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Appendix

Abstracts & Titles, No. 52–No. 54

BULLETIN
OF THE
FACULTY OF ENGINEERING
HOKKAIDO UNIVERSITY

NOTICE

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Study on Surface-finishing with an Elastic Wheel (III)

—On a Trial Scratch Dynamometer—

Satoru IGARASHI
Katsumasa SAITO
Koichi HOSHI

Abstract

As a fundamental study to determine the mechanism of surface-finishing of soft metals with an elastic wheel, one of the effective and appropriate methods is to perform scratch tests for soft metals with some elastic supported tool which may be considered as a model of active single grain on the surface of the elastic wheel, and to analyze the behavior of this elastic supported tool. In order to analyze the behavior of this tool, it is necessary to clarify the dynamic characteristics of the elastic materials used and especially to measure the scratch force accurately. For this purpose, a scratch dynamometer using a barium titanate ceramic as the piezoelectric transducer was made experimentally to measure the normal and horizontal components of scratch force. However, the scratch phenomenon is subtle and rapid. Thus, a thorough consideration must be made for the scratch dynamometer.

Hence in this paper, a simple theory of piezoelectric transducer was introduced and an application of this theory was made in the trial scratch dynamometer, thereafter the dynamic sensitivity and frequency characteristics were investigated. The results of calibration of sensitivity on the trial scratch dynamometer performed by means of static load and a trial electrometer tube circuit with high input resistance were reported. In addition, the results of the above mentioned static calibration were compared with that of calibration by means of dynamic load due to impact of a rubber ball on the dynamometer.

Geometrical Analysis of the Snagging Method (I)

HirotaKa YOKOUCHI
Koichi HOSHI

Abstract

A geometrical analysis of the snagging method is carried out in this report. The distribution of chip volume on the working surface of the grinding wheel and the distribution of grinding force on the same surface are obtained. By equating the vertical grinding force obtained and the vertical load, an equation which yields the relationships between the metal removal rate M and scratch depth d and other variables e.g. grinding velocity V , wheel diameter D , feed speed v , vertical load L , working radius of wheel section r and specific grinding energy u is obtained and critically discussed. It is concluded that metal removal rate M may be given approximately by $\frac{1}{k} \frac{L}{u} V$, ($k = \text{const.}$), when the ratio of d by r is practically small.

This analysis includes none of the effect of vibration of the system nor that of elastic behavior of the wheel.

A Study of Laser Machining

—On Micro Drilling Characteristics—

Noriko KUSANAGI
Toshio YUTA
Toshikazu SATO

Abstract

This paper describes some experiments on drilling by laser machining using steel as the specimen. The results obtained are as follows:

1. When the work surface is set inside of the focal plane, the drilled hole resembles a cone, and when set on the focal plane or outside of it, the hole resembles a cylinder. As regards the depth of the hole, the former was deeper than the latter.
2. When a single laser pulse is repeated to the same working point in a sequence, the hole becomes deeper, and the depth reaches a certain limit.
3. When the work surface is set close to the focal plane, the depth and the mass removal increase with the increase in the output energy, but the hole diameter

on the surface remains almost constant.

4. The thickness of the hardened surface layer caused by laser machining varies with the output of energy and the number of pulses.

On the Transitional Phenomena of Micro-Cutting

—On the Rubbing Phenomenon—

Toshio YUTA
Masaoki YAMAMOTO

Abstract

At the beginning of cutting with abrasives or cutting tools, a phenomenon which no chips are formed because the tool-edge is rubbed appears.

In order to grasp the essentials of this phenomenon and to find the factors influencing the beginning phenomenon of cutting an experiment was carried out on low speed orthogonal-cutting with a gradual increasing depth of cut using carbon steel as the specimen.

The experimental data shows that the state of contact between the tool and workpiece, in the rubbing region the tool-edge comes in contact with the surface roughness of the workpiece, in the ploughing region the tool-edge comes fully in contact with the surface and in the cutting region it perfectly contacts with the bulk, and there is a critical normal force which has a linear relation to the tool-edge roundness at the stage when the rubbing phenomenon changes into ploughing phenomenon.

The critical normal force is influenced by the nature of the workpiece and rigidity of the machine tool.

A Study on Automated Design Systems

Norio OKINO

Abstract

In recent years, a new use of computers in the field of design and drawing has been developed. This is known as "CAD", "DAC" or "ADE" etc.

This paper is a proposal on a new form of a system for automated design in

which the following two main points are included.

- (1) Man-Machine communication system managed by the computer side.
- (2) Expression of the design information by the "Pattern Matrix Method"

The first is a method of communication from designer to computer and in this paper, this new system is compared with the CAD system which has CRT and a light pen. A description of the process to realize the design was made.

The second is a concept concerning the mathematical system of the automated design, in which all patterns for designs, e.g. graphic patterns, constraints for the engineering design or characteristics of the Materials etc. are expressible in the simple mathematical formulae by the Pattern Matrix Method.

Some merits of this system are as follows:

- (1) A small computer may be sufficiently used to organize the system.
- (2) This system makes expert technical knowledge of designers unnecessary.
- (3) The results of the automated design are directly linked with numerically controlled machine tools and drafters.

Investigation of High Speed Adaptive Control Systems

—A High Speed Adaptive Control System with an Inverse
Transfer Function Type Adaptive Compensation—

Shoichi KOYAMA
Ryoichi MIURA
Hiroshi KUBO
Koji SAKURADA
Katsuhisa SHIBUKAWA
Masaru TAKAHASHI

Abstract

In this paper, the general theory of an adaptive control system employing a high speed identifier with one adjustable parameter is studied.

First, the dynamics and stability of the high speed identifier are analysed and its equivalent transfer function is derived, using a small perturbation technique. Then, the effectiveness of the high speed adaptive compensation is discussed as an ideal case. It is also shown that the adaptation system can be efficiently applicable even for the processes that are disturbed by some disturbances, being regarded them as the cause of parameter variations.

In cases where some mismatches between the actual processes and their

mathematical models exist, there are some phenomena which are unique in the high speed adaptive control system. Thus, theoretical considerations on the behaviors of the system in such cases and a detailed discussion on the effects of the mismatchings are made.

Some Considerations on the Problem of Learning Discrimination and a Formulation for the Character Extraction of Visual Pattern Recognition

Yoshiki HAGIWARA
Ryoichi MIURA
Syoichi KOYAMA

Abstract

1) In learning discrimination, the problem estimation of probability distribution and the decision in the stratiform structures of accepted information are problematic. In this paper some methods were considered and examined for application to cope with such problems.

These methods were considered for the purpose of application to such situations where the process of transforming from one stratum to the next is not merely a process of subdividing or unifying given sets, which were made in the previous division in the former stratum, but a more general process was proposed.

2) A new method of character extraction of visual patterns was presented. As microscopic transformation, the mapping φ analogous to the transformation by T. Iijima were applied. Next a macroscopic transformation was attained by means of variation of parameters of φ according to the characters of a given pattern.

