady decrease of cladding ductility with increasing carbon content of the fuel in contact with the cladding. This decrease in ductility is even more pronounced when sodium bonding is used.

Fig. 11 and 12 demonstrate the effect of temperature and type of fuel on the carbon uptake and weight increase of a stainless steel cladding. Carbon uptake increases with temperature and carbon content of the fuel. The influence of sodium bonding seems to play a role at higher temperatures and at high carbon contents of the fuel.

Microhardness measurements (Fig. 13 and 14) also revealed the greatest effect of the carburization at 600°C. At 500°C the penetration zones of carbon into the cladding seem to be smaller than 100 µm. At 700°C hardness values decrease again.

For vanadium base alloys, 600°C seems to be the critical temperature, too. However, we do not have many results on the behaviour of vanadium alloys. Due to a shortage in sheet material, we have carried through tests with vanadium base alloys only at 600 and 800°C. 800°C was looked upon as an interesting temperature since vanadium base alloys have a potential as cladding materials only when high coolant temperatures are to be aspired. Hence, statements upon the behaviour on vanadium alloys are more or less preliminary. Fig. 15 shows the room temperature mechanical properties of two vanadium alloys annealed at 6oo and 8oo°C for 1ooo h. After annealing at 6oo°C there is practically no ductility left. The tensile strength values are still high, however, so no total embrittlement was caused by the carbon uptake. After 8oo°C, ductility is rather high for VTi3Si1 but very low for VTi2Cr15. High temperature ductility, however, is still good. In Fig. 17 and 18 the effect of bonding medium and carbon content of the fuel on the room and high temperature properties of the alloy VTi3Si1 is demonstrated. With gas bonding an increase in carbon content of the fuel has only a slight effect on the ductility of the cladding when annealed at 8oo°C. With sodium bonding an effect can be seen.

In Fig. 19 and 2o, finally, a comparison is given on the behaviour of the different cladding materials when annealed in contact with carbide fuel at 6oo°C. Fig. 19 shows the carbon uptake by the cladding. Steel of type 1.4988 encounters the least increase in carbon content among the types of stainless steels tested. The carbon uptake by VTi3Si1 is not much higher. Also the decrease of ductility is the least for type 1.4988 steel as shown in Fig. 2o. Ductility values after annealing in contact with carbide fuel at 6oo°C are similar, however. A very low value is measured for VTi3Si1.

## 4. Conclusion

6oo°C seems to be the most critical temperature for the cladding of a carbide fuel pin. Fracture elongation drops to very low values especially in connection with sodium bonding. However, tensile and yield strength are very high, so no total embrittlement has occured. Hardened zones in the cladding reach 1oo to 2oo µm after annealing for 1ooo h depending on temperature. Most of the effect on the mechanical properties due to chemical interaction with carbide fuels is achieved already after a relative short period of time of approximately 2oo h. The vanadium base alloys demonstrate a satisfying behaviour.

## Literature

[1] P. Hofmann, O. Götzmann, KFK 1718, 1973

[2] P. Hofmann, KFK 1832, 1973

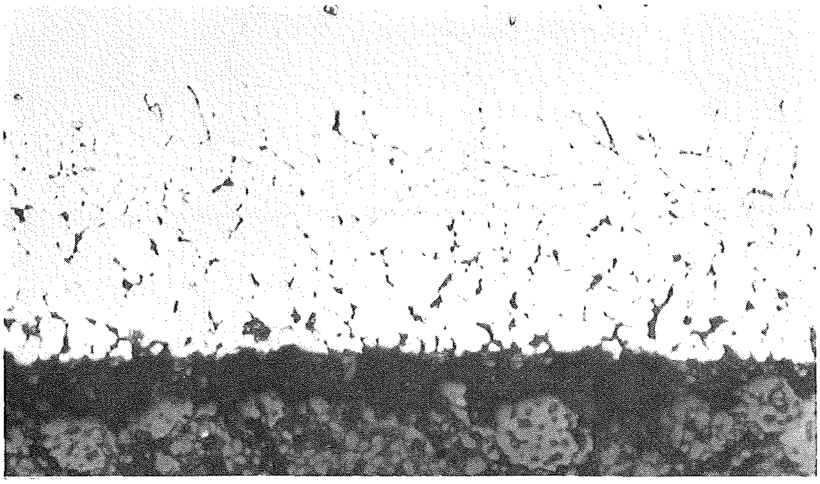
[3] O. Götzmann, P. Hofmann, paper presented at the Fuel and Fuel Elements Meeting of the American Nuclear Society, Brussels, 1973 (IAEA-SM-173/28)

Tab. 1: Composition of cladding materials (wt.%)

| | C | Si | Mn | P | S | Cr | Ni | Co | Mo | V | Ti | Al | Fe | Ta+Nb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1.4988 | 0,08 | 0,27 | 1,1 | 0,019 | 0,009 | 16,1 | 13,6 | 0,055 | 1,28 | 0,7 | - | - | bal. | 0,87 |
| 316 | 0,045 | 0,45 | 1,7 | 0,23 | 0,017 | 17,0 | 13,25 | 0,14 | 8,2 | - | 0,05 | 0,01 | bal. | 0,05 |
| 304 | 0,048 | 0,55 | 1,54 | 0,035 | 0,02 | 17,7 | 4,5 | 6,17 | 0,2 | - | 0,05 | 0,03 | bal. | |
| VTi3Si | 0,023 | 0,81 | | | | | | | | bal. | 2,75 | | | |
| VTi2Cr15 | | - | | | | 15 | | | | bal. | 2 | | | |

Tab. 2: Composition of fuels (wt.%)

| | O | N | C (total) | C (free) | aquiv C |
|---|---|---|---|---|---|
| UC (1) | o,o37 | o,o38 | 4,76 | | 4,82 |
| UC (2) | o,17o | o,o5o | 4,76 | o,o34 | 4,96 |
| UC (3) | o,12o | o,o23 | 4,99 | o,o53 | 5,15 |
| U(C,N) | o,22 | 2,43 | 2,65 | o,o5 | 4,9 |



$UO_{2.o8}$+simulated fission products 2o µm

$UC_{1.o5}$+simulated fission products 2o µm

$UC_{1.o5}$ 2o µm

Fig. 1: Attack on cladding of type 1.4988 steel by fuel and simulated fission products (corresp. to burn-up of lo at.%) sim. fission products: Cs,J,Te,Se, Ba,Zr,Ce,Nd,Mo,Ru,Rh,Pd.

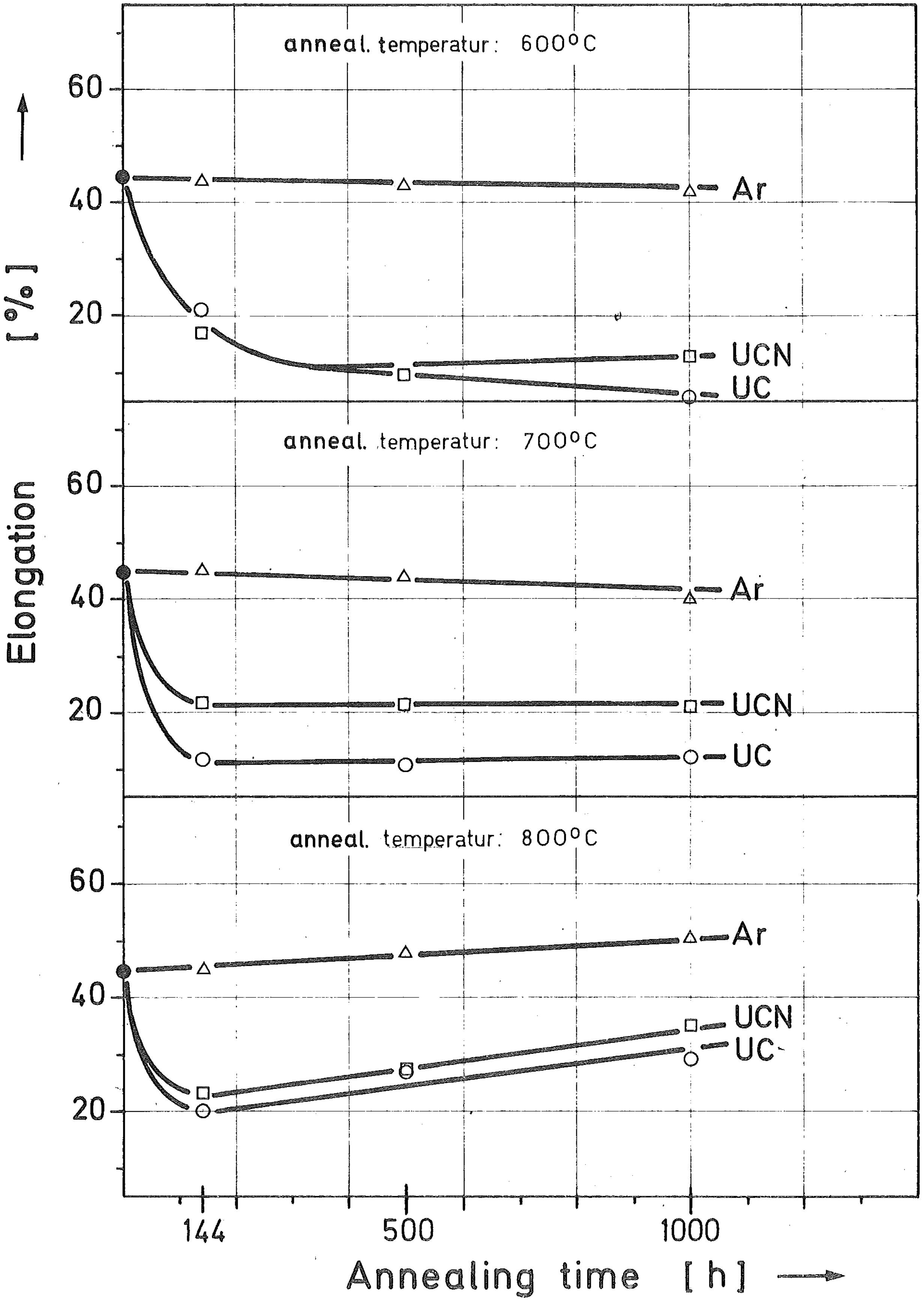


Fig. 2 Elongation of steel 1.4988 at 20°C annealed in Ar, UC, UCN, resp.

