Synthesis of leucite for application in dentistry

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Introduction.

Leucite is a potassium alumino-silicate ($KAlSi_2O_6$) and represents a very promising material for dental applications. It was initially introduced into dental porcelain compositions to match the thermal expansion behaviour of the ceramic to that of the metal reinforcement when cooled. In addition in last few years it was found that the presence of leucite, thanks to its phase-transition behaviour, also improves the composite strength and fracture toughness. As the amount, average crystal size and structure of the leucite phase directly affect the thermal and mechanical properties of the final composite material; suitable technology of its preparation was necessary to be developed.

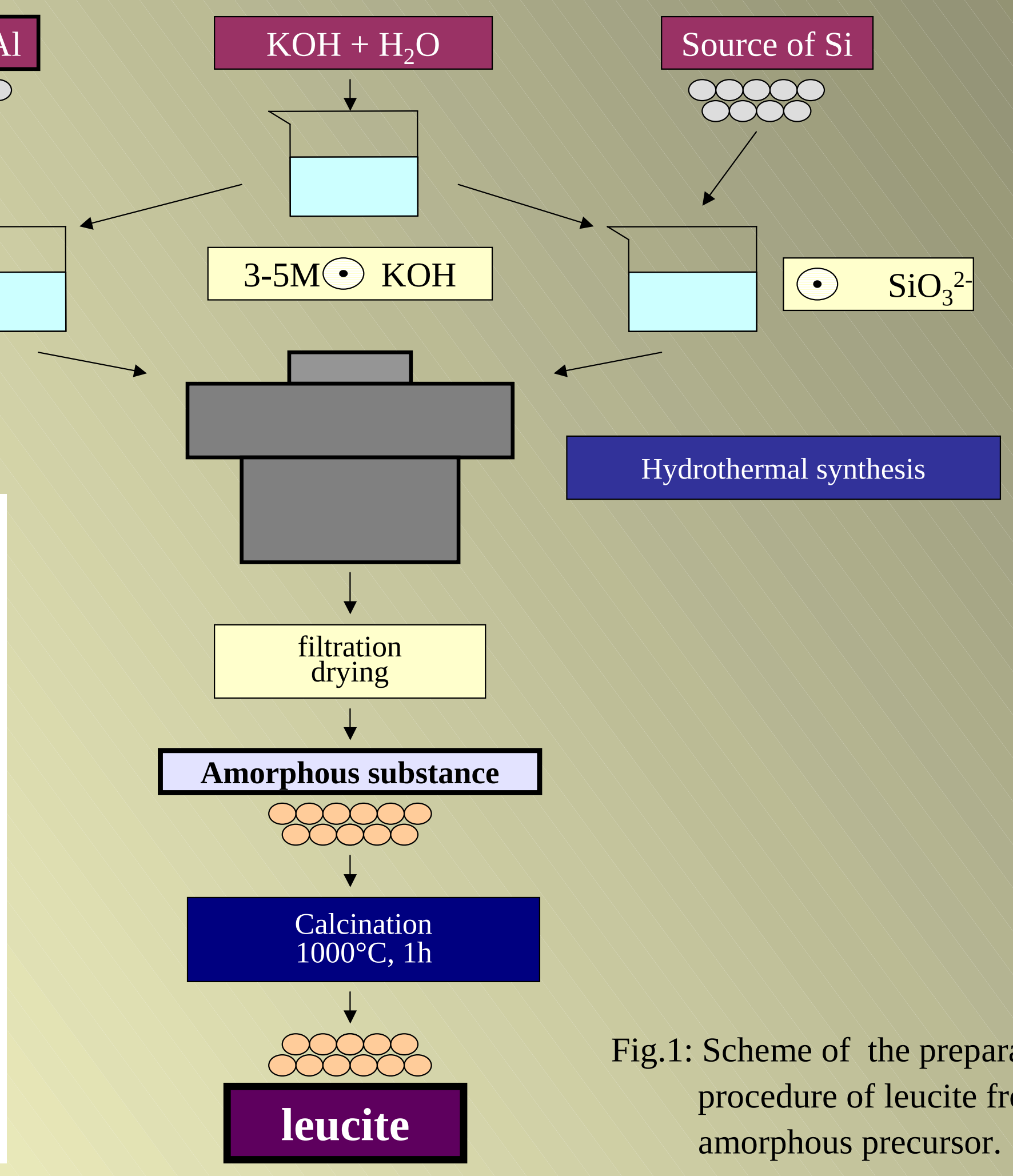
Preparation procedures of leucite ceramics were up to now proceeded from the classical technology of dental porcelain based on the crystallisation from a melt. However, the nucleation and crystal growth are very slow processes [3, 4]. Consequently the thermal treatment requires very long time, tens of hours or even several days. In addition using this procedure it is very difficult to control final microstructure of a material, especially homogenous distribution of particles in glassy matrix. Neither glass-ceramic preparation technology based on melting glass of suitable composition followed by controlled nucleation and crystal growth of leucite has not brought a desired solution [3]. The nucleation takes place only on the glass surface and so a very fine grinding of the glass is necessary to achieve sufficient degree of conversion.