3) The distance, which is introduced in the character space corresponding to the method described in 2), should be set up with the intent that the separating surface in the character space shall be as simple as possible.

From this requirement, a special distance is proposed.

Numerical Solution of Optimal Control Problems

—Consideration from two examples—

Yuzuru KUBOTA

Ryōichi MIURA

Abstract

This paper extends the variable metric minimization method of Davidon to optimal control problems. The technique is directly applicable to unconstrained and control variable constrained problems, because it carries out the problem formulation by the Maximum Principle. If terminal conditions and inequality constraints of state variables are presented, problem must be converted to an unconstrained form; e.g. by penalty functions.

Two point boundary problems must be finally solved to formulate optimal control problems by the M.P. But this is usually difficult. Hence the problems were treated by extremum seeking methods of Hamiltonian functions. Consequently, it becomes a problem of extremum seeking methods in functional space carried out in such a way as to obtain gradients by solving the initial value problems. According to the two examples presented, the authors compared the present method with Davidon's method and the Modified Steepest Descent method.

A good approximate solution was obtained by iterative calculations.

On the Adaptive Control to Cutting Process

—A Study on the Influence of the Errors
in the Used Tool-life Equation—

Yasuki SEKIGUCHI

Shōichi KOYAMA

Ryōichi MIURA

Abstract

It is well known that the cutting condition is one of the most important factors determining the economy of machining. Many papers have been written on the mode of determination of the optimal cutting condition, using such performance indexes as machining cost, machining efficiency or profit-rate. The off-line method was used in most of these papers and the adaptive control technique has been applied in more recent papers. In any event, an experimental tool-life equation must be used. This equation invariably has some errors against the real tool-life owing to the variety of tools, materials and conditions etc. These errors cause the estimated optimal cutting condition to veer from the actual optimal cutting.

The authors paid special attention to this discrepancy and studied its influence in the case where the machining cost per piece was used as a performance index for the adaptive-controlled turning operation by digital simulation. And the following results were made clear;

1) When the machine operation cost is low and the tool-life is high and, moreover, the high precision in machining is required, the more detailed investigation on the tool-life equation would be required.

2) And in such a case, a study on the relation between cutting temperature and tool-life by a logarithmic graph showed that the temperature intercept and the mutual relation between the intercept and the slope is especially important.

In conclusion, if the economy of machining is aggravated to such an extent that it becomes impossible to neglect the increased part of the cost against the cost of production, a higher adaptive control loop might be needed to identify and correct the parameters of the tool-life equation on line.

Design of the Linear Feedback System Considering the Parameter Variation

Hiroyuki KANNO
Shōichi KOYAMA
Ryōichi MIURA

Abstract

Generally a process or plant has variable parameters which affect the performance of the control system. When parameters of a process (i.e. gain, pole and zero) are varied, the influence on the feedback system is represented by the relative variation $V(s)$ of a closed loop system, which is defined as follows,

$$V(s) = \ln T(s) - \ln T_0(s)$$

$T(s)$ and $T_0(s)$ are over-all transfer functions with varied parameters and nominal parameters respectively. $V(s)$ is formulated on the basis of Horowitz's graphical design in order to fit the analytical design. If $L_0(s)$ is the loop transmission the design procedure is simply a matter of choosing a sufficiently large $L_0(s)$ over the real frequency range for which small $V(s)$ is desired.

It is emphasized by using $V(s)$ that a linear feedback system has a considerable adaptive capability, if it is properly designed according to our new analytical method. And the limitation of the reduction of parameter variation has been also discussed.

An Observation of Magnetic Domains by the Kerr Effect and its Application to the Analysis of Residual Stress

Hitoshi NAKAE
Iwao ISHIDA
Juichi NODA
Kunihiko KONOMA

Abstract

The observation of magnetic domains by the magneto-optic Kerr effect was carried out on single crystals of 3% Si-Fe alloy.

In the magnetization process, a small number of both edge-shaped 90°-domains

on (100), and spike-shaped 180°-domains on (110) crystals remained unchanged by considerably high magnetic fields. The formation of these domains might be affected by the discontinuity of grain boundary migration during growth.

Several forms of domain configurations were observed after mechanical working, presumably resulting from its residual stress. Under certain assumptions, the residual stress was estimated by the change of domain pattern with the applied magnetic field.

The Effect of Heating Rate on the Grain Growth of 3.25% Si-Fe Alloy Sheets

Hitoshi NAKAE
Noriaki GOSHI
Yoshihiro BANNO
Akira OKADA

Abstract

The effect of heating rate on texture formation after grain growth were investigated on relatively high purity 3.25% Si-Fe alloy sheets. The specimens were heated at (1) 24°C/h, (2) 150°C/h and (3) rapid heating from 600°C and were held for 1 hr at 800°C, 900°C, 1000°C and 1100°C. Each annealing texture was determined by optical microscopic observation, magnetic torque meter and x-ray diffractometer.

In a slow heating up process, the grain growth was retarded and a large amount of (100) [011] components still remained unrecrystallized even in specimens annealed for 1 hr at 800°C. By decreasing the heating rate, the annealing texture developed well. Thus, it may be deduced that the heating rate has an alternative effect as impurity on the texture formation.

Experimental Investigation to Produce Single Crystal of Fe-Si Alloy by Strain-Anneal Method (III)

—On the (100) single crystal—

Kōsuke TAGASHIRA
Nobuaki ENOMOTO
Hitoshi NAKAE

Abstract

In preparing single crystals of 3.25% Si-Fe alloys, a new method with a high temperature-gradient (500°C/cm) furnace specially designed for this purpose was adopted. The modifications of the conventional strain-anneal method were as follows: (1) specimens were strained by a slight rolling and not by tension, (2) after which the surface layer resulting from rolling was removed by chemical etching.

By this process, large grained single crystals with various orientations were obtained, in which the most suitable reduction rates for the growth of (110) and (100) single crystals were about 2% and 4%, respectively.

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Statistical Analysis of the Monthly Runoff for the Ran Young River, Formosa

CHANG Yu Tien
Ken-ichi HIRAYAMA
Tsutomu KISHI

Abstract

The sequence of the monthly runoff for the Ran Young River, Formosa observed from 1950 to 1966 was analyzed by the theory of stationary time series.

The mathematical structures of the sequence and the possibility of the statistical prediction are discussed.

The sequence was first separated into the periodic deterministic component and the stochastic component. Then, four mathematical models were applied for the stochastic component to compare the efficiency of analysis, i.e., (1) a purely random process, (2) the 1st order Markov process, (3) the 2nd order Markov process and (4) the moving average scheme (which is equal to the 6th order Markov process in the present example).

The following facts were found from the results of calculation:

- a) For the periodic component, 12, 6 and 4 month periods were detected at 95% level of confidence by means of the auto-correlation analysis.
- b) Mathematically speaking, the correlogram of the stochastic component did not indicate pure randomness, but showed a weak persistency of the process.
- c) The efficiencies of analysis were 11% for the 1st order Markov process, 12% for the 2nd order Markov process and 13% for the moving average scheme, respectively.