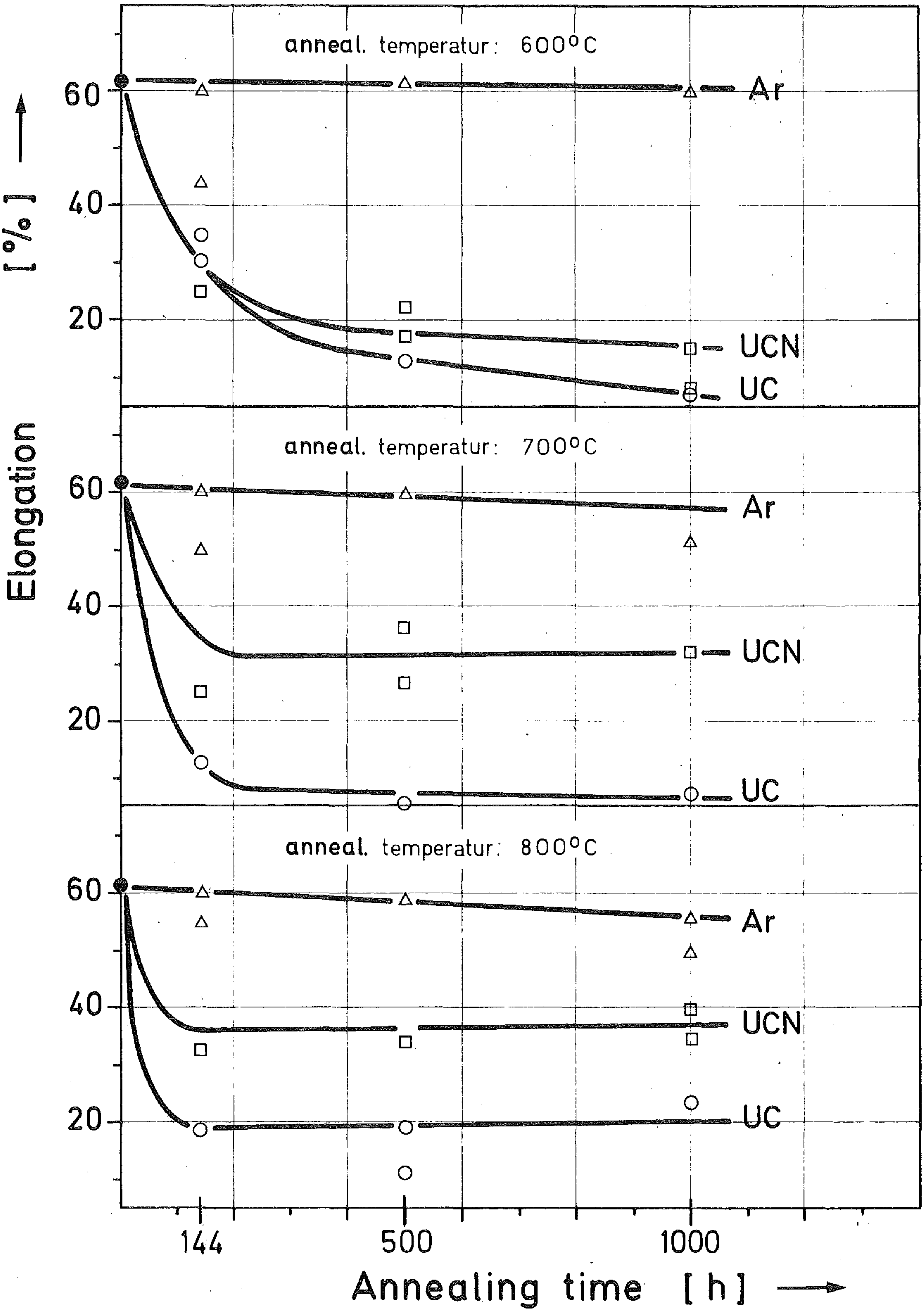
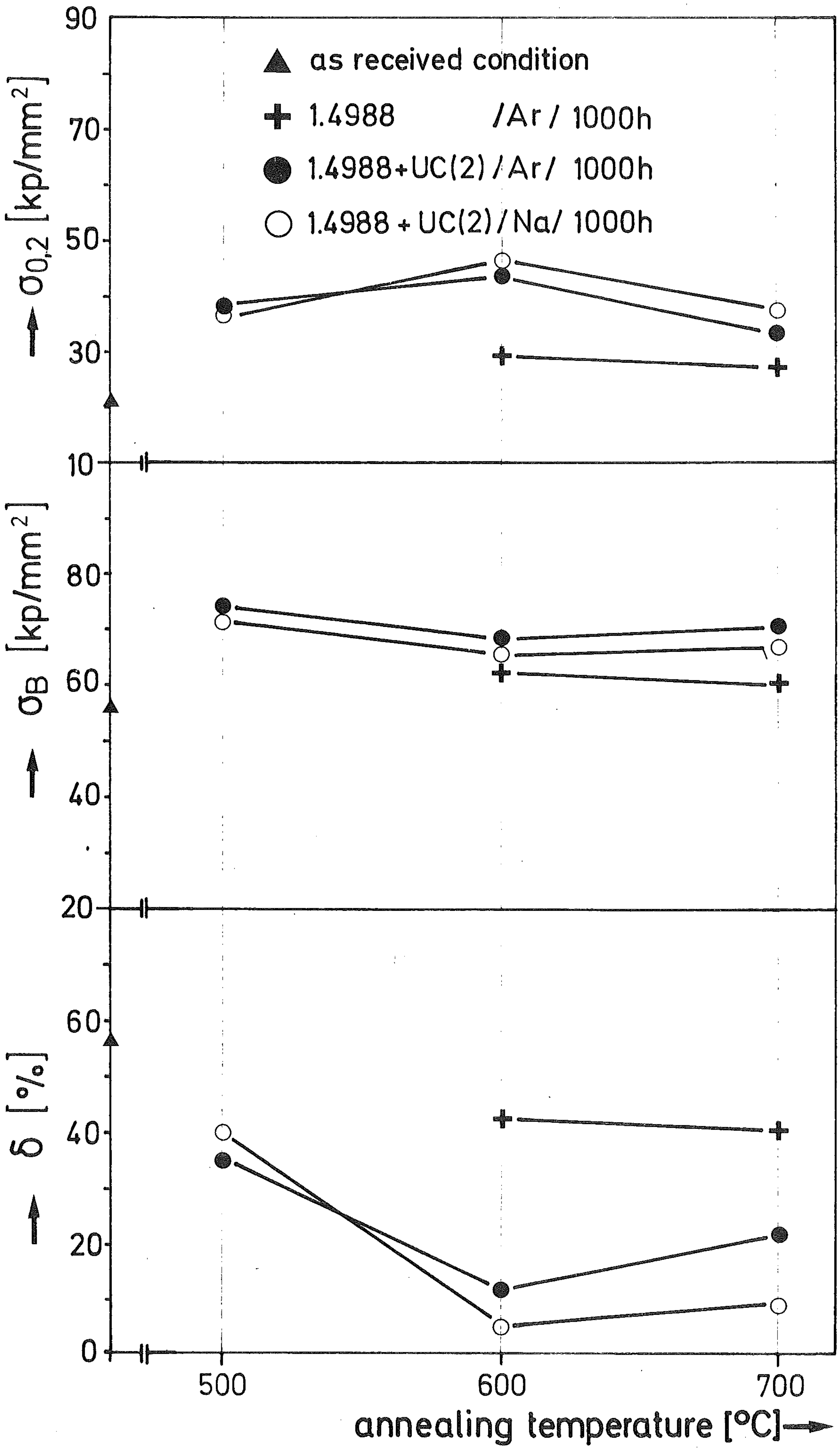
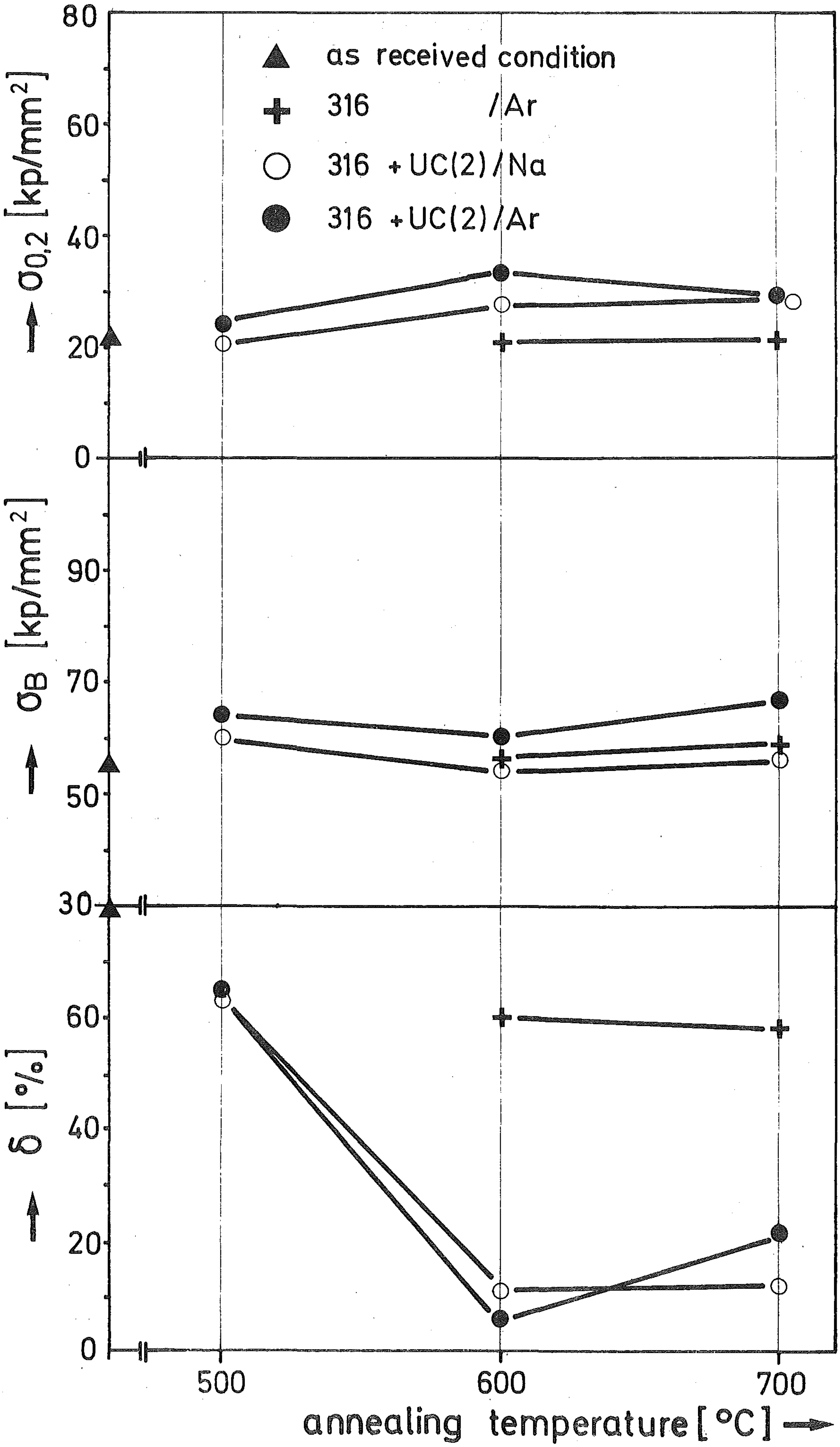