On the contrary a perspective line seems to be the hydrothermal synthesis, which is used for preparation of various zeolites. This method enables to prepare homogenous leucite powders at relatively low temperature and pressure (200°C, 2 MPa) and in a relatively short time of several hours. In this presentation we report two possible technological procedures of leucite preparation, both based on the hydrothermal reaction conditions.

1. Preparation of leucite from an amorphous precursor and its subsequent calcination.

Experimental.

Preparation procedure using hydrothermal synthesis of an amorphous precursor and its subsequent high temperature treatment is schematically illustrated at Fig.1. The precursor was synthesized from a gel containing aluminium, amorphous silica and 3M potassium hydroxide solution. Reaction was carried out in a Teflon lined stainless steal autoclave at a temperature of 200°C for 2h. After the hydrothermal treatment, the content of the autoclave was washed with boiling distilled water, vacuum filtered and dried in an oven at 100°C. Leucite was then prepared by calcination at temperature of 1000°C.



Source of Al

KOH + H_2O

Source of Si

⊙ AlO_2^-

3-5M ⊙ KOH

⊙ SiO_3^{2-}

Hydrothermal synthesis

filtration
drying

Amorphous substance

Calcination
1000°C, 1h

leucite

Fig.1: Scheme of the preparation procedure of leucite from amorphous precursor.