A Study of Traffic Assignment to Arterial Street Network

Etsuo YAMAMURA

Abstract

In this paper, two problems were considered.

One is the method of solution for arterial street network capacity and the other is the traffic assignment method that the inter-zone trips are assigned to a network of arterial streets.

We applied the network flow theory and the discrete maximum principle in order to formulate the traffic assignment process.

According to a combination of two methods, we applied them to the traffic assignment of the arterial street network in Sapporo City.

The Effect of Anions and Cations on the Kinetics of Anodic Oxide Film Growth on Iron in Neutral Solution

Takenori NOTOYA
Norio SATO

Abstract

The growth kinetics of anodic oxide film on iron in neutral solutions were examined using electrochemical techniques to examine the effect of anions and cations. The growth rate observed by galvanostatic and potentiostatic oxidations was analysed in the form, $i = k \exp(\beta E - Q_T/B)$, leading to $E = E' + KQ_T$, the polarization potential at constant growth rate, and $i = A \exp(-Q_T/B)$, the growth rate at constant potential, where i is the growth rate, E the potential, Q_T the film thickness, and where k , β , B , and K are constants.

Kinetic parameter K was found to change depending on the kind of anions in the order, $\text{NO}_3^- > \text{SO}_4^{2-} > \text{ClO}_4^- > \text{CH}_3\text{COO}^- > \text{BO}_3^{3-}$, while kinetic parameter B was ordered $\text{ClO}_4^- > \text{NO}_3^- > \text{CH}_3\text{COO}^- > \text{PO}_4^{3-} > \text{SO}_4^{2-} > \text{BO}_3^{3-}$. We found no effect of cations on the growth kinetics, the cations examined were Li^+ , K^+ , Na^+ and Ca^{2+} .

The Effects of Buoyancy on the Velocity and Temperature Distributions in a Vertical Wake of a Heated Body

Shoichiro FUKUSAKO
Masaru KIYA
Mikio ARIE

Abstract

The effects of buoyancy on the laminar and turbulent wakes of a heated body placed in a vertically ascending flow of an incompressible fluid are described on

the basis of similarity solutions. By applying Prandtl's mixing length hypotheses, a closed solution was obtained for the turbulent wake, while a closed solution for the laminar wake was obtained only when Prandtl number σ was equal to unity.

The buoyancy was found to give a considerable effect on the flow in the wake.

It was found from the present analysis that the width of the laminar wake increases in proportion to $x_1^{1/2}$, x_1 being the longitudinal distance from the heated body for both two-dimensional and axisymmetric cases. In the case of a turbulent wake, on the other hand, the width increases in proportion to x_1 (two-dimensional) and $x_1^{2/3}$ (axisymmetric).

The decrease of temperature excess on the axis of the laminar wake will be proportional to $x_1^{-1/2}$ (two-dimensional) and to x_1^{-1} (axisymmetric), and that of the turbulent wake to x_1^{-1} (two-dimensional) and to $x_1^{-4/3}$ (axisymmetric).

The maximum velocity-defect on the wake axis was found to change in proportion to $x_1^{1/2}$ (two-dimensional) and to x_1^0 (axisymmetric) for the laminar case, and to x_1^0 (two-dimensional) and to $x_1^{-1/3}$ (axisymmetric) for the turbulent case.

A Contribution to the Spreading of an Axisymmetric Wake with Streamwise Pressure Gradient

Shoichiro FUKUSAKO

Masaru KIYA

Mikio ARIE

Abstract

A Similar solution of axisymmetric laminar or turbulent wake of a body placed in a flow with streamwise pressure gradient was obtained on the assumption that the velocity outside the wake is proportional to x_1^p .

It was shown that the width of the laminar and turbulent wakes increases in proportion to $x_1^{(1-p)/2}$, and $x_1^{(1-3p)/3}$, respectively. On the other hand, the maximum velocity-defect in the wake decreases in proportion to $x_1^{-(1+p)}$ and $x_1^{-2/3}$ in laminar and turbulent wakes, respectively.

However, the velocity profile in the wake shows an equivalent from with that in the wake without a pressure gradient for both laminar and turbulent cases.

On a Few Trials of an Engine Pressure Indicator using a New Piezo Element

Hiroaki KIKKAWA
Tadashi MURAYAMA
Shoichi FUKAZAWA

Abstract

Pressure indicators which have a high performance are frequently required for studying the combustion phenomena of an engine.

Indicators such as strain gauge types or quartz types have been used commonly.

However, the later type is lacking in mechanical strength and in maintaining an electric charge, and the former is not sufficiently sensitive and is not built for durability. Some kinds of piezo electric elements with excellent characteristics over those of quartz have been developed recently. They are, for example, $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$, $\text{Pb}(\text{Mg}_{1/3}, \text{Nb}_{2/3})\text{O}_3$ - PbTiO_3 - PbZrO_3 . Engine pressure indicators using such elements were made and their characteristics were investigated in this paper. As a result, they were found to have superior properties.

A Study on the Melting Characteristics of the Coal Ash

Shoichi FUKAZAWA
Hiroshi TANIGUCHI
Masaharu KOBAYASHI
Kiyotaka YAMANE

Abstract

The melting characteristics of coal ash is a very important factor in the design and operation of the coal fired furnaces. It is known that coal ash consists of many kinds of mineral components, and it is difficult to predict the melting characteristics of the coal ash from its chemical composition.

In this study, using a Leitz heating microscope, the melting characteristics of the coal ash samples were measured by means of JIS (M 8801) or DIN (51730). The experimental results by the above two methods showed identical values. It was found that the melting characteristics of artificial ashes, which had the same composition as the coal ashes, were approximately equal to those of the coal ashes.

From the above mentioned results, artificial ashes may be used as models of the coal ashes. We attempted to express the relationships between the melting points and chemical compositions of the ashes by an empirical formula. The calculated results by this formula agreed well with the experimental values for the coal or artificial ashes.

The SCR Frequency Tripler

Shoji FUKUDA
Hajime FUJIWARA

Abstract

One circuit of constant ratio frequency changers utilizing SCRs was proposed and in this paper this circuit was treated as the frequency tripler.

The operating principle of the frequency tripler was clarified and the operating characteristics were analyzed. The calculated values and the tested values were compared.

The operating principle of this device is very simple. The conducting SCRs are turned off by the applied voltage becoming zero or minus and the mechanism of the commutation is also very simple. Therefore the commutation reactors and capacitors in the case of the self-excited inverter are unnecessary. The possibility of a short circuit when the choice of the firing angle fails is the inherent disadvantage. The number of SCRs in this device is six when the neutral point of the transformer is used otherwise twelve.

**Development of a General Program
concerning a Numerical Method for Stability Analysis
of Time-Varying Linear Systems with
Periodic Coefficients**

Ichiro SUGIOKA
Ryozauro TAGAWA

Abstract

In various engineering fields, we often have a case in which the behavior of a system under consideration is described by a set of linear ordinary differential equations with periodic coefficients, and up to the present, various studies have been presented related to the stability analysis of such a system.