Fig. 3 Elongation of 316ss at 20°C annealed in Ar, UC, UCN, resp.



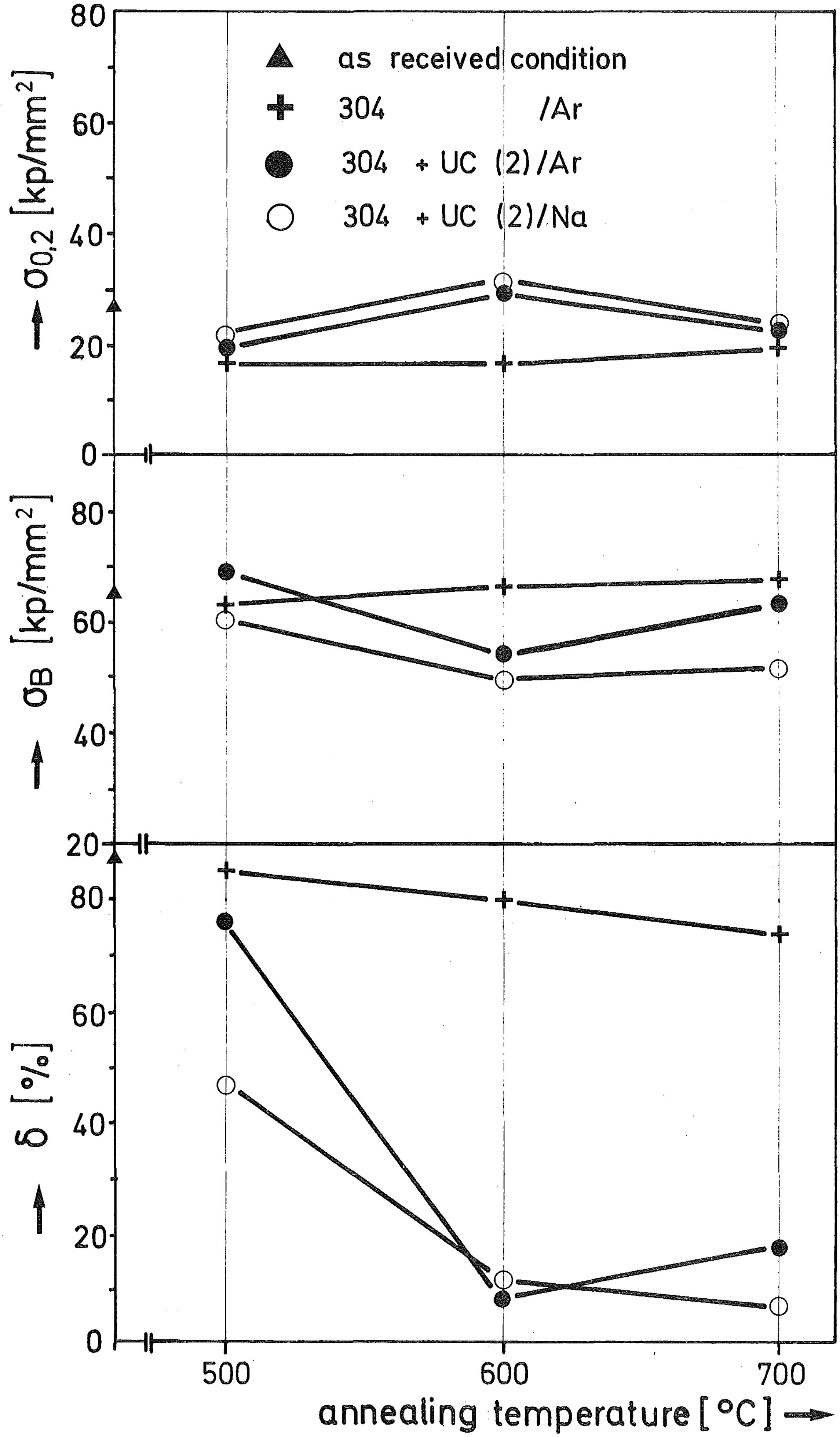
room temperature properties

Fig. 4



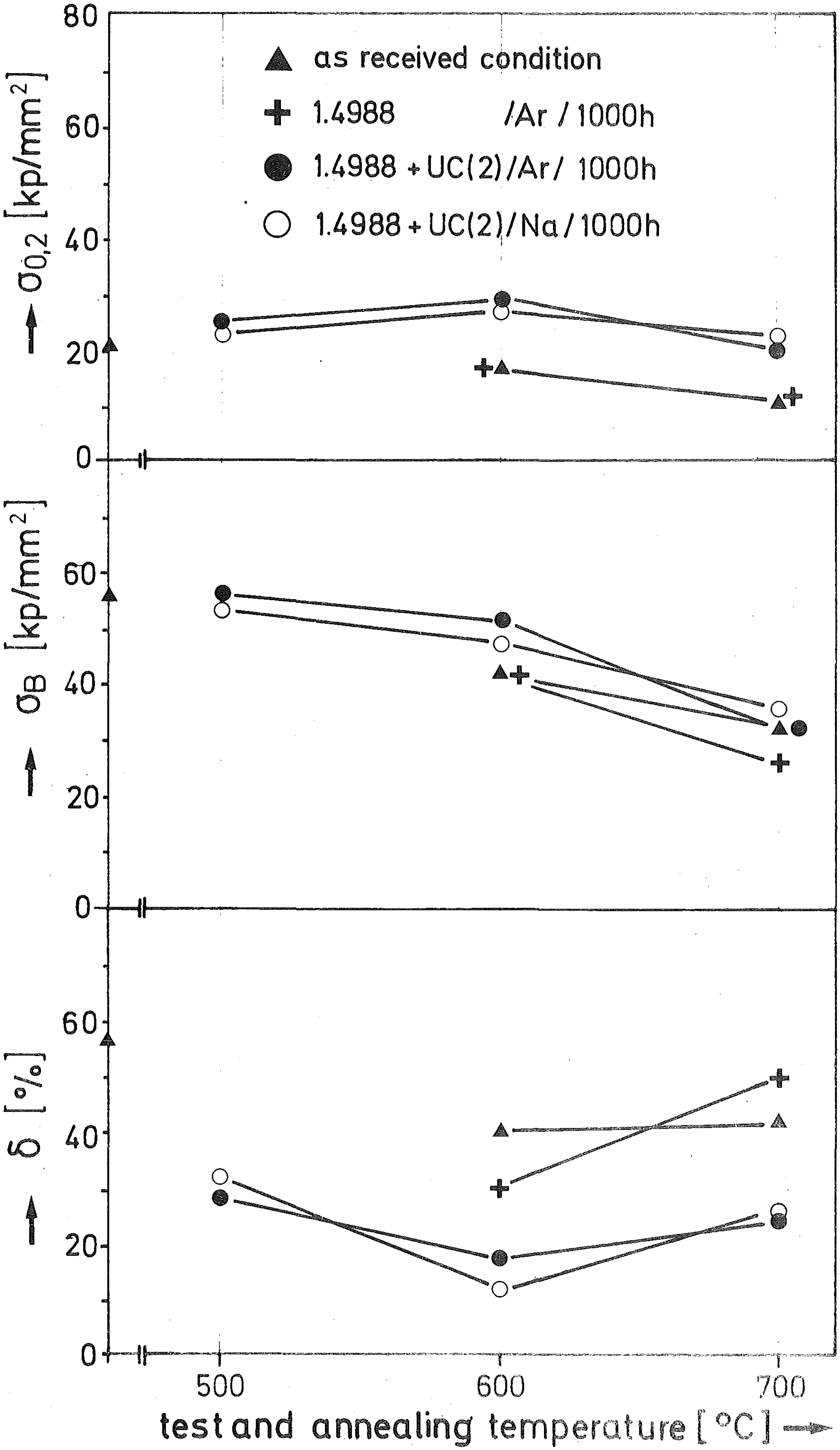
room temperature properties

Fig. 5



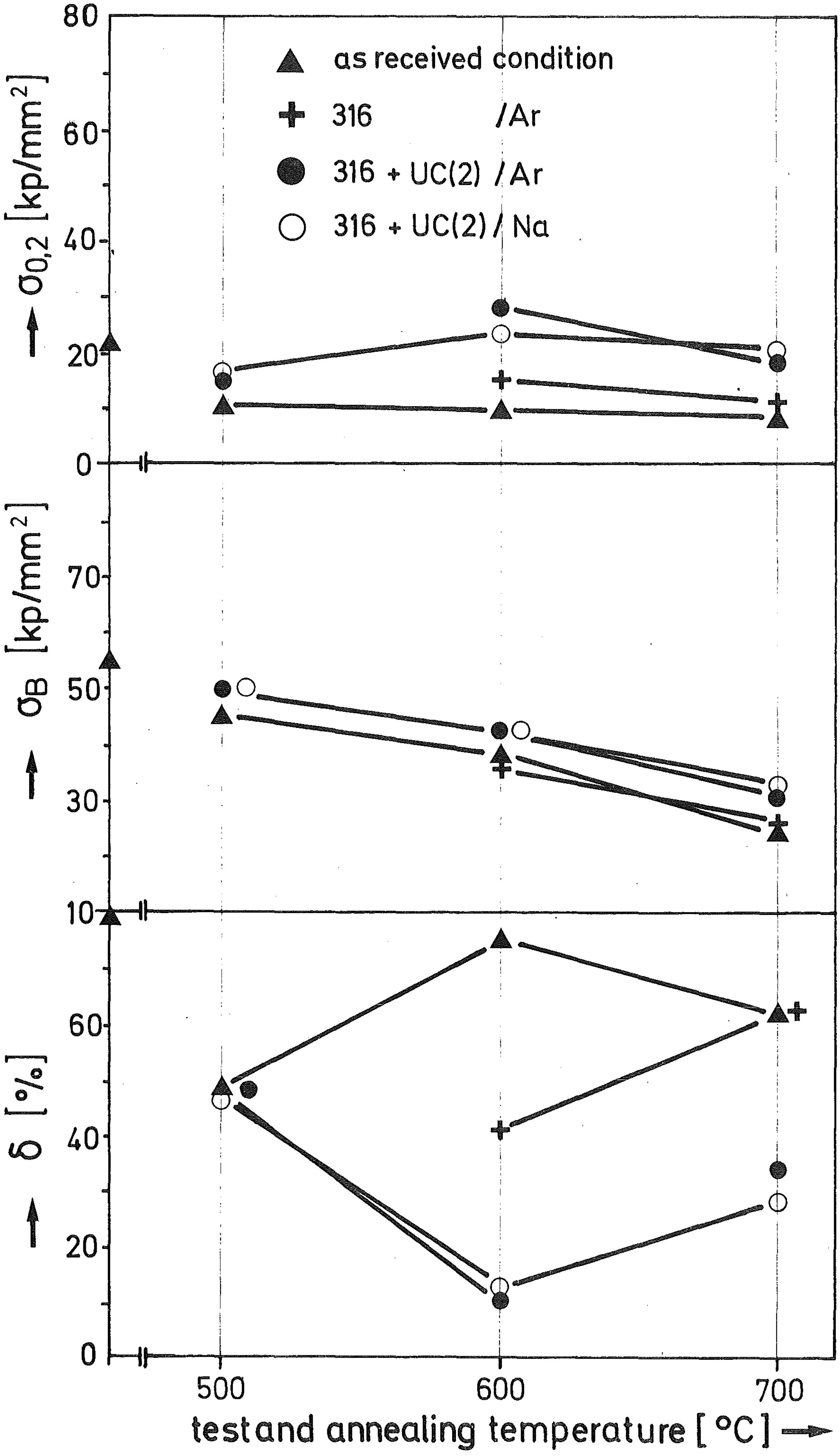
room temperature properties

Fig. 6



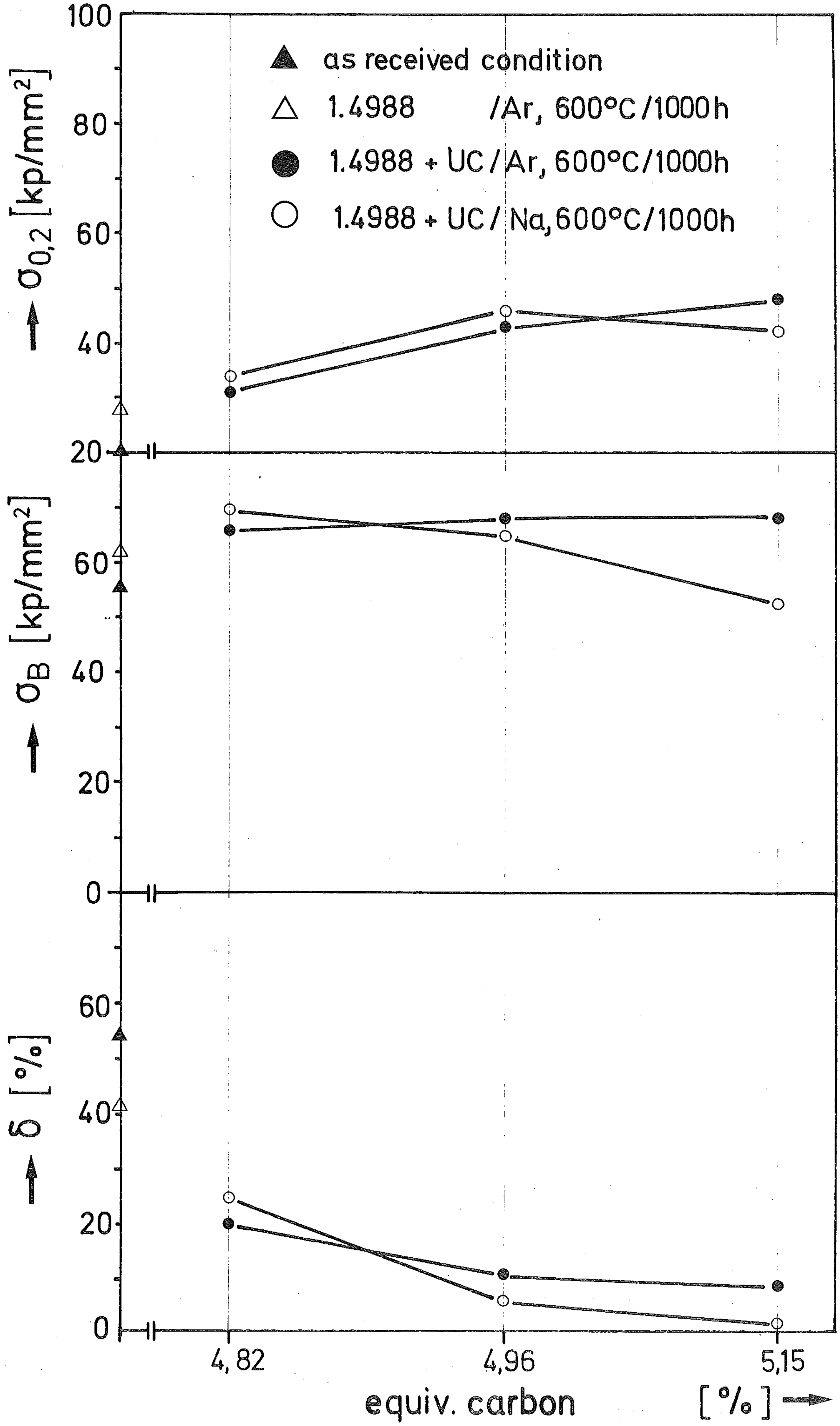