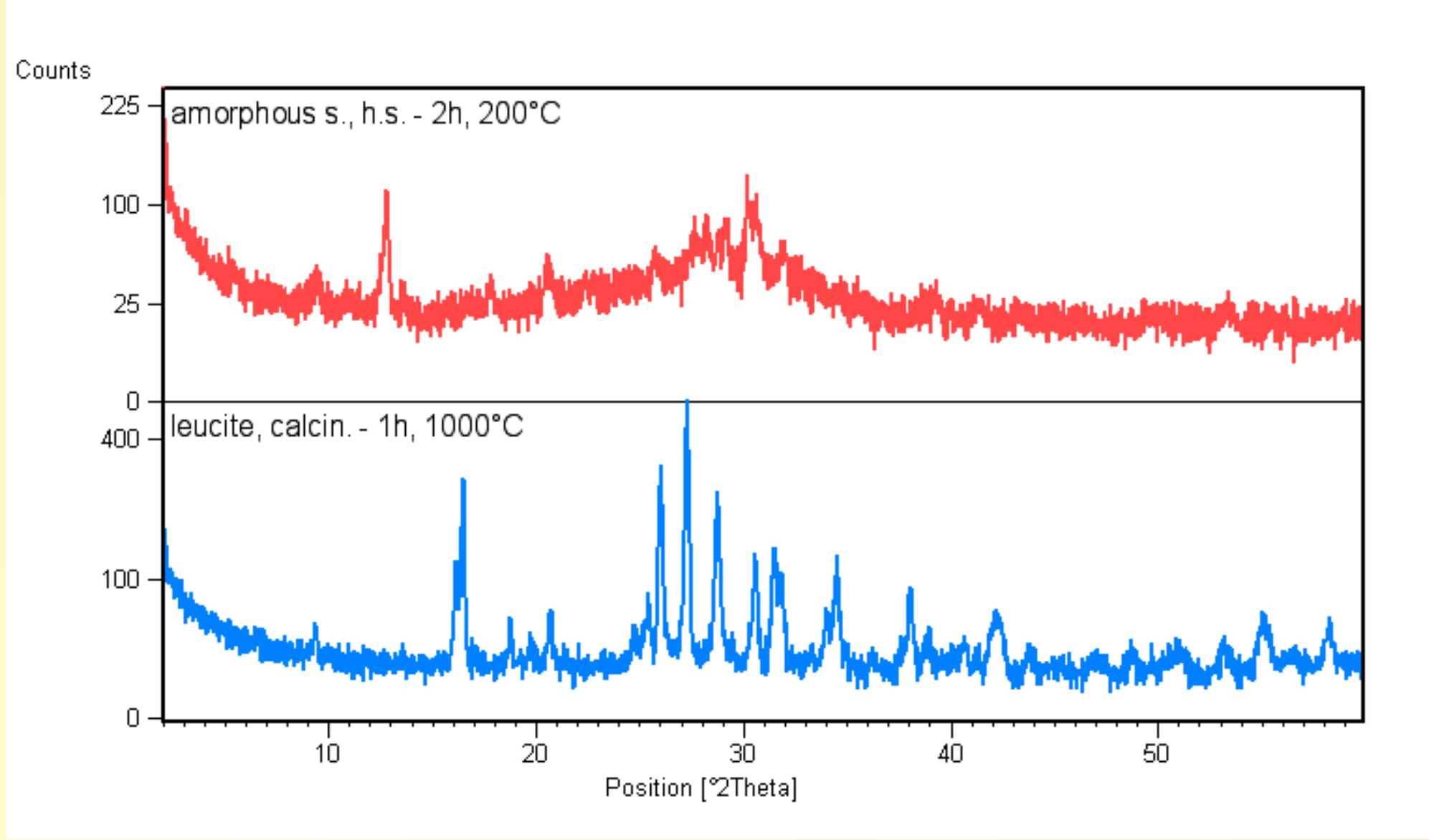


Fig.2: XRD spectra of amorphous substance and resulting leucite.