In this paper, a general program using especially the method of reference (1) is proposed, and also, the relation between the complexity of the system and the memory capacity of a computer were examined, and further, the relation between the complexity of the system and the computation time were examined.

Estimation of the Linear System Parameters

Osamu ISHIKAWA
Ichiji YASUDA

Abstract

The object of this paper is to estimate linear system parameters (coefficients) from given sampled data.

This procedure consists of two stages. The first stage is the estimation of the parameters in the higher order difference equation from given sampled responses. In this case, for process noise and error of measurement included in the data, the method of least squares is applied, and a compensation method for linear dependence induced sampled data matrix was also discussed.

The second stage is parameter conversion of difference equation to differential equation, in which the formula of derivative function is approximately used.

Measurement of Directivity Factors in Photomixing with Coherent Light

Kojiro KOYANAGI
Ichiro SAKURABA

Abstract

This paper deals with measurements of directivity factors in photomixing with coherent light.

In the case of unfocused light beams the directivity factor of a rectangular detector of sides a and b , $D(\theta_1, \theta_2, \varphi_1, \varphi_2)$ is $4 \sin(pa/2) \cdot \sin(qb/2) / pqab$ and the directivity factor of a circular detector of radius ρ_0 is $D(\omega, \rho_0) = 2 J_1(\omega, \rho_0) / (\omega\rho_0)$. A comparison of calculated and experimental values was made with rectangular and circular detectors of MS 9 S, M 7316, M 7324 and M 7696 photomultipliers and LSD 39 B photodiodes in optical homodyne receivers and a good experimental comparison was obtained.

A Proposal of a New Type of Energy Density Mobile Radio Antenna

Kiyohiko ITOH
Tadashi MATSUMOTO

Abstract

This paper deals with the analysis of the fading phenomenon due to motion through the coherent standing wave pattern in the mobile radio electromagnetic field and a new type of energy density mobile radio antenna which is helpful in overcoming this fading was proposed.

The energy density antenna is an antenna which samples the energy density, that is, $1/2(\epsilon E^2 + \mu H^2)$ of the mobile radio field, rather than the electric field. W. C.-Y. Lee proposed the energy density antenna system which samples E field by a monopole antenna and H field by a loop antenna. This paper proposes a new type of energy density antenna system which samples H field by a slot antenna or a magnetic current antenna and E field by a conductor antenna or a electric current antenna. Also, it was reported here that the new type of energy density antenna system (slot unipole antenna system) functions efficiently.

Mode Conversion of Gaussian Beams Propagating Through a Random Lens-Like Medium

Shōichi KIKUCHI
Masaaki IMAI
Tadashi MATSUMOTO

Abstract

The mean-square value of a mode conversion coefficient was analytically obtained for a gaussian light beam propagating through a lens-like medium, i.e., a gas lens, that is considered as a focusing element for a laser beam waveguide in which the dielectric-constant varies essentially with quadratic law but has a term varying at random.

The mode conversion coefficient is evaluated by calculating the scalar-field propagating through a lens-like medium with fluctuations by means of a perturbation method and then by expanding at any point in the medium the scattered-field in terms of the scalar-field in the absence of fluctuations.

Here, assuming that the randomly homogeneous medium has gaussian covariance of the dielectric-constant fluctuations, the mode conversion coefficient is obtained as a mean-square value.

In such a realistic guiding medium with aberrations, the conversion into parasitic modes increases a diffraction loss, and it influences the efficiency of a heterodyne system.

The mode conversion caused by a gaussian light beam scintillation in the atmosphere was also discussed.

Study of Propylene Oligomerization on Nickel Oxide-Silica-Alumina Catalyst

Kunimasa TAKAHASHI

Osamu SASAKI

Kazuo AOMURA

Hiroshi OHTSUKA

Abstract

In this paper, the oligomerization of propylene was studied in the presence of nickel oxide on silica-alumina catalyst. The effect of the catalyst carrier and reaction conditions on the composition of propylene oligomers, especially of dimers, were observed and discussed.

Silica-alumina used as catalyst carrier was a commercial catalyst for catalytic cracking. Its alumina content was 13 wt-% (dry base). The mixture of silica-alumina and bentonite was suspended in nickel nitrate solution. To this solution, ammonium carbonate solution was added to precipitate nickel hydroxide-nickel carbonate mixture on the carrier surface. This catalyst precipitate was filtered, washed and dried. The dry powder was pelleted. The catalyst pellets were activated by treating with hot air (600°C) for 6 hours.

A high pressure continuous flow system reactor was used for propylene oligomerization. The optimum reaction conditions were as follows: React. temp. 70~80°C, Press. 30~35 kg/cm²G, S. V. 750 g-propylene/ ℓ -cat·hr. Under these conditions, the reaction proceeded in liquid phase, when using the catalyst having 9.7 wt-% nickel, 91 wt-% of propylene conversion, 51 wt-% dimer yield and 58 wt-% dimer selectivity were obtained.

Optimum activation temperature of the catalyst was 500-600°C. A good parallelism was observed between the catalyst activity and acidity measured by the quinoline adsorption method and between the acidity and the nickel content of the catalyst.

The existence of nickel on the acid catalyst was found remarkably to favor the formation of normal olefins. N-hexenes content in propylene dimer was around 40 wt-% in the presence of the nickel oxide-silica-alumina catalyst in various ranges of nickel content. Isomerization of 4-methylpentenes was also carried out to confirm that n-hexenes were not the products of skeleton isomerization of isohexenes. The isomerization product mainly consisted of 2-methylpentenes and contained no normal components.

Studies on the Boron Trifluoride Catalyst (IV)

—On the Active Form (HBF_3OH) of $\text{BF}_3\text{-H}_2\text{O}$
Complex Catalyst in the Alkylation Reaction—

Eishi HASEGAWA
Norihiko YONEDA
Takao YOTSUYANAGI
Kazuo AOMURA
Hiroshi OHTSUKA

Abstract

The active form of $\text{BF}_3\text{-H}_2\text{O}$ complex catalyst in the alkylation of benzene with propylene was considered to be a proton donor substance ($\text{H}^+\text{BF}_3\text{OH}^-$) formed by the combination of BF_3 with H_2O .

In this paper, the durability of $\text{BF}_3\text{-H}_2\text{O}$ complex catalyst was studied with special reference to the behaviors of the active form $\text{H}^+\text{BF}_3\text{OH}^-$ in the catalyst.

The experimental results were summarized as follows:

1) The catalyst was inactive at the reaction temperature of $10\sim 50^\circ\text{C}$, when the $\text{BF}_3/\text{H}_2\text{O}$ mole ratio of the catalyst was 0.5 or lower. However, at the reaction temperature of 70°C , the $\text{BF}_3\text{-H}_2\text{O}$ complex having low $\text{BF}_3/\text{H}_2\text{O}$ mole ratio such as 0.4 showed some catalytic activity.