high temperature properties

Fig. 7



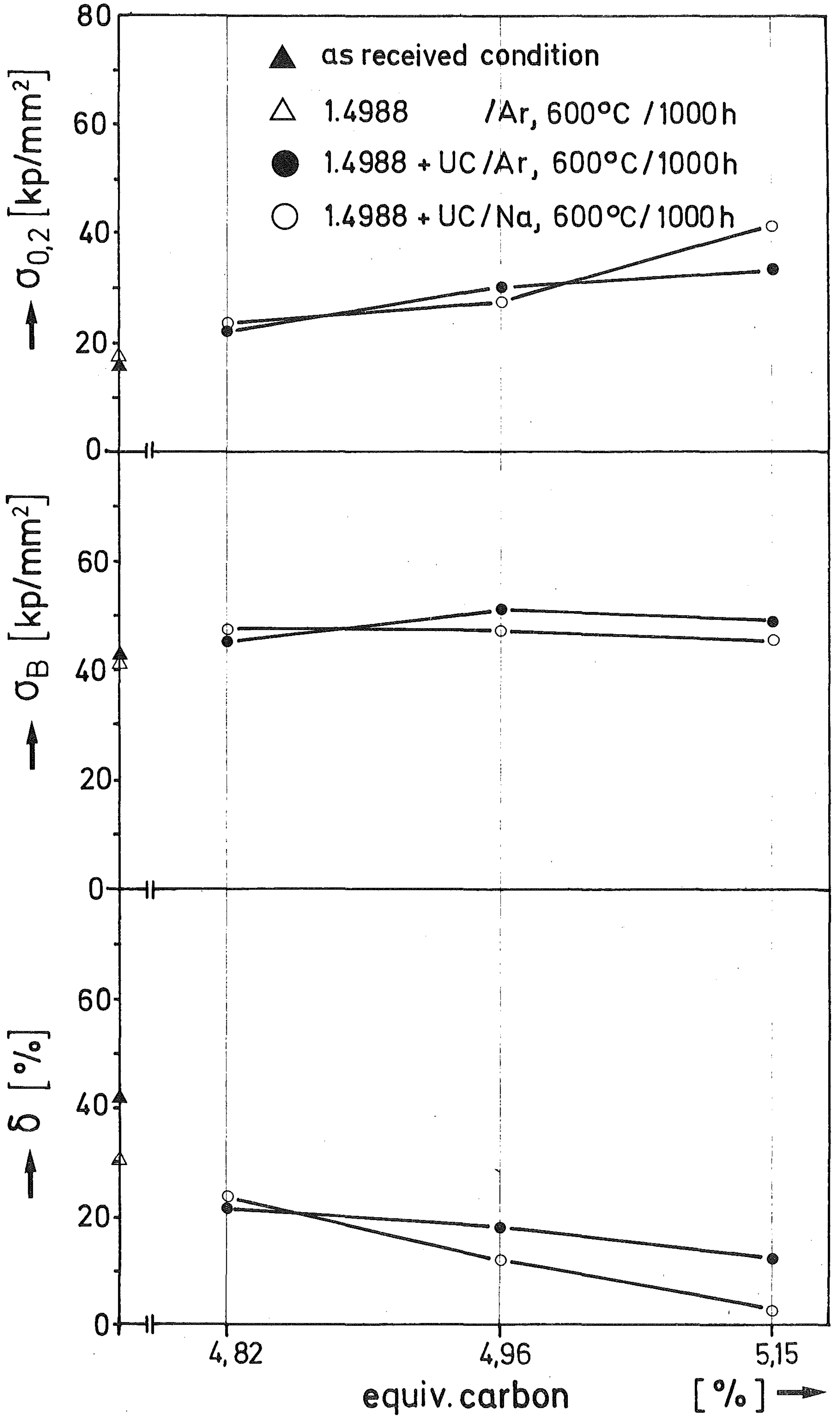
high temperature properties

Fig. 8



room temperature properties

Fig. 9



high temperature properties

Fig.10

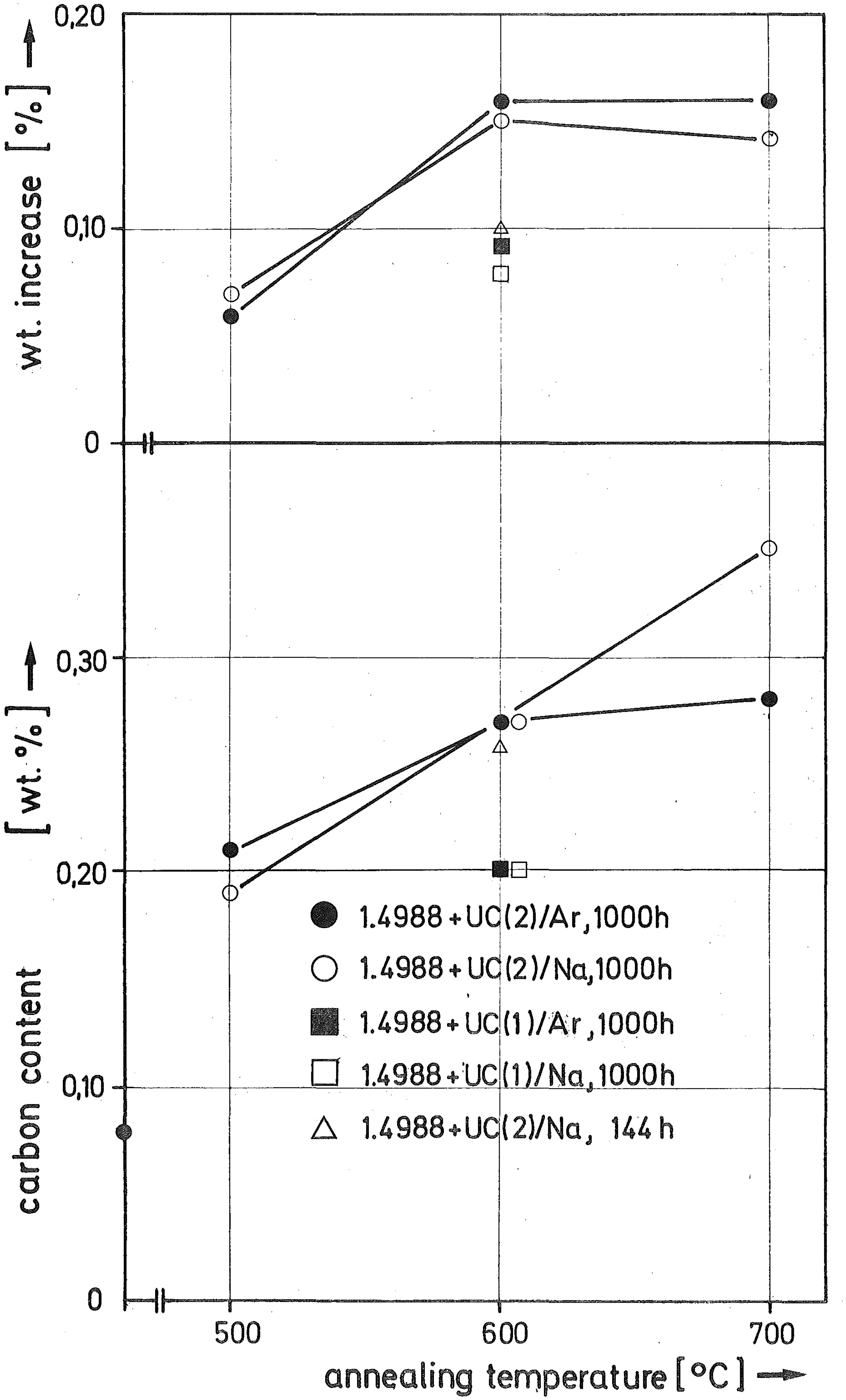

0,20
wt. increase [%]
0,10
0
0,30
[wt. %]
0,20
carbon content
0,10
0
500
600
700
annealing temperature [°C]