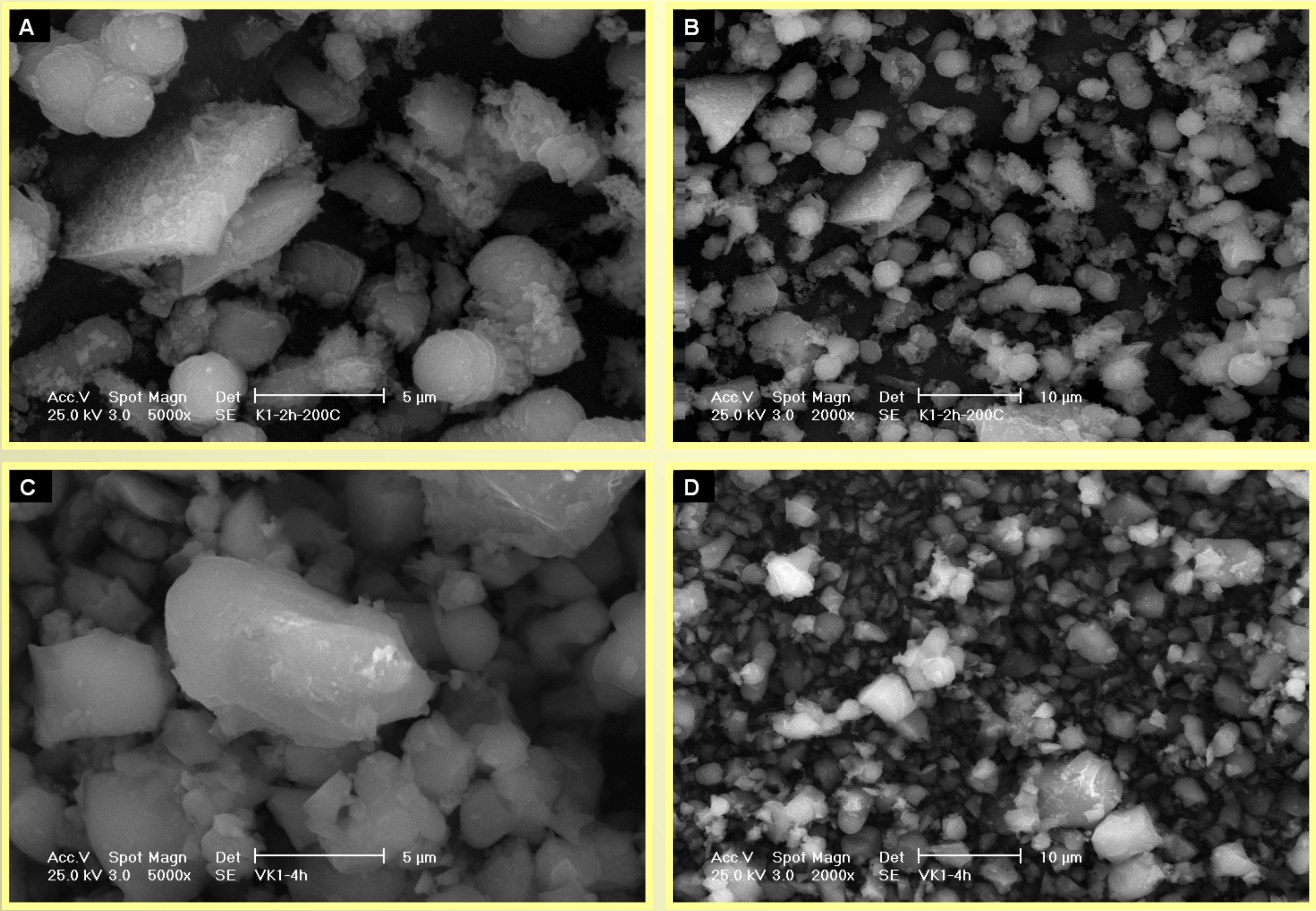


Fig.3: SEM images of the amorphous precursor (A, B) and resulting leucite (C, D).

Results.

Phase composition was identified by X-ray powder diffraction method (XRD, Philips). Particle size and their distribution were observed by scanning electron microscope (SEM, Philips XL 30 CP).

Hydrothermal treatment of the starting mixture led to the suspension homogenisation and preparation of