2) when the content of HBF_3OH in the catalyst was kept constant, the total amount of absorbed propylene was greatly affected by the BF_3 concentration in the catalyst and the reaction temperature.

3) At the reaction temperature of 10°C , the higher the BF_3 concentration in the catalyst, the longer the life of the catalyst. Generally speaking, high BF_3 concentration in the catalyst favored the durability of the catalyst. For example, 0.9 and higher $\text{BF}_3/\text{H}_2\text{O}$ mole ratio at the reaction temperature of 30°C and 0.80 and higher $\text{BF}_3/\text{H}_2\text{O}$ mole ratio at the reaction temperature of 50°C gave a long life to the catalyst, respectively.

4) These values of the $\text{BF}_3/\text{H}_2\text{O}$ mole ratio in the catalyst corresponded to the compositions of the stable $\text{BF}_3\text{-H}_2\text{O}$ complexes at 30°C and 50°C .

5) In the case of reaction temperature of 70°C , the total amount of reacted propylene increased with the decrease of the BF_3 concentration in the catalyst contrary to the results obtained at lower reaction temperatures. However, in this case, hydration of propylene accompanied the alkylation reaction to a remarkable extent.

6) The total amount of absorbed propylene also increased with the decrease of the feed rate of propylene. This tendency was remarkable especially when using

the catalyst having high $\text{BF}_3/\text{H}_2\text{O}$ mole ratios.

7) The main cause of the deterioration of $\text{BF}_3\text{-H}_2\text{O}$ catalyst in the alkylation of benzene with propylene was considered to be the chemisorption of propylene to the catalyst to form the ester-like substance ($\text{C}_3\text{H}_7^{\delta+}\cdots\text{BF}_3\text{OH}^{\delta-}$).

Studies on Density Current (III)

—A Comparative Study of the Outflow Pattern of the Density Current with a Single Ideal Fluid Flow—

Masakazu KASHIWAMURA

Abstract

As described in the previous paper, there are five flow patterns of a fluid, when it passes through an outlet into a heavier fluid. The example can be found at a river mouth.

The first pattern, which is called A pattern, appears in a condition where the density difference of the two fluids is large and a velocity of the lighter fluid is small while the breadth of the outlet is long. The last, called E pattern, on the other hand, occurs in a reverse condition, and it gives approximately the same feature with a turbulent jet.

The A pattern has stable stream lines, which extend over the heavier fluid while spreading gradually with the distance from the outlet, and eventually stretch in radial directions. It seems to resemble a flow pattern of a single ideal fluid which issues from an outlet.

This paper describes the extent of conformity between the A pattern flow and the ideal fluid flow. The velocity and the stream lines of the ideal flow have been calculated with conformal representation, while the A pattern was obtained through the experiment.

As a result, it appears that the stream lines of the two flows offer an approximate resemblance with the each other except for a point where the A pattern flow spreads over more rapidly after passing through the outlet. It appears, however, that there are differences between the two flows in velocity. The most remarkable difference is that the velocity along the center line decreases gradually in the ideal flow but, in the A pattern flow, accelerates temporarily at the outlet. The cause has been discussed by emphasizing that a depth of the lighter fluid plays an important part in the motion of the A pattern flow.

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Basic Investigations on the Cracking and Deformation Behavior of Reinforced Concrete Beams

Yoshio KAKUTA

Abstract

The present study deals with the basic properties of reinforced concrete (RC) beams with high-strength deformed bars in cracking and deformation due to bending.

Crackings of RC beams in the initial state as well as in the stationary state were analysed on the basis of bond characteristics of deformed bars between cracks.

The increase of deformation and crack widths of RC beams under sustained and repeated loads were analysed taking into consideration the effect of restriction of steel against delayed deformation of concrete and the effect of the gradual relaxation of bond stresses.

Furtherthea cracking and deformation behavior and mode of design of PRC beams, which are RC beams prestressed to improve its crack-resisting ability or to keep crack widths below the value permitted, were described.

Statistical Study on the Inflow into the Kanayama-Dam

Tsutomu KISHI
 Ken-ichi HIRAYAMA
 Akio MORI

Abstract

The time series $X(t)$ of ten-day-discharge of the Sorachi-River in Hokkaido was analysed. Records of observation from 1961 to 1966 are available but no observations were made from January to March. However, they were represented by

the first discharge of January.

The correlogram $\rho_{X'}$ of the time series $X'(t)$ which exclude the periodic components from $X(t)$ was approximated as $\rho_{X'}(k) = 0.514\rho_{X'}(k-1) + 0.130\rho_{X'}(k-2)$.

Normalised time series $Y(t)$ show an apparent periodicity of 4 to 5 years. A time series which exclude this apparent periodic component and discharge from January to March was expressed as $Y''(t)$. This was regarded as a stationary stochastic time series. $\rho_{Y''}$ was approximated as $\rho_{Y''}(k) = 0.575\rho_{Y''}(k-1) + 0.036\rho_{Y''}(k-2) - 0.222\rho_{Y''}(k-3)$. $\eta(t)$ is defined as a stationary stochastic time series, $s(t)$ as a periodic function and $\xi(t)$ as $\eta(t) \cdot s(t)$. Then there is such a relation among these serial correlations which can be written as $\rho_\eta = \rho_\xi / \rho_s$.

This relation was examined using the present data between $\rho_{X'}/\rho_s$ and $\rho_{Y''}$, but the results seemed to indicate that both were different types. This may be attributed to the limited data which was not sufficient to obtain an accurate $s(t)$. However $X'(t)$ showed little or no influence of such an effect, and generally ρ_ξ' can be accepted as an approximated value of ρ_η . Thus it may be possible to presume a type of ρ_Y from $\rho_{X'}$.

Statistical Analysis of the Daily River Discharge Sequence for the Ishikari River

Tsutomu KISHI
Ken-ichi HIRAYAMA
Masahiko HASEBE

Abstract

The sequences of the daily river discharge during August, September and October at Naka-Aibetsu and Inou, both are located in the upper reaches of the Ishikari River basin, were analyzed from a stochastic point of view by data recorded from 1952 to 1964. Such analyses concerning numerous statistical characteristics enable us the construction of a numerical simulation model for calculating the daily river discharge at a down stream station based on the assumption that up-stream station discharge is known.

In the first model the Inou discharge was calculated utilizing the regression line between Inou and Naka-Aibetsu, but the computed hydrograph was not so similar to the observed one.

In the second model the daily run-off was divided into two types, i.e., rainy day and fine day. When on a certain day the discharge is larger than that of the previous day, such a day is referred to as 'a rainy day', and the opposite is referred

to as 'a fine day.' If F_N indicates a fine day at Naka-Aibetsu and R_I is a rainy day at Inou and so on, the probability of the next four pair i.e., a) R_N-R_I , b) R_N-F_I , c) F_N-R_I and d) F_N-F_I were estimated from the actual data, and in addition the persistency of both rainy days and the fine days for each station were also considered. If the event has a persistency the probability of the same pair to persist should be increased.