● 1.4988 + UC(2)/Ar, 1000h
○ 1.4988 + UC(2)/Na, 1000h
■ 1.4988 + UC(1)/Ar, 1000h
□ 1.4988 + UC(1)/Na, 1000h
△ 1.4988 + UC(2)/Na, 144 h

Fig. 11

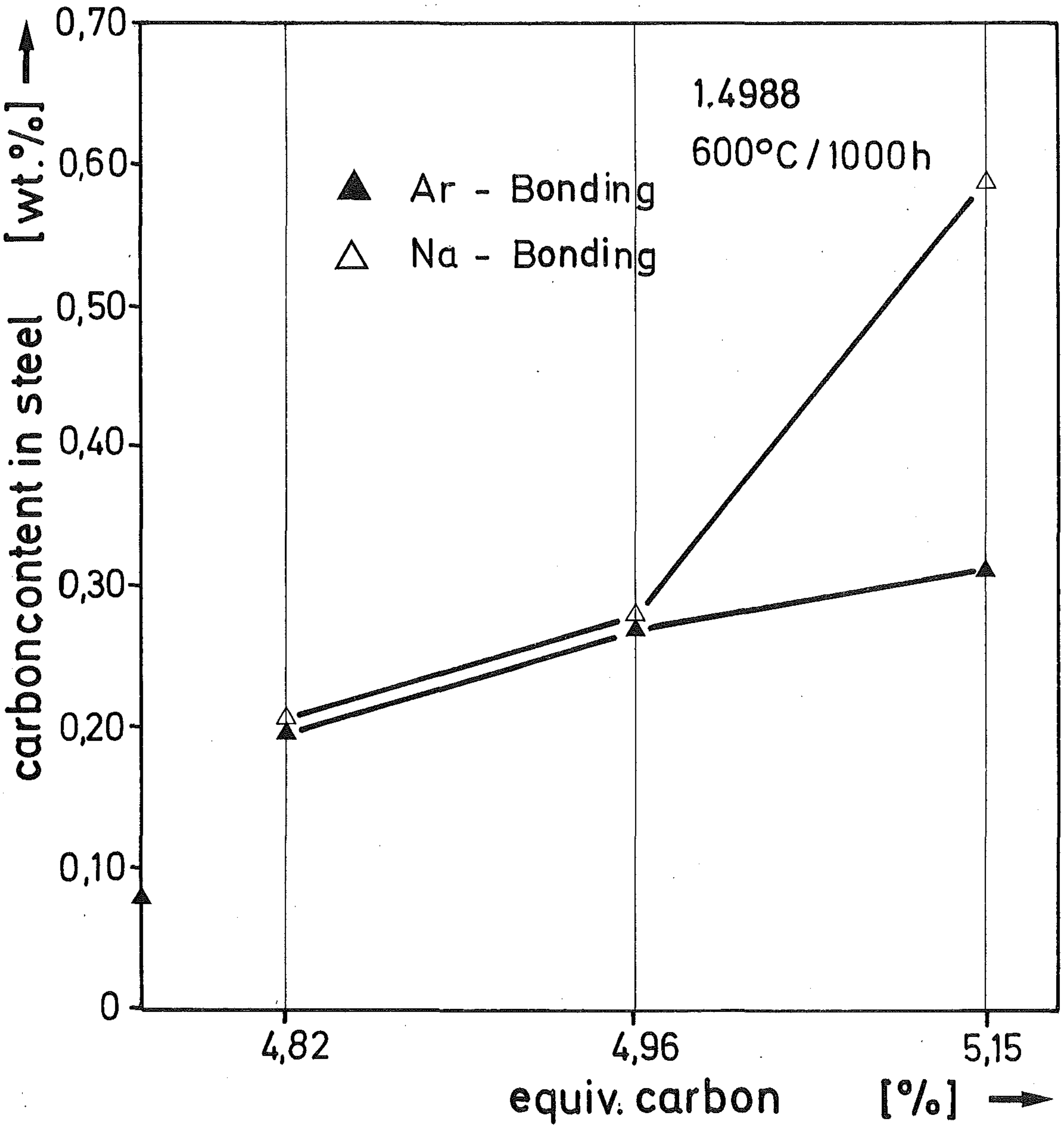

1.4988
600°C / 1000h
▲ Ar - Bonding
△ Na - Bonding
carboncontent in steel [wt.%]
0,70
0,60
0,50
0,40
0,30
0,20
0,10
0
4,82
4,96
5,15
equiv. carbon [%]