an amorphous powder with the composition similar to leucite and particle size of 3-7µm. Subsequent calcination at temperature of 1000°C resulted in the obtaining of crystalline leucite already at dwell of 1 hour (Fig.2). Particle size and their distribution did not significantly change during the high temperature process (Fig. 3).

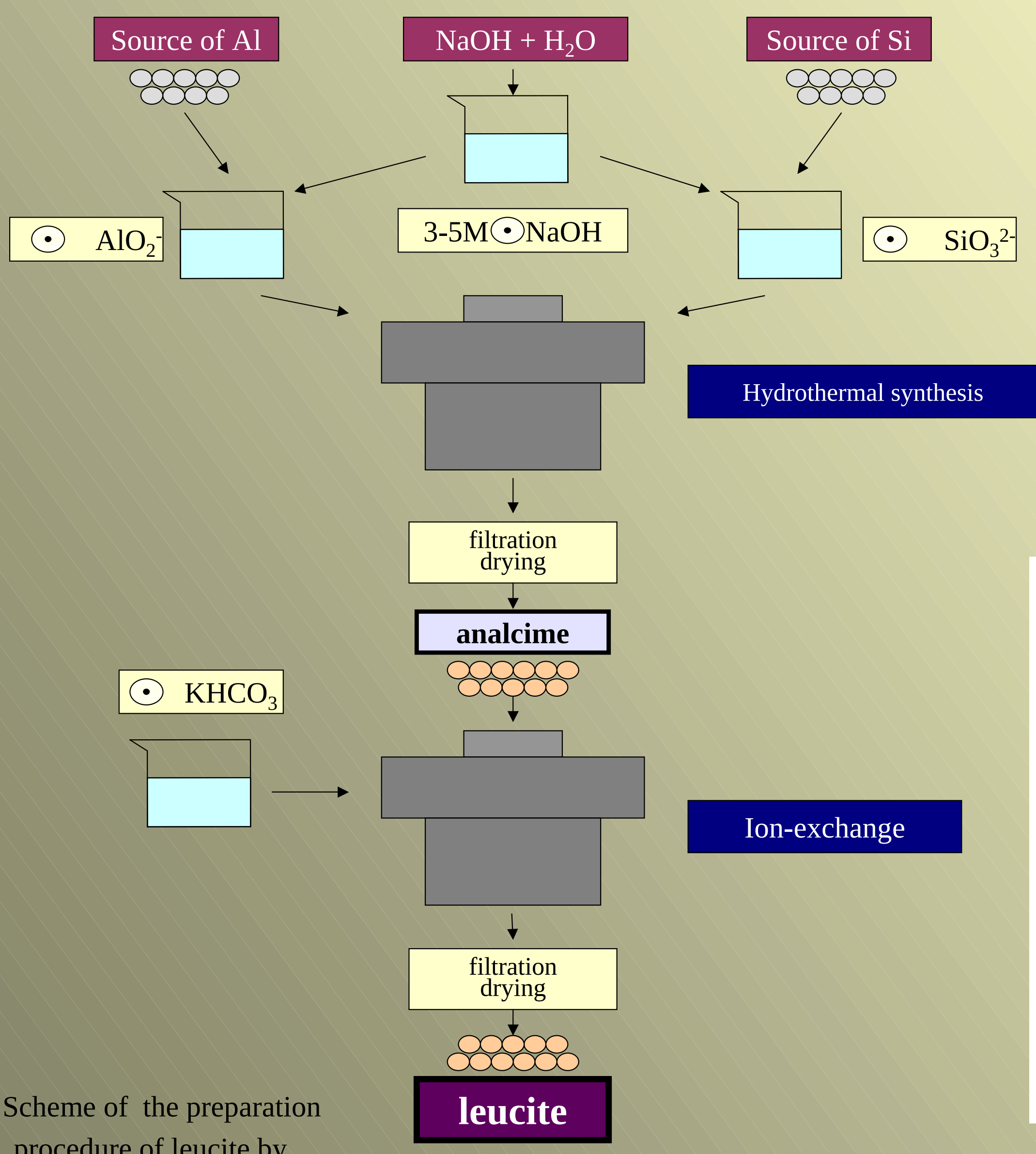
2. Preparation of leucite by ion-exchange of analcime