Following the indication of R or F for the station the discharge was calculated by means of one of these four patterns which indicated statistical relation between the stations

In case of a) and c) the decay ratio Q_t/Q_{t-1} , and in b) and d) the increment in discharge (Q_t-Q_{t-1}) was determined statistically using the fitted probability density function. After the river run-off sequence was generated using the above simulation model, the difference between the calculated values and the observed values tested. It was shown that this method would be useful for planning and for statistical appraisal of water resource problems.

Study on the Mechanics of a River Confluence

Tadaoki ITAKURA

Chimataro ISHII

Abstract

Various types of river confluences with different geometry are known where the separation zone is formed immediately downstream from the confluence and has various sizes and shapes. It follows that analyzing the flow around a river confluence is a complicated problem.

An analytical and experimental study was carried out on the mechanics of a river confluence in this report. A water flow discharging normally into a main water flow was taken as a simplified model of a river confluence.

The following is an outline of the results obtained from the analysis:

a) The trajectory of positions of maximum velocity along the tributary were normalized to an identical elliptic arc by parameters. These parameters were calculated by the analysis of water-surface profile and velocity distribution.

b) A maximum value of the depth of water appeared immediately upstream from the confluence. On the contrary, the minimum depth occurred at the cross section of the deepest tributary penetration. These values were calculated by a similar mode of analysis as a).

c) The separation zone which was formed immediately downstream from the confluence had similar shapes. Its maximum width and length were normalized by a new parameter. Rouse's parameter⁴⁾ was modified by the ratio of widths of the tributary to the main flow and applied to normalize its maximum width and length.

d) Görtler's solution¹⁰⁾ for a diffusion of a jet was applied to the velocity distribution of the flow in a modified version. A parameter occurring in the solution was obtained from experiments; $\sigma=5.5$ to 6.0 for $l/D \leq 10$ and it increased with distance as $\sigma \propto l^{1/2}$.

e) An attempt was made to evaluate the energy acquisition of the main flow by an analysis which used to be carried out on a junction of pipes. Values of the coefficient of loss of the velocity head were small enough to be neglected as compared with the values in pipes.

Longitudinal Distribution of Surface Salinity in an Estuary

Isao YAKUWA
Morimasa OHTANI

Abstract

The relation between the behavior of a salt wedge and salinity diffusion at the interface of salt and fresh water have been observed for many years at the mouth of the Ishikari River.

In this paper, the longitudinal distribution of salinity in a fresh water layer and the role of diffusion coefficients of the river flow on salinity distribution are discussed by solving the differential equation of salinity diffusion under adequate boundary conditions. The calculated distribution of surface salinity was compared with the observed results in the Ishikari River.

Velocity and Temperature Distributions in a Heated Turbulent Vertical Jet

Shoichiro FUKUSAKO

Masaru KIYA

Mikio ARIE

Abstract

A steady, turbulent flow in a heated vertically ascending jet is considered within the framework of the boundary layer theory by taking the effect of buoyancy caused by the temperature difference into account. By assuming a similar velocity and temperature profiles, a two-dimensional and an axisymmetric case were treated. The buoyancy was found to produce a considerable influence on the flow in the vertical jet.

By applying Prandtl's mixing length hypothesis, $\overline{Lv} = kB(U_{\max} - U_{\min})$, it was found from the present analysis that the width of the heated vertical jet increases in proportion to the streamwise distance, for both two-dimensional and axisymmetric cases. The maximum temperature on the axis of the jet is proportional to x^{-1} (two-dimensional) and to $x^{-5/3}$ (axisymmetric). The maximum velocity on the jet axis was found to change in proportion to x^0 (two-dimensional) and to $x^{-1/3}$ (axisymmetric). These results of the development of the vertical jets are equivalent to those obtained by Schmidt who applied Prandtl's other mixing length hypothesis, $\overline{Lv} = l^2 |dU/dy|$.

In the two-dimensional case, the velocity profile is almost the same as that of Schmidt, but the temperature profile was considerably different. In the axisymmetric case, on the other hand, the situation was reversed. The temperature profile obtained by the present analysis for the axisymmetric jet represents the experimental data more closely than the analytical results of Schmidt.

An Experimental Study on a Two-Dimensional Wake of a Rectangular Cylinder Attached to a Plane Wall

Shoichiro FUKUSAKO
Masaru KIYA
Mikio ARIE

Abstract

Two flow visualization techniques using dye and aluminium flakes were applied to investigate the flow around a rectangular cylinder attached to a plane wall. The length of the standing eddy to the lee of the cylinder was found to have a tendency to increase linearly with respect to $\log R$, R being the Reynolds number in terms of the height of the rectangular cylinder and the oncoming fluid velocity. The ratio of the length of the rectangular cylinder to its height was varied from 0 to 3 and the Reynolds number from 19 to 2500.

Studies on the Secondary Injection Phenomena of Bosch Type Fuel Injection Systems

Yoshihiro MAEDA
Tadashi MURAYAMA
Shôichi FUKAZAWA

Abstract

The results of some measurements on the secondary injections of Bosch type fuel injection systems are reported and these experimental results were compared with the calculated results.

It was shown that the present experimental results were in good agreement with the calculated values.

In addition, the effects of temperature on the secondary injection of fuel injection pump were investigated.

As a result of these experiments, it was shown that the empirical limit of secondary injection varied considerably.

On a Performance Test for a Snow Melting Machine (I)

—Intermittent Feeding of Snow—

Shôichi FUKAZAWA

Takeshi SAITO

Hiroshi TANIGUCHI

Ryôji ISHIGURO

Ken-ichi ITO

Abstract

It is an important practice to remove the snow from a surface of highway and a city zone in a every country where is cold and snowy in a period of winter. Many efforts have been devoted to increase the efficiency of the work by engineers in the related fields.

The present study gives the results of a performance test for a stationary snow melting machine which is becoming increasingly important in city zones. In this report the performance of the machine is analyzed under conditions of intermittent feeding of snow.

The snow is fed into the melting machine at fixed intervals of time by dump trucks after weighting procedures. The test conditions are widely varied by the feeding rate and the quality of snow. The measured items are the quality of snow, feeding and melting quantity of snow, the heat input, the temperature distribution, and so on. Each performance is analyzed based on the data obtained and the following results are concluded: The capacity of the machine should be measured by the melting quantity and not by the feeding quantity. The efficiency of the machine should be evaluated by using the melting quantity of snow and not by the temperature increase of the water. Some counterplan to the air or water pollution by CO gas, hydrocarbon and soot in the combustion gas should be considered. It is also noted that steps should be taken to minimize noise with respect to the public.

Azimuthal-Velocity-Prebunching Effects on Electron-Wave Interactions in CEF-Type Devices

Ichiro SAKURABA
Teruo HAGA

Abstract

This paper is concerned with the azimuthal-velocity-prebunching effect on electron-wave interactions in CEF-type devices in a special case where $b=0$, $d=0$, $Q=0$, $\beta_e \approx 20$, and $C \approx 0.05$.