Fig.12

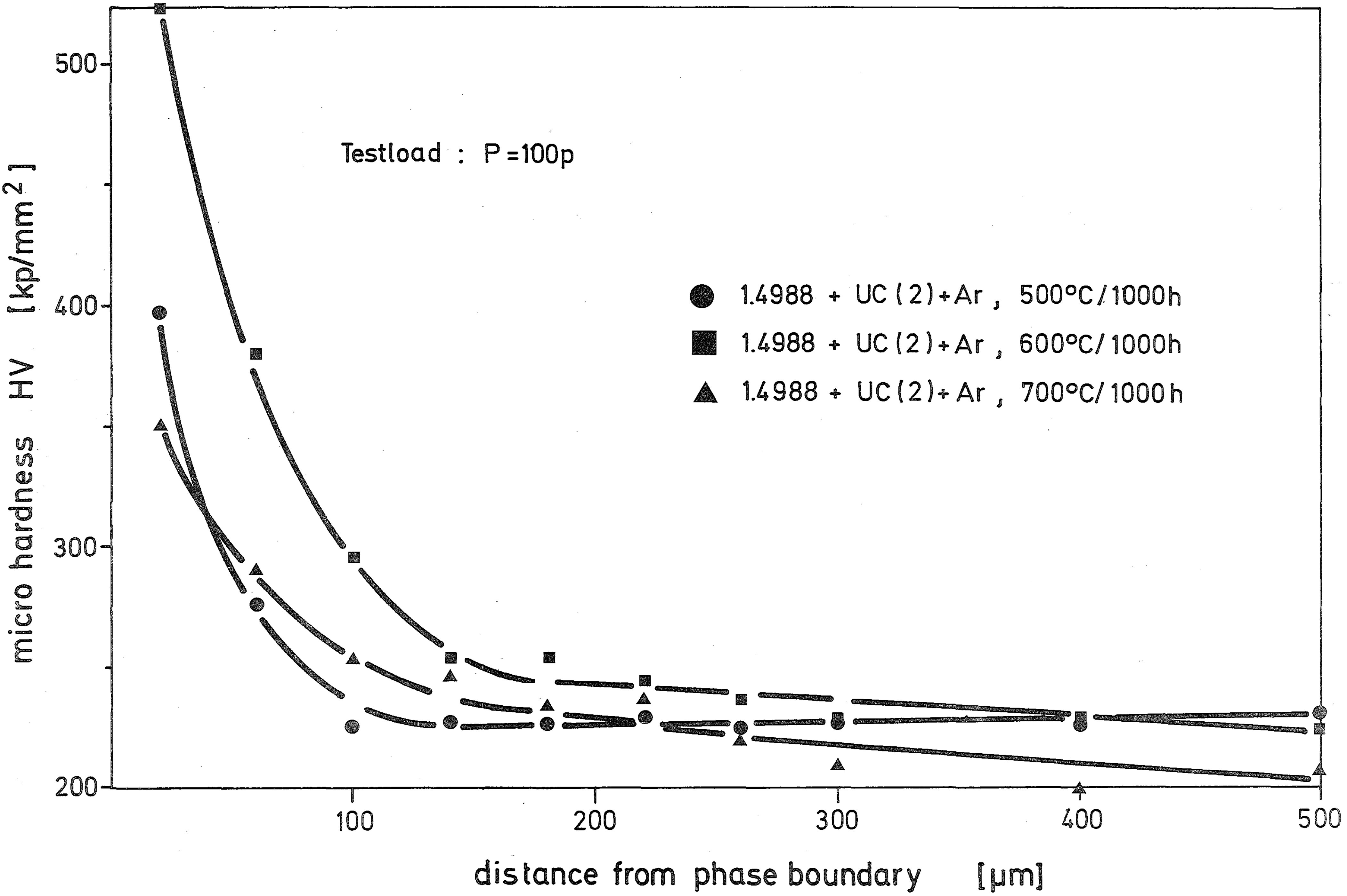

Testload : P = 100p
● 1.4988 + UC (2) + Ar , 500°C / 1000h
■ 1.4988 + UC (2) + Ar , 600°C / 1000h
▲ 1.4988 + UC (2) + Ar , 700°C / 1000h
micro hardness HV [kp/mm²]
500
400
300
200
100
200
300
400
500
distance from phase boundary [µm]

Fig. 13

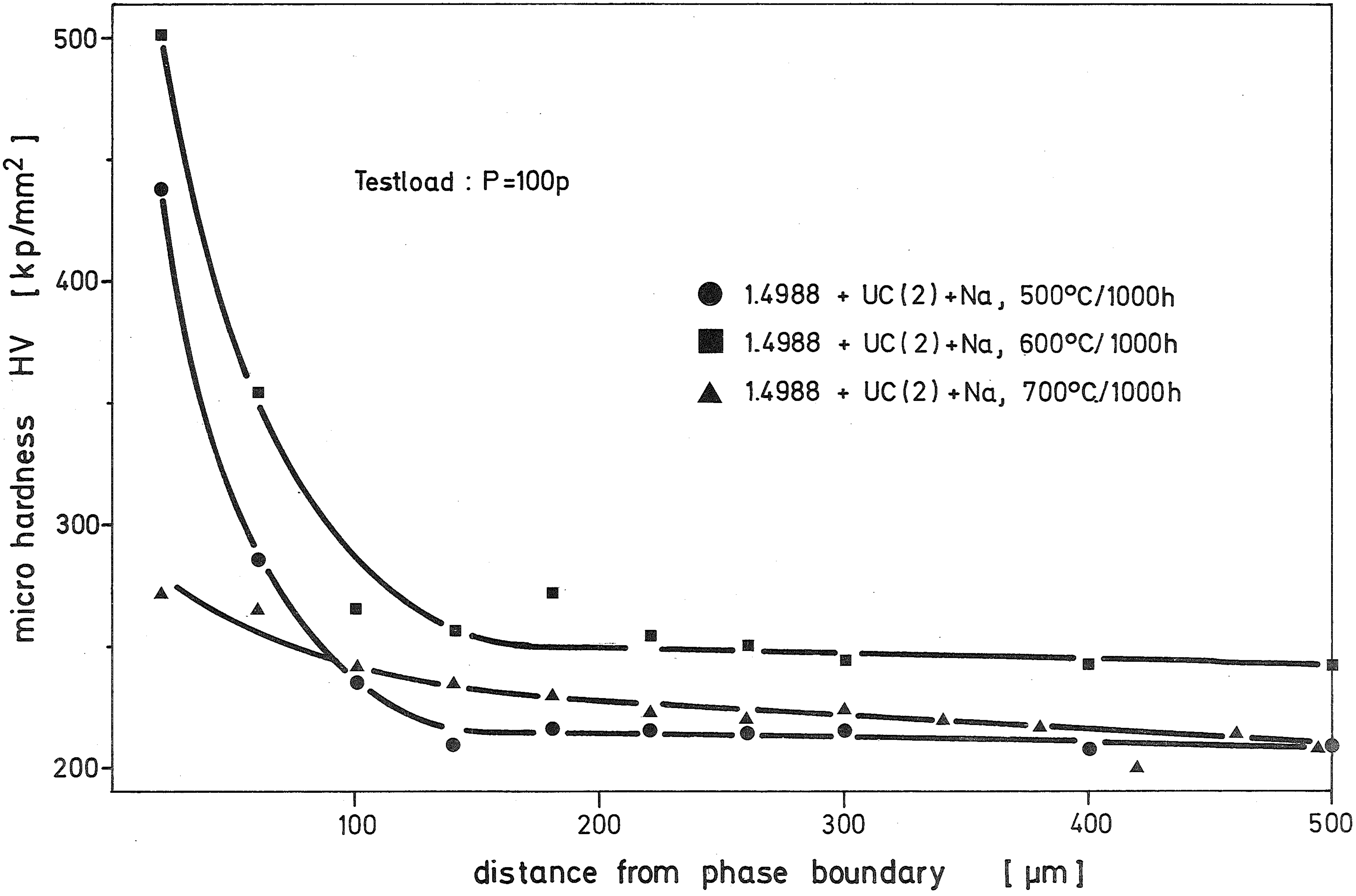
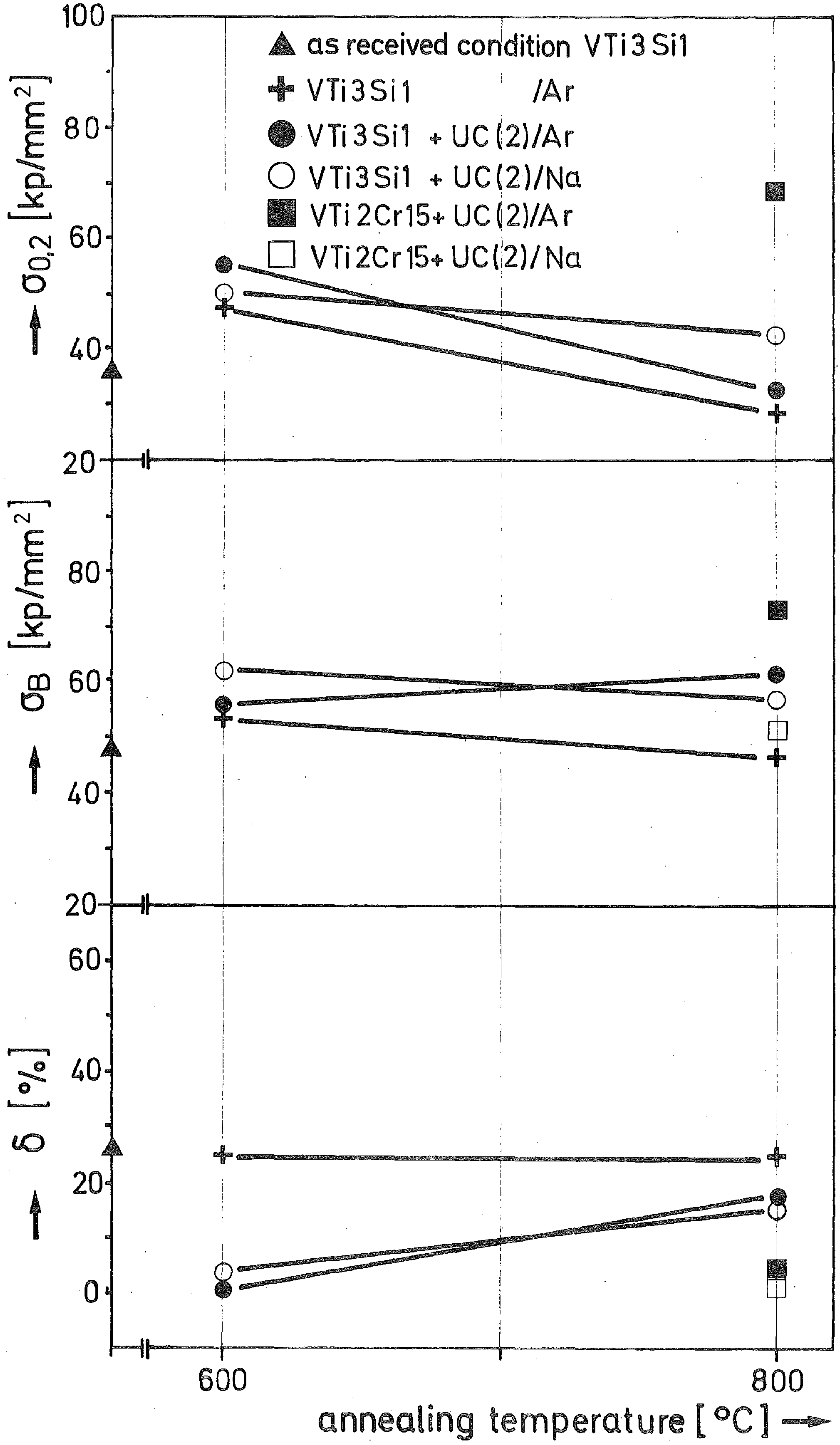
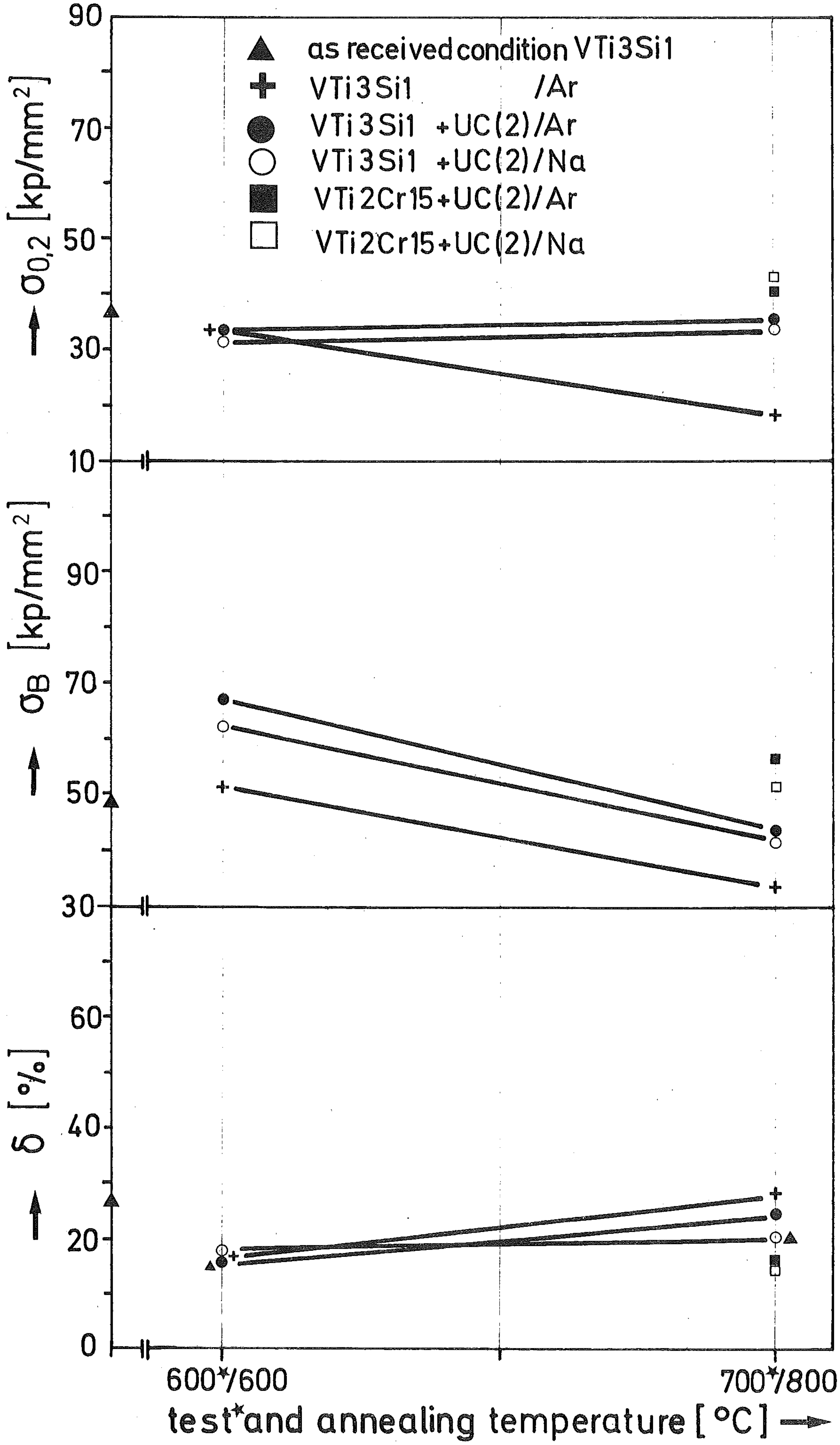