Experimental.

Preparation procedure of leucite by ion.exchange of analcime is schematically illustrated at Fig. 4.

Starting sols were prepared from fine-ground amorphous silica powder, sodium hydroxide solution and aluminium powder. These materials were preceded hydrothermally at a temperature of 200°C for 2h in Teflon lined stainless steal autoclave. Resulting analcime powder with the particle size of about 3-4µm was used for the ion-exchange reaction. Samples were treated with a solution of 5M KCl at a temperature of 200°C for 2h.

After the hydrothermal treatment, both intermediate and final product were thoroughly washed with boiling distilled water, vacuum filtered and dried in an oven at 100°C. Phase composition was identified by X-ray powder diffraction method (XRD, Philips). Particle size and their distribution were observed by a scanning electron microscope (SEM, Philips XL 30 CP). The degree of ion-exchange was established using Energy dispersive X-ray analysis EDAX.



Source of Al

NaOH + H_2O

Source of Si

⊙ AlO_2^-

3-5M ⊙ NaOH

⊙ SiO_3^{2-}

Hydrothermal synthesis

filtration
drying

analcime

⊙ $KHCO_3$

Ion-exchange

filtration
drying

leucite

Fig.4: Scheme of the preparation procedure of leucite by ion- exchange of analcime.

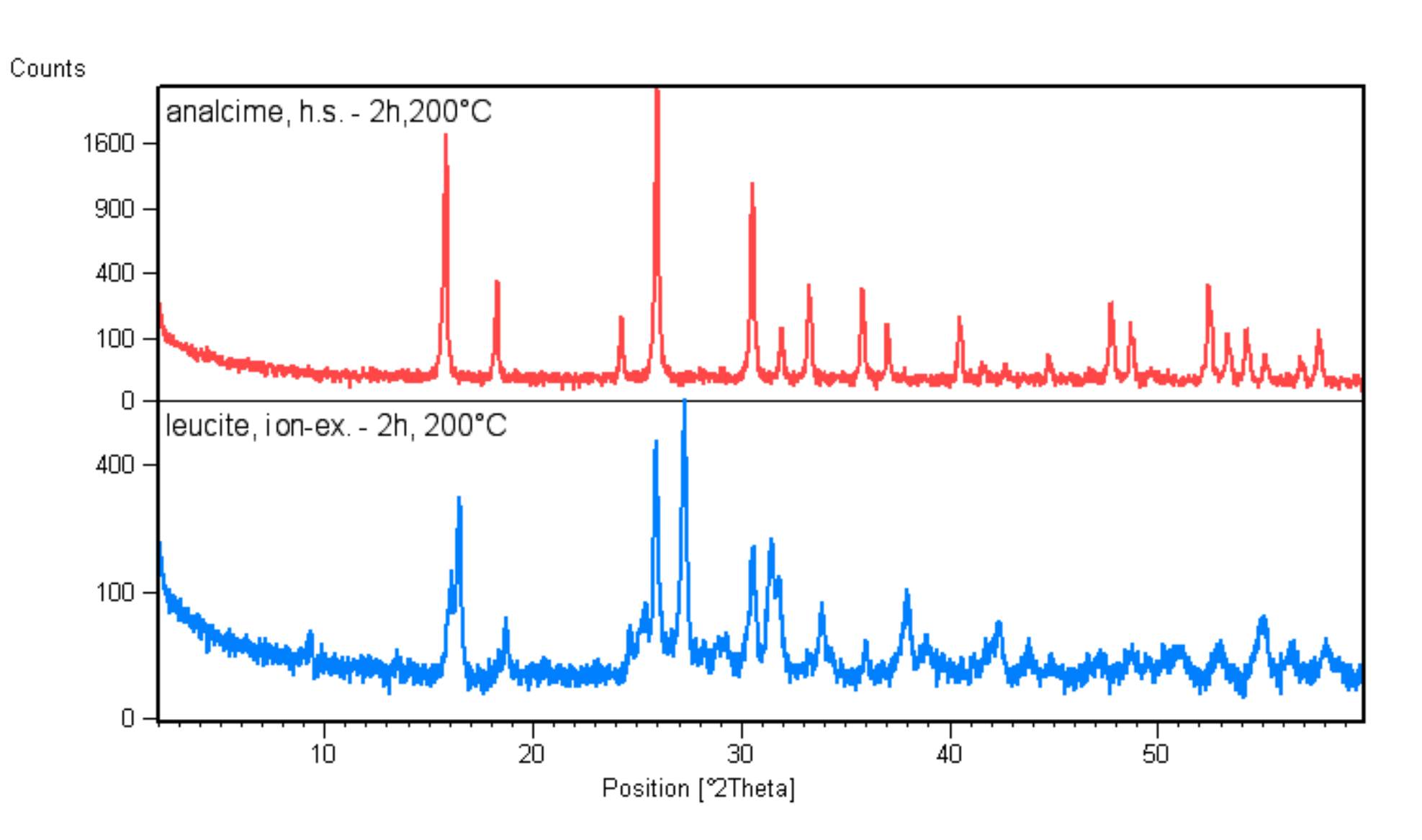


Fig.5: XRD spectra of analcime opening ion-exchange reaction and resulting leucite.