The small-signal forward-wave output power was given by

$$P(\theta) = \frac{1}{2} G_{\text{eq}} |r_0^2 \Omega_0 \Omega_1(0)/\eta|^2,$$

and the equivalent conductance G_{eq} was yielded by

$$G_{\text{eq}} = \frac{9}{8} \frac{1}{k_r^2 \beta_e^4} \frac{1}{K} \left[2 \left(\cosh \frac{1}{2} \sqrt{\beta_e C} \phi - \cos \frac{\sqrt{2}}{\beta_e C} \phi \right)^2 + \sin^2 \frac{\sqrt{2}}{\beta_e C} \phi \right].$$

The term of $\cosh(\sqrt{\beta_e C} \phi/2)$ is due to the growing and decreasing waves in the device and the terms of $\cos(\sqrt{2} \phi/\beta_e C)$ and $\sin(\sqrt{2} \phi/\beta_e C)$ are due to the characteristic ripple in the CEF-type focusing systems. The ripple effect on the equivalent conductance in the azimuthal-velocity-prebunching case is larger than that on the equivalent resistance in the radial-current prebunching case.

Crystal Growth and Electrical Characteristics of Vacuum-Deposited Germanium Films

Mamoru TADAKI
Yoshihiko OGAWA
Kazuo TOKIWANO
Teiichi KUROBE

Abstract

The effects of formation conditions on the crystalline quality of thin Ge films, vacuum evaporated onto a mica substrate were investigated. The crystalline quality was found to depend markedly upon the substrate temperature. Amorphous films formed on the substrate in a temperature range of 20~300°C while polycrystalline

films were formed in a temperature range of 400~700°C. It was also noted that several orientations were obtained depending on varied substrate temperatures. At temperatures above 650°C the films with large particle sizes were obtained, indicating a very strong [110] texture in electron-diffraction patterns.

In addition, an experiment on the temperature dependence of the conductivity and the Hall effect was preformed. Further, it was shown that evaporated Ge films have inductive properties. The relaxation time for hole trapping was computed in terms of the inductance.

Optical Heterodyne Detection of a Focused Gaussian Beam Propagating Through a Random Medium

Shigeo FUJIKI
Michio SUZUKI

Abstract

An investigation on optical heterodyne detection of a plane wave propagating through a randomly inhomogeneous medium was made by D. L. Fried and others. Further it was shown by Kinoshita, et al, that the fluctuations of a focused gaussian beam decreases near the focal point.

In this paper, a significant reduction of scintillation in the optical heterodyne detection of a focused gaussian light beam propagating through a random medium was theoretically investigated. The investigation was made on the assumption that the beam was emitted from a coherent and monochromatic source extended over a pupil plane with a circularly symmetric gaussian amplitude distribution and a curved wavefront which characterizes the beam shape, and the refractive index fluctuations of the medium were statistically homogeneous and isotropic.

**Some Properties of the Higher-Order Images Reconstructed
from Grating-Like Acoustical Holograms and
their Applications for Multiplexing
and Multi-Color Acoustical Holography**

Yoshinao AOKI

Abstract

In this paper grating-like acoustical holograms are constructed by scanning the acoustical fields with a microphone. The optical reconstruction of image is conducted using laser light and the higher-order images were observed. The grating-like acoustical holograms were constructed by a Gabor's coherent background method, a two-beam interference method and an electronic reference method. The resulting higher-order images reconstructed from these holograms were discussed. Some properties of the higher-order images, contrast enhancement and contrast inversion, were analyzed from a point of non-linearity of the hologram recording system and the property of the grating.

For the applications of grating-like acoustical holograms, a space division multiplexing acoustical holography and a multi-color acoustical holography were proposed. An experiment to construct space division multiplexing acoustical holograms was conducted, where information of different objects was recorded in a single grating-like acoustical hologram by scanning with a microphone. As a preliminary experiment to reconstruct a multi-color image from acoustical holograms, an experiment to reconstruct a single-color image from a grating-like acoustical hologram was conducted.

Liquid Laser (I)

—Spectroscopic Property of Tb^{3+} : $POCl_3$ - $SnCl_4$ Solution—

Kenji NAGASHIMA
Michio SUZUKI

Abstract

The absorption, fluorescence emission and excitation spectra of Tb^{3+} : $POCl_3$ - $SnCl_4$ solution and the fluorescence decay time of this solution are described in detail.

Our experiment shows that the $\text{POCl}_3\text{-SnCl}_4$ solvent is transparent above $300\text{ m}\mu$ but the $\text{SeOCl}_2\text{-SnCl}_4$ is above $400\text{ m}\mu$, and the $\text{Tb}^{3+}:\text{POCl}_3\text{-SnCl}_4$ solution has a comparative strong absorption band from $300\text{ m}\mu$ to $400\text{ m}\mu$ and the lifetime of terbium in the solution was 3.3 mS at room temperature.

It is conjectured that a long lifetime as compared with that of terbium β -diketone chelete at room temperature would be due to the heavy atom effect, but the exact mechanism remains unknown.

A possible application as a laser material is briefly considered.

Coexistence Curves in Regular Solutions

Masaji ONODERA

Abstract

Four types of coexistence curves in regular solutions were obtained by application of the asymptotic method. Two of them are ordinary coexistence curves and the others are anomalous coexistence curves, which have not been mentioned in the literature based on existing theories. One of the anomalous coexistence curves was connected with the excess heat capacity.

Glass Transition

Masaji ONODERA

Abstract

The thermodynamic properties of a glassy state were obtained from statistical mechanics by applying an asymptotic method which allows for a variation of volume with temperature. The glass temperature is defined as the point which maximizes the curvature of the curve of volume versus the temperature. The volume, the excess heat capacity, and entropy at the glass temperature were obtained and compared with experimental results. The present theoretical predictions were found to be in excellent agreement with the experimental results.

Direct Determination of Organic Oxygen in Coal and Coal Derivative by means of the Unterzaucher Method

Susumu YOKOYAMA
Kazuo MAKINO
Yoshihisa HASEGAWA
Gen TAKEYA

Abstract

The oxygen containing groups in coals and its pyridine extracts of different ranks of 12 Japanese coals were studied to determine the structure of coal. In such studies, the exact content of oxygen in coals and its pyridine extracts must be known. Thus, it follows that a direct determination method of oxygen in coals and coal derivatives must be developed.

The present report deals with an analysis of oxygen in pyridine extracts of coal by the Unterzaucher method using a semi-micro apparatus. Coal derivative samples of 20 to 30 mg were pyrolysed at 950°C in nitrogen stream. Water, carbon dioxide and other oxygen containing gases produced by pyrolysis of the coal sample were converted stoichiometrically to carbon monoxide allowed to pass over platinum-charcoal heated to 950°C. The carbon monoxide was oxidized completely to carbon dioxide by iodine pentoxide and was measured gravimetrically.