Testload : P=100p
● 1.4988 + UC(2)+Na, 500°C/1000h
■ 1.4988 + UC(2)+Na, 600°C/1000h
▲ 1.4988 + UC(2)+Na, 700°C/1000h
micro hardness HV [kp/mm²]
500
400
300
200
100
200
300
400
500
distance from phase boundary [µm]

Fig. 14



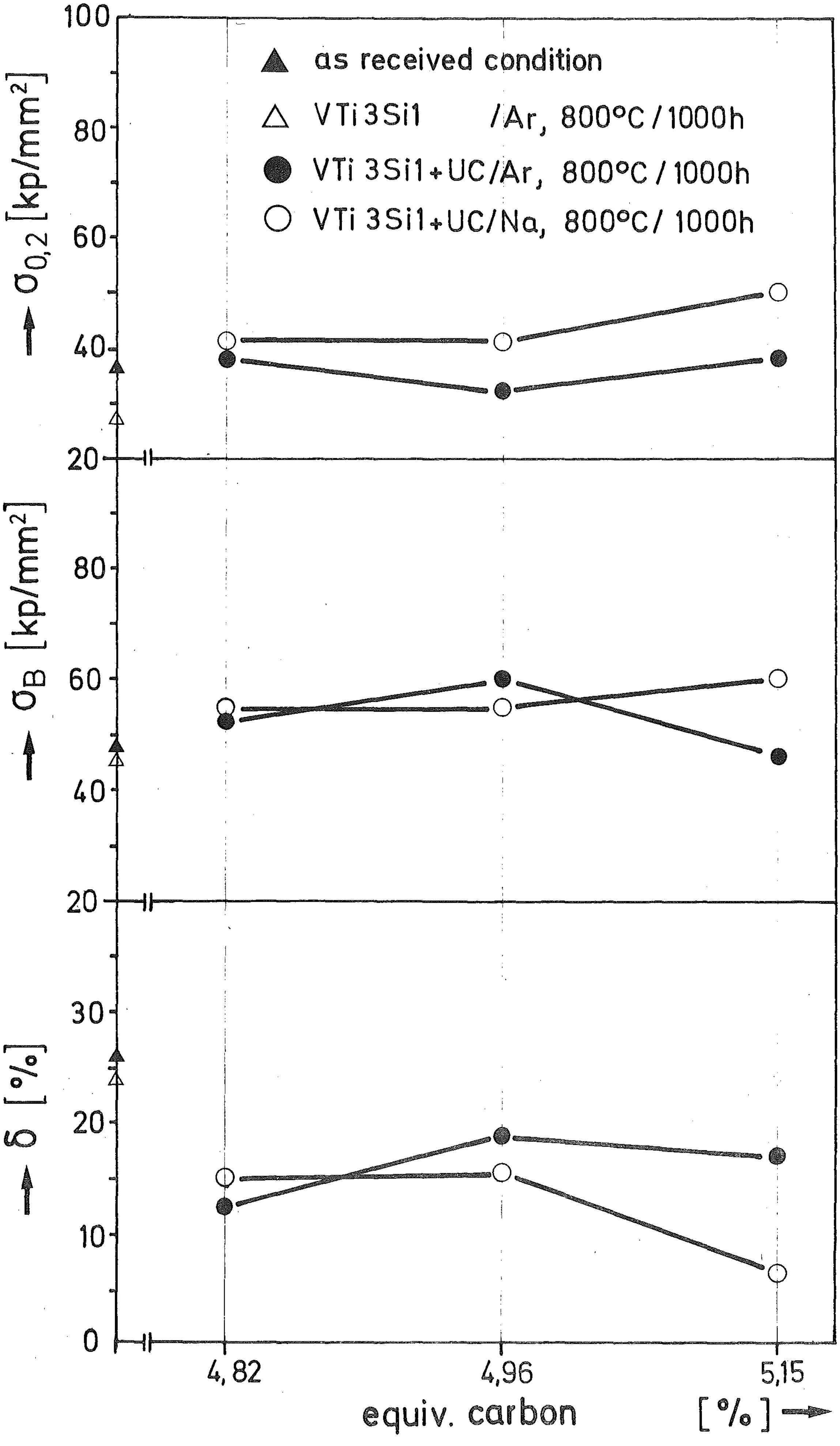
room temperature properties

Fig. 15



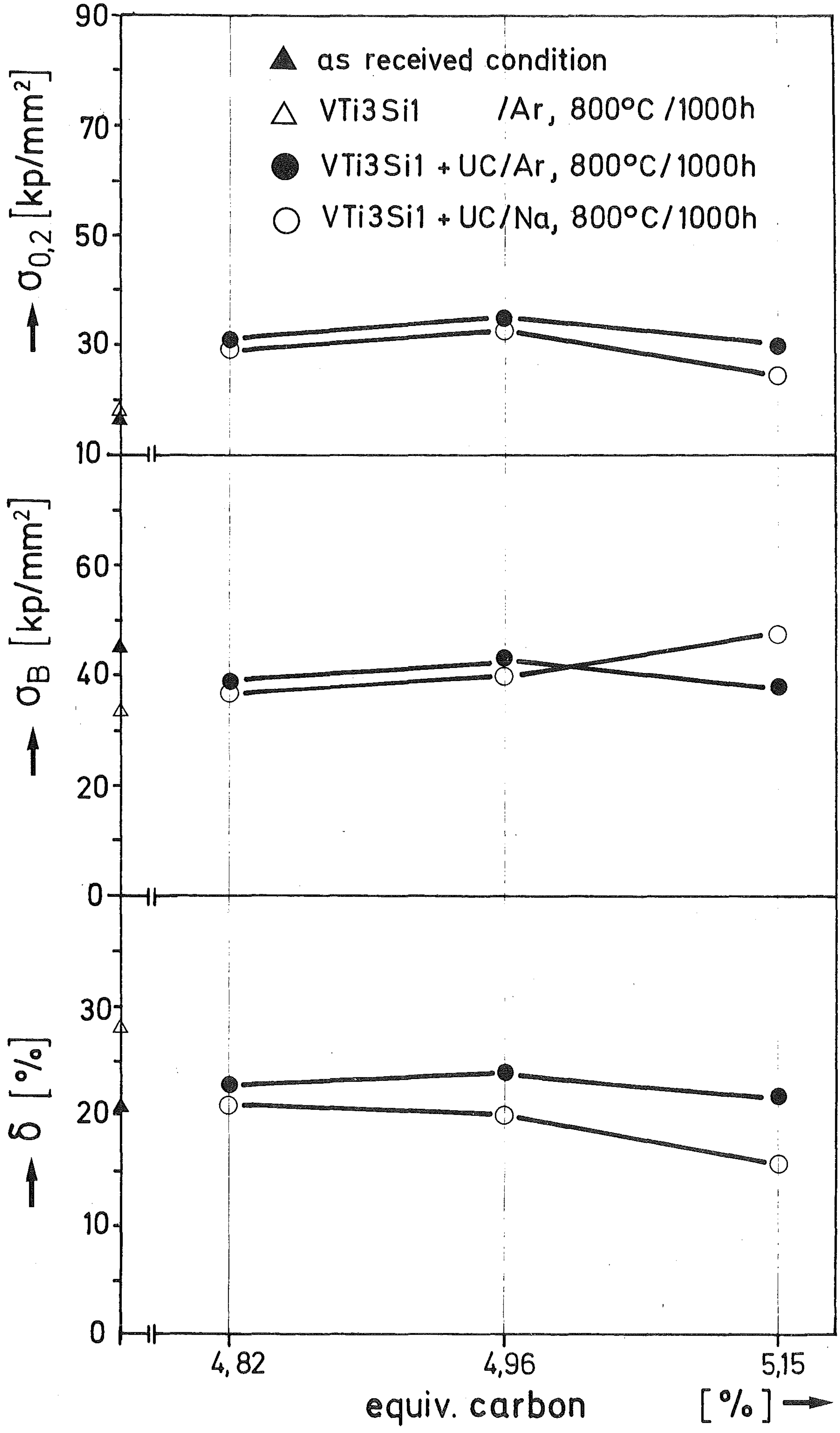
high temperature properties

Fig.16