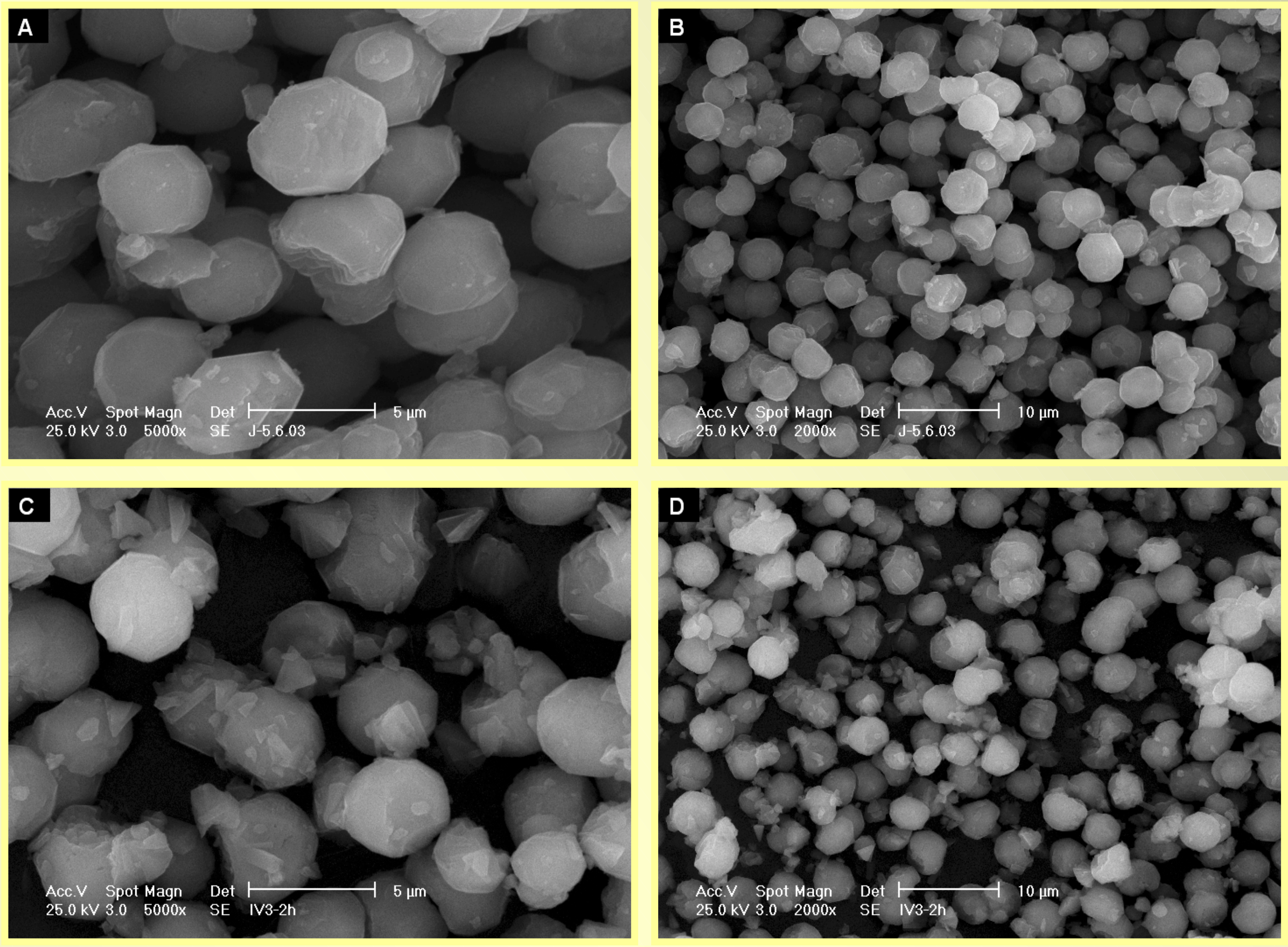


Fig.6: SEM images of analcime (A, B) and resulting leucite (C, D) after 2h oh ion-exchange reaction at 200°C.

Results.

Leucite was prepared by ion-exchange of analcime having particle size of 3-4µm. The reaction proceeded two hours at 200°C. X-ray powder diffraction analysis proved that leucite was the only crystalline phase presented in the final product (Fig. 5).

Energy dispersive X-ray analysis was used to determine chemical composition of samples and confirmed that all Na^+ ions were removed from analcime structure by ion-exchange for K^+ ions.

Scanning electron microscopy of the reacted analcime grains showed no significant changes in their size or shape. On the other hand distinct cracks were observed on grain surfaces leading to disintegration of some particles (Fig. 6). These cracks are probably due to the transformation from cubic analcime to tetragonal form of leucite accompanied by a significant decrease in solid volume.

Acknowledgements.

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References.

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Conclusions.

New technology of the synthesis of leucite was studied. Two preparation procedures were proposed, both using hydrothermal conditions. The first method, consisting in the hydrothermal synthesis of an amorphous precursor and its subsequent high temperature treatment, led to the preparation of leucite with the particle sizes ranging from 3 to 7µm. The second method - ion-exchange of analcime, resulted in homogenous leucite powder with the particles of 3-4 µm.

Up to now realised experiments show that the hydrothermal synthesis could be a suitable method for the preparation of leucite powders with the properties desired for further dental applications. Especially technology using ion-exchange of analcime was found very promising. This technology enables to prepare homogenous leucite powders at relatively low temperature and pressure (200°C, 2 MPa) and in a relatively short time of several hours.