Some authentic pure substances with known oxygen contents were previously measured to confirm the accuracy of the apparatus, and the values thus obtained were in close agreement with the theoretical oxygen content values. After this test run the coal derivatives were studied for the analysis of their oxygen contents. The results of oxygen determination for pyridine extracts of coal showed high reproducibility and were in approximate agreement with values obtained by indirect calculations for the difference of C, H and N. The contents of oxygen in pyridine extracts of coal determined by the Unterzaucher method seem to show a good reliability.

Baeyer-Villiger Reaktionen der Benzophenonkarbonsäuren (I)

—Eine Synthese der Terephthalsäure und p-Oxybenzoesäure
aus Benzophenon-4, 4'-dikarbonsäure durch
Oxydation mit Peroxysäure—

Yoshiyuki TAKATA
Kiyoshi ICHIMURA
Katsuyoshi KONDO

Zusammenfassung

Mit Hilfe der Baeyer-Villiger Reaktion versuchten wir die gleichzeitigen Herstellungs-verfahren der Terephthalsäure und p-Oxybenzoesäure aus Benzophenon-4, 4'-dikarbonsäure und dem entsprechenden Äthylester.

Zuerst durch Oxydation in konzentrierter Schwefelsäure, dann bei der Hydrolyse mit Natronlauge, aus Benzophenon-4, 4'-dikarbonsäure gewinnt man Terephthalsäure. Ausbeute daran 50 proz. der Theorie. Hierauf wird nicht p-Oxybenzoesäure entstehen machen. Mit dem Gemisch von konz. Schwefelsäure, Wasserstoffperoxyd und Eisessig wird Benzophenon-4, 4'-dikarbonsäure kaum oxydiert. Durch Oxydation mit dem Gemisch von konz. Schwefelsäure, Wasserstoffperoxyd, und Eisessig wird Benzophenon-4, 4'-dikarbonsäurediäthylester leicht verändert. Nach Hydrolyse es entstand gleichzeitig beide Reaktionsprodukten, Terephthalsäure und p-Oxybenzoesäure, deren Ausbeuten daran respektiv 95-und 40 proz. waren.

Versuchung über Triazin-derivaten (I)

—Hydrolyse des Benzoguanamin—

Yoshiyuki TAKATA
Kazuo TAN
Hiroyuki ÔMORI

Zusammenfassung

Wir versuchten die Hydrolyse des Benzoguanamin. Beim Erhitzen mit Salzsäure wird das Benzoguanamin immer leichter hydrolysiert und bildet eine Verbindung mit Fp. 267°C (I), 2-Amino-4-hydroxy-6-phenyl-s-triazin, 2, 4-Dihydroxy-6-phenyl-s-triazin und Benzoesäure. Wir beobachteten, dass der Triazinring mit Salzsäure ziemlich leicht zerfällt, sowie tritt Benzoesäure durch Abspaltung dieses Triazin-ring

des Benzoguanamin ein. (I) bildet 2-Amino-4-hydroxy-6-phenyl-s-triazin mit Natronlauge, oder 2, 4-Dihydroxy-6-phenyl-s-triazin mit Salzsäure, auch (I) bildet mit Essigsäureanhydrid gleichzeitig 2, 4-Dihydroxy-6-phenyl-s-triazin und N,N-Diacetyl-benzoguanamin.

Durch diese Resultaten auffassten wir (I) als eine Molekül-Verbindung von 2, 4-Dihydroxyl-6-phenyl-s-triazin und Benzoguanamin. Im weiteren es gelang uns aus 2, 4-Dihydroxy-6-phenyl-s-triazin und Benzoguanamin die Struktur des (I) synthetisch feststellen.

Wenn man Benzoguanamin mit Natronlauge erhitzt, soentsteht 2-Amino-4-hydroxy-6-phenyl-s-triazin allein mit guter Ausbeute. Das beweist sich zum Herstell-
ung des 2-Amino-4-hydroxy-6-phenyl-s-triazin als eine taugliche Verfahren.

A Spectrophotometric and Potentiometric Study of the Formation Equilibrium of Catechol- 4-Sulfonate-Aluminum Complexes

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Yasuo KUDO

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Abstract

The formation equilibrium of catechol-4-sulfonate (PCS)-aluminum complexes was investigated by spectrophotometric and potentiometric method. PCS is a useful reagent for the classification of the hydrolysis products of aluminum ions in an alcohol solution, into mononuclear and polynuclear species by the difference in their rate of formation of the colored complex.

PCS reacted with aluminum to form stable complexes, with a molar ratio of Al to PCS 1:1 in pH range from 4.5 to 6.5, 1:2 in pH range from 6.5 to 8.5 and 1:3 in pH range greater than 9.0. The stability constants of these complexes determined by potentiometric titration were $\log K_1=16.5$, $\log K_2=12.8$ and $\log K_3=8.19$, and acid dissociation constants of hydrogen complexes were $pK_{Al(O_2Pc)_2(OH)Pc^+}^H=3.24$, $pK_{Al(O_2Pc)_2(OH)Pc^{2-}}^H=4.94$ and $pK_{Al(O_2Pc)_2(OH)Pc^{3-}}^H=7.53$ in 0.1N KNO_3 solution at $25.0 \pm 0.1^\circ C$.

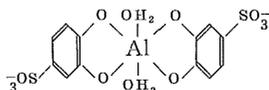
By a detailed comparison of an absorbance \sim pH curve and an equilibrium diagram for the aluminum complexes evaluated from these constants, the following conclusions were obtained:

- 1) 1:1 and 1:2 complexes showed two absorption peaks at $255 m\mu$ and $298 m\mu$,

and 1:3 complex showed a peak at 305 m μ ,

2) The molar absorptivity of the 1:2 complex was the largest among these complexes and it was 1.25×10^4 at 298 m μ ,

3) The complex employed for the spectro-photometric determination of aluminum in our previous paper was found to be 1:2 complex.



Detailed discussions were also made on the effect of the sulfonate group on the ability of the complex formation of catechol-OH groups. The increase of the amount of acid complexes of PCS compared with that of tiron was observed and it was well explained by the pronounced electron-withdrawing effect at their para position in the benzen ring having only one sulfonate substituent.

Selective Hydrogenation of α,β -Unsaturated Aldehyde to Unsaturated Alcohol (I)

—Selective Hydrogenation of Crotonaldehyde to Butyraldehyde over a Copper Catalyst—

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Takao YOTSUYANAGI
Kazuo AOMURA

Abstract

Selective hydrogenation of crotonaldehyde to produce butyraldehyde over a copper catalyst



was studied with special reference to the nature of the active sites on the catalyst by the poisoning and the heat treatment method.

The selectivity of the hydrogenation increased by poisoning the catalyst with thiophene and the same behavior was also observed in the case of the heat treatment. The increase of butyraldehyde in the product was due to the preferential inhibition of the hydrogenation of $\text{H} \diagup \text{C}=\text{O}$ group of butyraldehyde. These results suggest that the active sites on the catalyst for the hydrogenation of butyraldehyde clearly differs from those for the hydrogenation of C=C group of crotonaldehyde.