room temperature properties

Fig. 17



high temperature properties (700°C)

Fig. 18

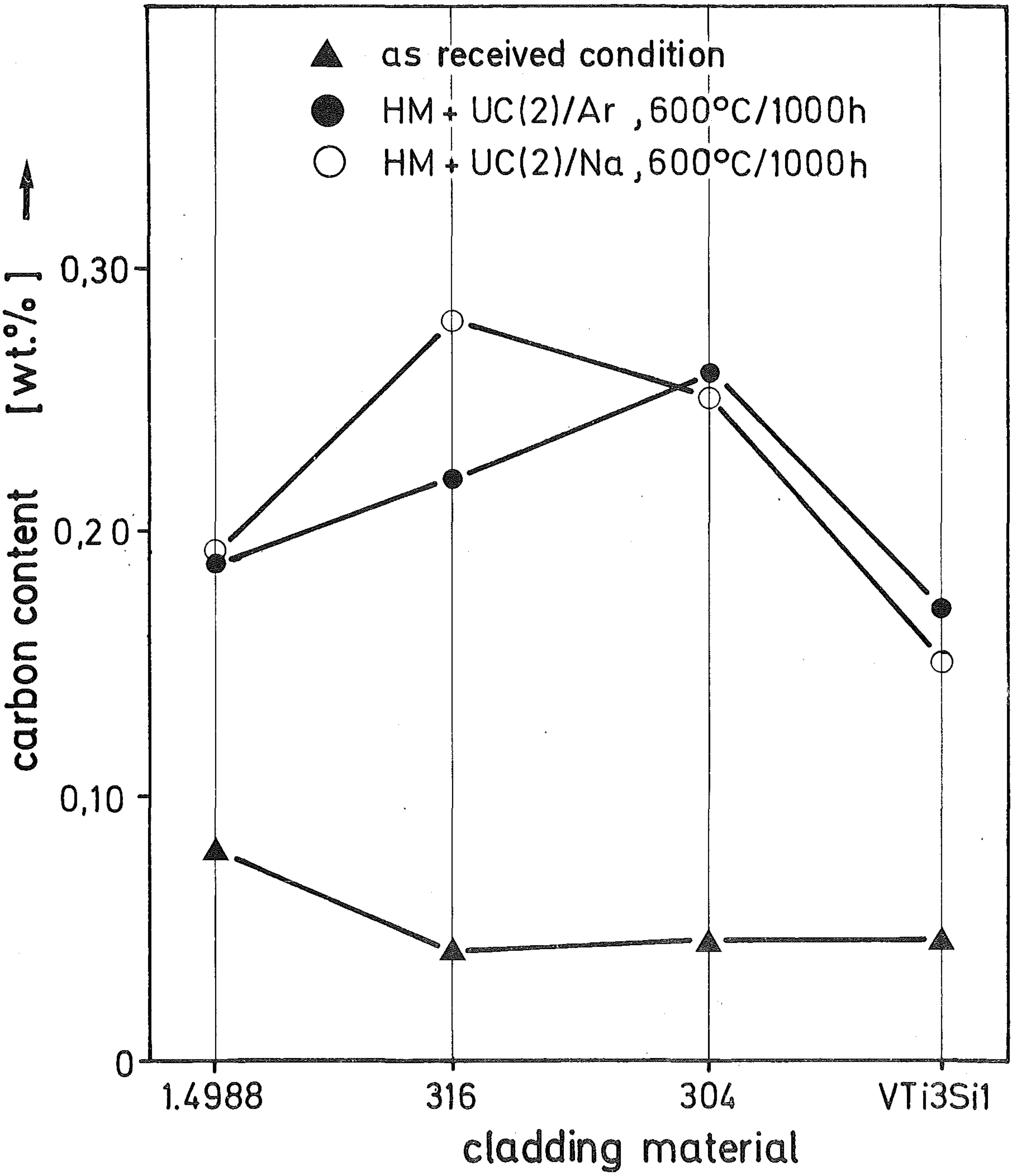
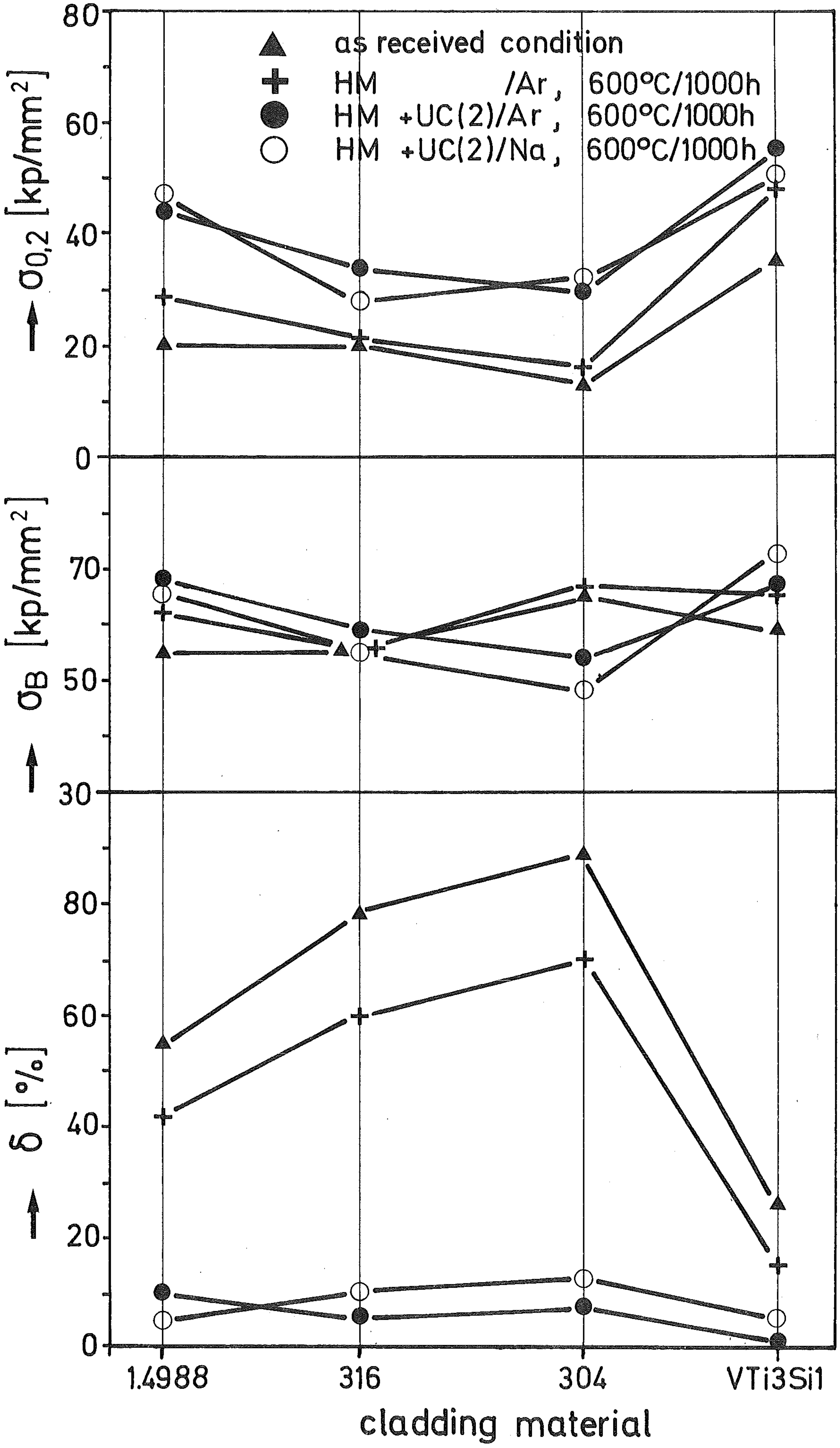

▲ as received condition
● HM + UC(2)/Ar ,600°C/1000h
○ HM + UC(2)/Na ,600°C/1000h
carbon content [wt.%] →
0,30
0,20
0,10
0
1.4988
316
304
VTi3Si1
cladding material

Fig. 19



room temperature properties

Fig. 20